Nuclear Development

Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles

A Comparative Study

NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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In these and related tasks, the NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has a Co-operation Agreement, as well as with other international organisations in the nuclear field.

FOREWORD

Partitioning and transmutation (P&T) aims at reducing the amount of actinides, and thus the radiotoxicity of the high-level waste (HLW) going to disposal. Its purpose is ultimately to facilitate the geological disposal of actinide-containing HLW. To make this technologically complex process worthwhile, a reduction of the long-term radiotoxicity of HLW by a factor of at least one hundred is desirable.

The first OECD/NEA system study, entitled "Status and Assessment Report of Actinide and Fission Product Partitioning and Transmutation" (1999), already studied the necessary technologies to achieve this goal. However, the more effective transmutation strategies with fully closed fuel cycles and the specific role of accelerator-driven systems (ADS) in these fuel cycles were the not addressed. The present, second P&T systems study closes this gap and compares fast reactor (FR) and ADS-based actinide transmutation strategies in order to highlight the specific role that ADS might play and the main differences between ADS and FR with respect tot reactor properties, fuel cycle requirements, economic aspects, and R&D needs.

P&T is introduced in the first two chapters. The comparative analysis using a consistent set of transmutation strategies is addressed in Chapter 3. The status of FR and ADS technologies is compared in Chapter 4; Chapter 5 analyses the safety aspects of both systems. The report addresses the economics of transmutation strategies in Chapter 6, the perceived R&D needs in Chapter 7 and fission product transmutation and alternative approaches to the selected strategies in this study are described in Chapters 8 and 9 respectively. Each technical chapter carries its own conclusions. The overall conclusions of this study are given in Chapter 10.

The present report has been prepared by the group of experts listed in Annex A and is published under the responsibility of the Secretary General of the OECD. It does not necessarily represent the official governmental opinion nor that of the international organisations involved.

Acknowledgements

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The group wishes to dedicate this report to the family of our colleague and friend, Dr. Mikael Björnberg, who passed away during the period of this study and who would surely have been a supporter of its outcome.

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EXECUTIVE SUMMARY

Scope of study and principal messages

The long-term hazard of radioactive wastes arising from nuclear energy production is a matter of continued discussion and public concern in many countries. By the use of partitioning and transmutation (P&T)[1] of the actinides and some of the long-lived fission products, the radiotoxicity of the high-level waste (HLW) and, possibly, the safety requirements for its geologic disposal can be reduced compared with the current once-through fuel cycle. To make the technologically complex enterprise worthwhile, a reduction in the HLW radiotoxicity by a factor of at least one hundred is desirable. This requires very effective reactor and fuel cycle strategies, including fast reactors (FRs) and/or accelerator-driven, sub-critical systems. The accelerator-driven system (ADS) has recently been receiving increased attention due to its potential to improve the flexibility and safety characteristics of transmutation systems.

The present study compares FR- and ADS-based actinide transmutation systems with respect to reactor properties, fuel cycle requirements, economic aspects, and R&D needs. The essential differences between the various systems are evaluated with the help of a number of representative "fuel cycle schemes". The strategies investigated include an evolutionary transmutation strategy in which the ADS provides additional flexibility by enabling plutonium utilisation in conventional reactors and confining the minor actinides to a small part of the fuel cycle, and two innovative transuranics (TRU) burning strategies, with an FR or an ADS, in which plutonium and minor actinides are managed together to minimise the proliferation risk. A novelty in the present study is that the analyses are carried out in a consistent manner using reactor and fuel cycle parameters which have been agreed upon by international experts.

Principle messages from the study which could influence P&T policy development are:

- Fuel cycles with multiple recycling of the fuel and very low fuel losses are required to achieve the desired hundred-fold radiotoxicity reduction.
- All transmutation strategies with multiple recycling of the fuel can achieve similar radio-toxicity reductions, but the choice of the strategy strongly influences fuel cycle requirements.
- The ADS is particularly suited as a "dedicated" minor actinide burner in steady-state scenarios and provides flexibility in transient scenarios.
- The ADS-based evolutionary, and the FR-based innovative, approaches appear to be attractive transmutation strategies, from both technical and economic viewpoints.
- The full potential of a transmutation system can be exploited only if the system is utilised for a minimum time period of about a hundred years.
- A considerable amount of R&D on sub-critical reactors, advanced fuels, and materials would be needed before ADS-based transmutation technology could be deployed.

1. A list of acronyms is given in Annex B.

General context

The world-wide increasing energy demand in general, and electricity demand in particular, call for a re-evaluation of fission energy as a long-term energy source. In this context, a recent OECD/NEA publication has investigated the extent to which nuclear energy is compatible with the goals of sustainable development, and how it can best contribute to them [1]. Although present light water reactors (LWRs) are capable of covering the nuclear energy demand for many decades to come, there is a longer-term need for integrating advanced reactors, including fast reactors, into the nuclear energy system. Important development goals for such advanced systems are environmental friendliness, resource efficiency, and cost-effectiveness, while accounting for socio-political concerns such as proliferation.

In the early days of nuclear energy, electricity generation in LWRs as well as FRs was estimated to be economically competitive with other forms of electricity generation. At that time, uranium resources were assumed to be the limiting factor for nuclear deployment, while the limited amount of radioactive waste was seen as less of a concern than it is today. This early perspective called for a rapid introduction of conventional, uranium-plutonium mixed-oxide (MOX) fuelled fast reactors with a fuel cycle which is fully closed for plutonium, but not for the minor actinides neptunium, americium and curium, which are at least as radiotoxic as plutonium. A complete closure of the fuel cycle by recycling the minor actinides as well was already envisaged at that time, but not given much attention because the utilisation of the energy content of the minor actinides is not economically attractive.

Today, while uranium is still abundant but radioactive waste is giving increasing rise to public concern, an attempt to progress towards the ultimate goal of a fully closed, FR-based fuel cycle via the intermediate step of a transmutation system is appropriate. The partitioning and transmutation of actinides and fission products which are now put to waste would allow the "radiological cleanliness" of nuclear energy to be improved, and thus one of the most important requirements for an environmentally friendly nuclear energy system to be addressed. It is clear that not only the technical but also the economic feasibilities of such a system must be demonstrated.

Previous studies and adopted approach

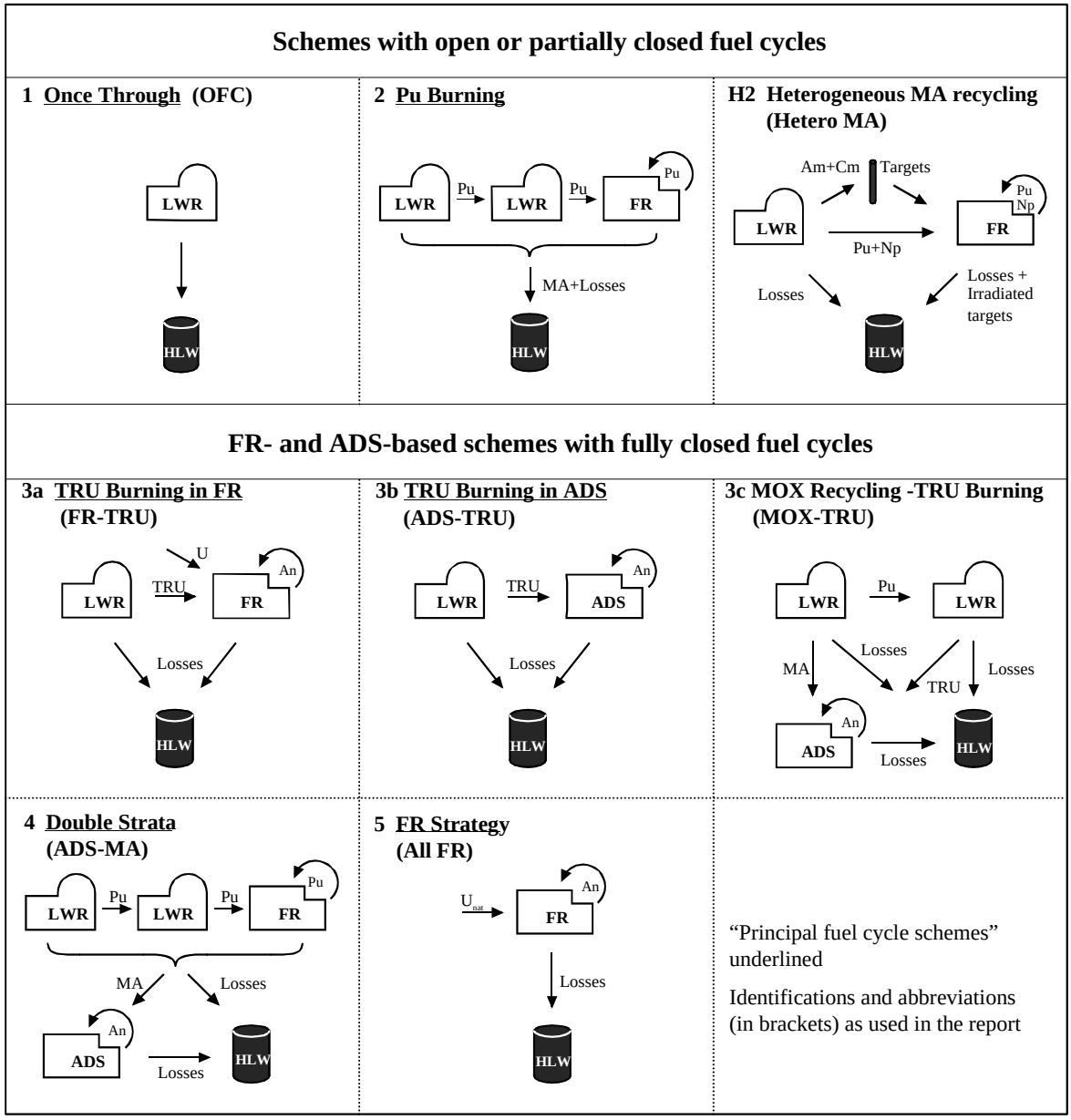
In response to the interest of Member countries, and recognising the activities pursued, the Nuclear Energy Agency initiated a long-term programme on P&T in 1989, addressing a wide range of technical and scientific issues.[2] An International Exchange Programme was established to strengthen international collaboration, and a first P&T systems study was carried out from 1996 to 1998 [2]. This systems study focused on a review of the progress in P&T and the possible benefits for waste management. Specific fuel cycle strategies were discussed, covering plutonium recycling and the additional burning of minor actinides in dedicated reactor systems; however, the more effective transmutation strategies with fully closed fuel cycles and the specific role of the ADS in these fuel cycles were not addressed. The present, second P&T systems study tries to close this gap and thereby complements the first study. Specific aims of this second study are the clarification of the roles and relative merits of the FR and the fast-spectrum ADS in closed fuel cycles by means of comparative analysis, as well as the assessment of the development status of the ADS with emphasis on reactor and fuel cycle technology, safety, economics, and general feasibility.

To quantitatively assess the advantages and drawbacks of different plutonium and minor actinide (MA) burning strategies, seven "fuel cycle schemes", shown in Figure 1, have been selected and compared with the current once-through fuel cycle (OFC). The schemes are generic and stand for

2. A historic overview of P&T activities in NEA Member countries and international organisations is given in Annex C.

groups of strategies with scope for variation according to national preferences. All reactors are assumed to be electricity producers.

Figure 1. **Overview of analysed fuel cycle schemes**



Note:
The "principal" fuel cycle schemes represent cornerstone strategies. Combinations of these are possible. For example, the MOX-TRU scheme combines elements of the ADS-TRU and plutonium burning schemes. The principal schemes were analysed using a single nuclear data library and a single reactor code system.

Schemes 3a, 3b, 3c and 4 allow the essential differences between FR- and ADS-based transmutation strategies with fully closed fuel cycles to be demonstrated. In addition, two schemes with partially closed fuel cycles are considered: scheme 2 is of interest because plutonium burning itself is an important issue and transmutation always involves plutonium burning as a preceding or simultaneous process. The heterogeneous recycling scheme H2 represents a possible alternative to the closed fuel cycle which, however, has an inferior transmutation potential. Finally, the all-FR strategy represents the long-term goal for nuclear development. Only "burner" reactors with solid fuels are considered, and these are optimised for a high burning efficiency so that they can support a large fraction of LWRs in the reactor park. The comparison is unique with regard to the use of consistent calculation methods, and reactor and fuel cycle parameters which have been evaluated specifically for this study.

Sustainability comparison

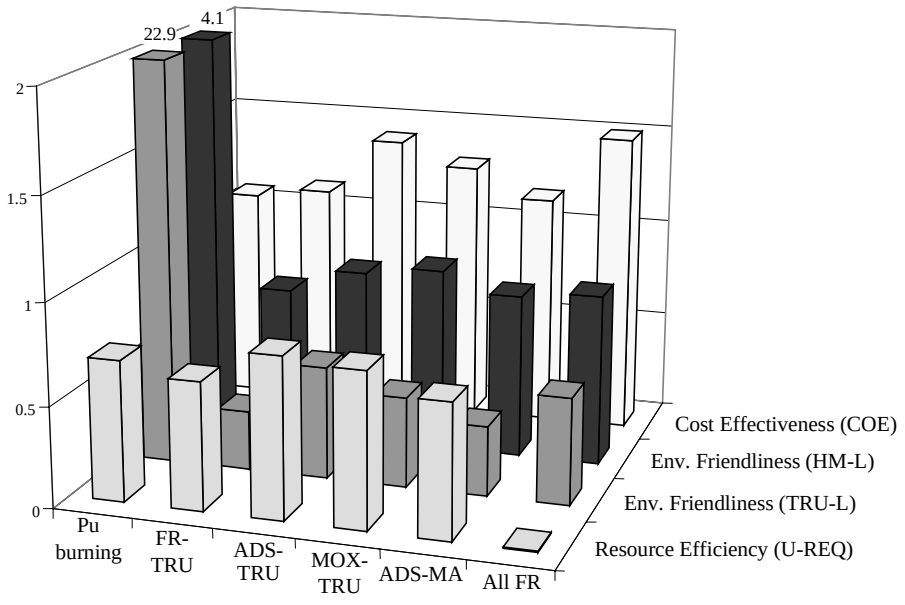
The comparison considers three axes of sustainability, namely resource efficiency, environmental friendliness, and cost-effectiveness. Key criteria along the second axis are the heavy metal and TRU losses, and the radiotoxicity of the losses, to repository. The principal results are illustrated in Figure 2 and can be summarised as follows:

- All transmutation strategies with fully closed fuel cycles can, in principle, achieve similar reductions in the actinide inventory and the long-term radiotoxicity of the high-level waste, and these are comparable with those of a pure fast reactor strategy. This implies that there are no distinct differences between the respective potentials of the FR and the ADS.
- With the assumed reactor and fuel cycle parameters, these strategies can achieve a more than hundred-fold reduction in the long-term waste radiotoxicity and even higher reductions in the heavy metal and TRU losses to repository, compared with the once-through fuel cycle. This applies for multiple recycling of the fuels, high fuel burn-ups, and very low reprocessing and fuel fabrication losses. For the latter, a value of 0.1% for all actinides is assumed, as already achieved for uranium and plutonium, though an ambitious target for the other actinides.
- Regarding actinide waste production and technological aspects, the FR-TRU and the ADS-MA scheme are similarly attractive. The first can gradually evolve to a pure fast reactor strategy, but requires higher initial investment in fast reactor and advanced fuel cycle technologies. The second confines the minor actinides to a small side-stream of the fuel cycle where, however, very innovative technology is needed. Here, the ADS has the advantage that it can burn pure minor actinides while avoiding a deterioration of the core safety characteristics.
- The economic analysis indicates that ADS-based transmutation technology can be made more competitive by burning as much plutonium as possible in conventional reactors, i.e. MOX-fuelled LWRs and FRs. This favours the ADS-MA scheme, which, together with the FR-TRU scheme, also features the lowest electricity costs of all transmutation schemes. In these cases, P&T is estimated to add a relatively modest 10-20% to the electricity costs of the once-through fuel cycle. Although such cost increases would be unacceptable to the market at present, they are limited and might be affordable in the future if price increases rendered fossil fuels less competitive or society placed a premium on reducing waste radiotoxicity.

The study also shows that plutonium burning alone is useful for the management of plutonium, but cannot qualify as a transmutation strategy because it reduces the long-term waste radiotoxicity by only a factor of about five. Recycling americium and curium heterogeneously in special "target" pins which are disposed of after irradiation, as shown in the hetero MA scheme H2, is technically less

demanding than a closed fuel cycle strategy, but is also about a factor of two less effective in reducing the radiotoxicity; this strategy is being explored as a near-term transmutation option.

Figure 2. **Sustainability comparison**



U-REQ: Natural uranium requirement relative to OFC.
TRU-L: Transuranics losses to repository (% of OFC).
HM-L: Heavy metal losses to repository (tenths of % of OFC).
COE: Cost of electricity relative to OFC (nominal case).

Note: For the Pu-burning scheme, TRU-L and HM-L are off-scale.

Regarding the utilisation period of transmutation systems, the study confirms that physical limitations associated with the production and destruction of in-pile and out-of-pile fuel inventories imply very long time constants for the introduction and final phase-out of such systems, and that P&T technology can, therefore, achieve its goal only if it is introduced with the intention of using it for at least a century. In particular, the full radiotoxicity reduction benefit can be realised only if the TRU inventory of the system is ultimately burnt and not put to waste.

Finally, it should be noted that all transmutation strategies including LWRs in the reactor mix require similar uranium resources and produce similarly large streams of residual uranium as the LWR once-through strategy. If the residual uranium is not considered as a resource for future fast reactors, its long-term radiological impact must also be assessed.

ADS technology and safety

Though the FR and the ADS perform similarly with respect to environmental friendliness criteria, they differ considerably from technology, operation, and safety viewpoints.

In this context, two advantages of the ADS are of particular interest:

- The sub-critical, ADS concept enables the design of reactor cores which would otherwise not have acceptable operating characteristics. In particular, the possibility of operating a sub-critical actinide burner with a uranium-free (or thorium-free) fuel supply allows burner effectiveness to be maximised and hence the fraction of specialised transmuters in the reactor park to be minimised.
- Moreover, the concept allows the adjustment, i.e. increase, of the reactivity margin to prompt criticality, thereby reducing the potential of the core for a power excursion. This is useful primarily for minor actinide burners, for which this margin is only about half of that of a normal fast reactor if the core is operated in a critical mode. TRU burner cores are less degraded in this respect.

The above-mentioned advantages of the ADS have to be balanced against the technological challenges arising from the coupling of a reactor and an accelerator, and the necessity to accommodate new types of operational and accidental transients. Regarding the former, the following problems require attention:

- Although the development of accelerators is well-advanced, with beam powers up to 10 MW for cyclotrons and 100 MW for linacs appearing feasible, beam losses and, most importantly, beam trip frequency must be further reduced to satisfy activation, fast temperature fluctuation and mechanical stress criteria for sensitive structures.
- Various problems related to accelerator-reactor coupling have still to be investigated. Thereby, special attention has to be given to the target and especially the beam window, as these components are subjected to complex stress, corrosion and irradiation conditions which are not encountered in normal reactors.

In the area of control and dynamic response, the following issues must be investigated:

- Controlling an ADS with beam power rather than an absorber-based reactivity compensation system reduces the potential of the core for reactivity-induced transients. For a sub-critical TRU burner, however, this advantage has to be balanced against the economic penalty arising from the high burn-up reactivity loss, which implies a higher beam current to maintain power at the end of the reactor cycle. The comparison is complicated because it also involves the balancing of safety-grade requirements for the two control systems.
- In contrast to the static behaviour of sub-critical cores, their response to reactivity and source transients is not yet well studied. The presence of an external neutron source which can vary very rapidly, in combination with very weak reactivity feedback, implies fast and (depending on the sub-criticality level) large responses to accelerator trips and control actions, which put additional demands on the control actuators, the fuel behaviour, and the heat removal processes. In particular, the fuel should be capable of buffering the respective heat balance disturbances.
- If a hypothetical core disruptive accident cannot be excluded deterministically, a prompt negative feedback mechanism must be developed to quench it.

Fuel cycle requirements

Important technological challenges also arise for the fuel cycle of a transmutation system. These are a direct consequence of the goal of transmutation, which implies the contamination of the fuel cycle by high concentrations of minor actinides. A central issue is the reprocessing of the fuel, but fuel fabrication and handling also pose new problems. The respective conclusions can be summarised as follows:

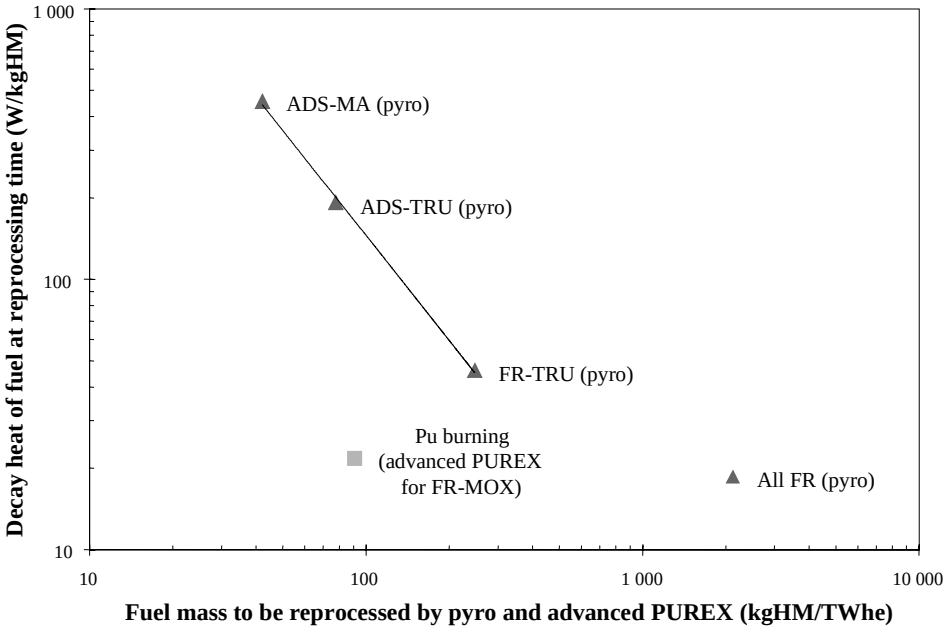
- Transmutation systems involve unusual fuels with high decay heat and neutron emission. A significant effort is required to demonstrate the manufacturability, burn-up behaviour, and reprocessability of these fuels. ADS fuels are particularly enriched in minor actinides and can probably be reprocessed only with the help of pyrochemical methods. These methods have to be further developed to tolerate from ten to more than twenty times higher decay heat levels than those encountered in the pyrochemical reprocessing of fast reactor fuels.
- The introduction of pyrochemical processing technologies at the industrial level will require the development of new process flowsheets and the use of potentially very corrosive reagents in hostile environments. These processes will generate chemical and radiological hazards which will have to be mitigated.
- PUREX aqueous reprocessing can be considered as valid for the FR-MOX fuel in the plutonium-burning and double strata schemes. Reprocessing of this fuel within short cooling times and with the required high recovery yields, however, will require the plutonium dissolution yield to be improved and the PUREX process to be modified.
- Due to the high radioactivity of FR-MOX fuel, its handling will require measures to be taken to reduce the radiation doses in the fabrication plant and during the transportation of the fuel assemblies. The increased requirements for shielding, and the preference for short transportation paths, of multiple recycled fuels also favour the pyrochemical reprocessing method at the reactor site.

During the past few years, many of these problems, especially in the separation area, have been addressed very successfully on a laboratory scale. The application of the processes on an industrial scale, however, still requires large extrapolations.

Figure 3 summarises the requirements of the different fuel cycle schemes for advanced aqueous reprocessing technology, as needed for the multiple recycled FR-MOX fuel, and for pyrochemical reprocessing technology. Regarding the pyrochemical reprocessing requirements of the transmutation schemes, there is some compensation for a high decay heat level by a low throughput, indicating that the reprocessing challenges are not very different (the trendline in the figure illustrates that the product of the two quantities is approximately constant).

A striking feature is that the pyroprocessing requirement of the all-FR scheme is much higher than that of the transmutation schemes. This is a consequence of accommodating the driver and the blanket fuel in the same fuel rod and blending the two components before processing. The blending has the advantage of reducing the decay heat of the fuel to be reprocessed and increasing the proliferation resistance of the system, but imposes high fuel throughput, and hence also economic, penalties on the scheme. These penalties could be reduced if the blanket were separated from the driver fuel and reprocessed using PUREX or UREX technology.

Figure 3. **Advanced reprocessing requirements**



Notes:
1. The ADS-MA scheme requires both pyro and advanced PUREX reprocessing. The requirements for the latter are very similar to those for the Pu-burning scheme.
2. Decay heats have to be compared with a "normal" decay heat of about 6 W/kgHM for LWR-MOX fuel.

Fission product transmutation

Fission product transmutation was already reviewed in the status and assessment report published in 1999 [2]. The present study indicates that, apart from its interesting potential as a powerful neutron source, the ADS does not open fundamentally new perspectives on this topic. Important general conclusions are:

- Excess neutrons produced by critical and sub-critical burners can, in principle, be utilised to transmute fission products. With the neutron fluxes available in these systems, it is theoretically possible to transmute the long-lived fission products; the transmutation of the more abundant short-lived fission products, however, is impracticable due to insufficient transmutation rates. This means that transmutation, in principle, allows the mitigation of the long-term risk from fission products in a geologic repository, but cannot significantly reduce the heat generation and mass of the disposed fission products.
- Minimising the fraction of specialised transmuters in the reactor park can result in an insufficient neutronic potential for transmuting the long-lived fission products of the entire park. If the transmutation would be limited to ^{129}I and ^{99}Tc, all TRU burning strategies could, theoretically, accomplish the task.
- In practice, the necessity of isotopic separation, as well as difficulties in the preparation of targets, present difficult obstacles for fission product transmutation, which currently reduces the number of candidate nuclides to only one or two, i.e. ^{99}Tc and, possibly, ^{129}I (so far, the feasibility has been established only for ^{99}Tc). This means that, for the remaining long-lived fission products, partitioning followed by immobilisation in a specially stable matrix may remain the only realistic method for reducing their radiological impact.

R&D needs

Developing advanced reactors and fuel cycles to a point where they could be deployed in a technically satisfactory and cost-effective manner can be expected to require long lead-times. The study concludes that, in order to keep the P&T option open, focused R&D should be continued on critical and sub-critical fast reactors, reprocessing technologies, advanced fuels, structural and coolant materials, and irradiation targets containing transmutable elements.

Thereby, emphasis should be placed especially on:

- Basic R&D is needed for the new FR and ADS in the fields of nuclear data and neutronic calculations, fuel technologies, structural materials, liquid metals, reprocessing technologies, target materials and high power accelerators (the last two only for ADS).
- Experimentation on fuels, as no concept can be considered seriously if the appropriate fuels are not defined and proven, i.e. fabricated, irradiated, and reprocessed.
- The continued availability of fast-spectrum irradiation facilities.
- The demonstration at appropriate scale of the performance of pyrochemical processes, in order to assess in more detail the technical-economic viability of the respective fuel cycle options.
- A clarification of the advantages and disadvantages of different coolants for fast-spectrum systems.
- Improved modelling tools to simulate materials behaviour under mixed irradiation conditions and, possibly, high temperatures.
- Safety analyses of ADS.

In addition to this R&D, countries embarking on an ADS-based fuel cycle strategy should envisage a demonstration experiment which allows the ADS concept to be validated from operation and safety viewpoints.

Finally, it should be emphasised that a satisfactory answer to the crucial question as to whether the benefits from P&T can outweigh the necessary technological and financial investments will require a substantial strengthening of the effort in the area of performance assessment studies for geological repositories using a P&T source term.

NOTE DE SYNTHÈSE

Champ de l'étude et principaux messages

Les risques à long terme que présentent les déchets radioactifs résultant de la production d'énergie nucléaire sont au cœur d'un débat de longue date et des préoccupations du public dans de nombreux pays. Par rapport au cycle ouvert, la séparation et la transmutation des actinides et de certains produits de fission à vie longue pourraient atténuer la radiotoxicité des déchets de haute activité, et éventuellement la rigueur des conditions à respecter pour leur stockage dans des dépôts géologiques. Pour que la mise en œuvre de cette technologie complexe en vaille la peine, il est souhaitable que la radiotoxicité des déchets de haute activité soit divisée par cent au moins. Ceci exige la mise en place de stratégies très efficaces de réacteurs et de cycles du combustible comprenant des réacteurs rapides et/ou des systèmes hybrides sous-critiques. Les systèmes hybrides, qui peuvent améliorer la souplesse et la sûreté des systèmes de transmutation, font actuellement l'objet d'une attention accrue.

On trouvera dans cette étude une comparaison entre divers systèmes de transmutation, systèmes hybrides et réacteurs rapides, établie en fonction des propriétés des réacteurs, des spécifications des cycles du combustible, des aspects économiques et des besoins de recherche. L'analyse des principales différences entre les divers systèmes est effectuée pour des cycles du combustible représentatifs. Les stratégies étudiées recouvrent un scénario de transmutation « évolutif » dans lequel le système hybride apporte une souplesse supplémentaire dans la mesure où il permet de consommer le plutonium dans des réacteurs conventionnels et de restreindre les actinides mineurs à une petite partie du cycle du combustible, et deux stratégies novatrices d'incinération des transuraniens consistant à utiliser un réacteur rapide ou un système hybride pour gérer ensemble le plutonium et les actinides mineurs de façon à minimiser le risque de prolifération. L'originalité de cette étude tient à l'homogénéité des analyses qui ont été réalisées avec une série de paramètres définis en commun par les experts internationaux pour les réacteurs et les cycles du combustible.

Les principaux messages émergeant de l'étude, qui pourraient marquer l'évolution des politiques en matière de séparation et de transmutation sont les suivants :

- pour diviser par cent la radiotoxicité des déchets, on a besoin de cycles avec multirecyclage du combustible où les pertes de combustible soient très faibles ;
- toutes les stratégies de transmutation avec multirecyclage du combustible autorisent une réduction du même ordre de la radiotoxicité, mais le choix de la stratégie conditionne fortement les besoins du cycle du combustible ;
- les systèmes hybrides constituent d'excellent incinérateurs d'actinides mineurs « dédiés » en régime permanent et offrent une certaine souplesse dans les scénarios de transition ;
- les approches évolutives fondées sur les systèmes hybrides et les solutions innovantes faisant appel aux réacteurs rapides se révèlent être les stratégies de transmutation les plus intéressantes, techniquement comme économiquement ;

- pour exploiter toutes les potentialités des systèmes de transmutation, il faut les utiliser au moins une centaine d'années ;
- le développement industriel de la transmutation dans des systèmes hybrides passe par des études et recherches considérables sur les réacteurs sous-critiques, les combustibles avancés et les matériaux.

Contexte général

La croissance de la demande d'énergie, et en particulier de la demande d'électricité, exige que l'on reconsidère la place de la fission en tant que source d'énergie pour le long terme. C'est dans ce contexte que l'Agence pour l'énergie nucléaire de l'OCDE (AEN/OCDE) a récemment publié une étude de la compatibilité de l'énergie nucléaire avec les objectifs du développement durable et de la meilleur façon de les atteindre[1]. Bien que les réacteurs à eau ordinaire actuels (REO) soient capables de répondre à la demande d'énergie nucléaire pendant de nombreuses décennies, on aura besoin à long terme d'intégrer des réacteurs avancés, dont les réacteurs rapides, dans le système énergétique nucléaire. Le développement de ces systèmes avancés devra viser des objectifs essentiels tels que la protection de l'environnement, l'utilisation efficace des ressources et la rentabilité tout en répondant à des préoccupations socio-politiques comme la prolifération.

Aux premiers jours de l'énergie nucléaire, la production d'électricité dans les REO et les réacteurs rapides était jugée concurrentielle par rapport aux autres modes de production électrique. À l'époque, on estimait que le déploiement de l'énergie nucléaire serait limité par les ressources en uranium, et les déchets radioactifs, en quantité limitée, étaient jugés moins préoccupants qu'ils ne le sont aujourd'hui. Cette conception initiale supposait l'introduction rapide de RNR classiques brûlant un mélange d'oxydes d'uranium et de plutonium avec un cycle entièrement fermé dans le cas du plutonium mais non dans celui des actinides mineurs, neptunium, américium et curium, qui sont au moins aussi radiotoxiques que le plutonium. La fermeture totale du cycle du combustible par le recyclage des actinides mineurs était également envisagée à l'époque sans recevoir toutefois beaucoup d'attention parce que la teneur énergétique des actinides mineurs ne présente pas d'intérêt économique.

Aujourd'hui, l'uranium est toujours abondant, mais les déchets radioactifs inquiètent de plus en plus le public. Le moment est donc venu d'essayer d'avancer vers l'objectif ultime, à savoir un cycle totalement fermé reposant sur des RNR, en passant par l'étape intermédiaire d'un système de transmutation. La séparation et la transmutation des actinides et des produits de fission aujourd'hui rejetés avec les déchets amélioreraient la « propreté radiologique » de l'énergie nucléaire et permettraient de satisfaire l'une des conditions majeures pour que le système énergétique nucléaire soit plus respectueux de l'environnement. Il conviendra bien sûr de démontrer non seulement la faisabilité technique d'un tel système mais sa faisabilité économique.

Études antérieures et démarche adoptée

Devant l'intérêt manifesté par les pays Membres et compte tenu des activités déjà entreprises dans ce domaine, en 1989, l'Agence pour l'énergie nucléaire a lancé un programme d'étude à long terme de la séparation et de la transmutation recouvrant un large éventail de problèmes techniques et scientifiques[2]. Pour renforcer la collaboration internationale, un programme d'échange d'informations a été mis sur pied, et une première étude systémique de la séparation et de la transmutation a été

1. L'énergie nucléaire dans une perspective de développement durable, OCDE, 2000.
2. On trouvera à l'annexe C un historique des travaux sur la séparation et la transmutation menés dans les pays Membres de l'Agence pour l'énergie nucléaire et dans les organisations internationales.

réalisée de 1996 à 1998 [2]. Cette étude consistait pour l'essentiel en une récapitulation des progrès dans ce domaine et des avantages éventuels de la séparation et de la transmutation pour la gestion des déchets. Elle comportait une analyse de certaines stratégies du cycle, dont le recyclage du plutonium et l'incinération complémentaire des actinides mineurs dans des systèmes de réacteurs dédiés. Cependant, elle n'abordait ni les stratégies de transmutation les plus efficaces dans des cycles du combustible entièrement fermés, ni le rôle spécifique des systèmes hybrides dans ces cycles. Cette deuxième étude systémique de la séparation et de la transmutation vise à compléter la première étude. Plus précisément, il s'agit cette fois de clarifier, au travers d'une étude comparative, les rôles et mérites relatifs des réacteurs rapides et des systèmes hybrides à spectres rapides dans des cycles fermés mais aussi d'évaluer l'état d'avancement des systèmes hybrides, l'accent étant mis sur la technologie (réacteur et cycle du combustible), la sûreté, l'économie et la faisabilité générale du réacteur et du cycle du combustible.

Pour quantifier les avantages et inconvénients des différentes stratégies d'incinération du plutonium et des actinides mineurs, sept cycles du combustible ont été choisis et comparés au cycle ouvert actuel (voir figure 1). Il s'agit de « cycles » génériques qui représentent différentes grandes stratégies avec des variantes possibles en fonction des préférences nationales.

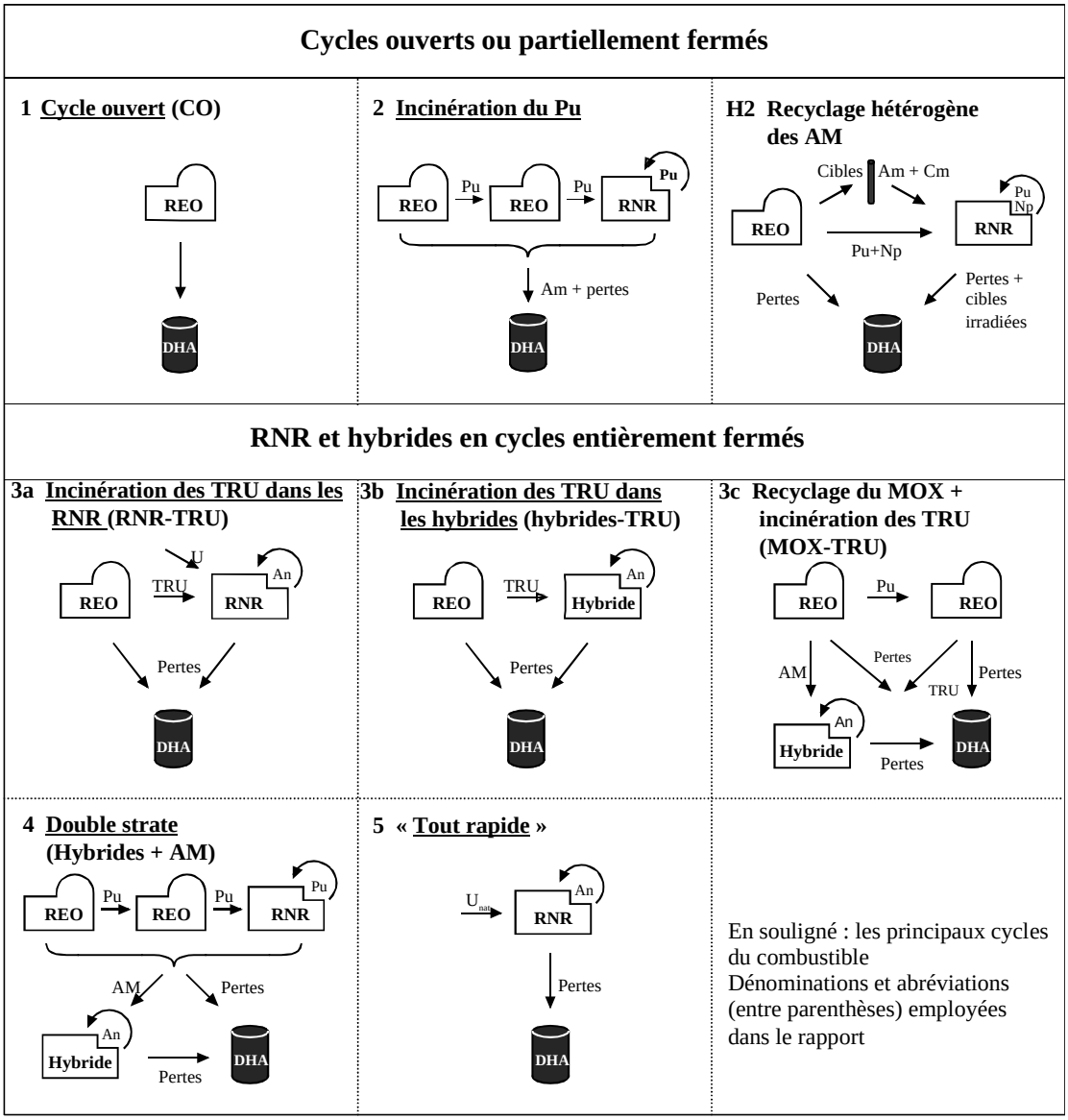
Les cycles 3a, 3b, 3c et 4 permettent de mettre en évidence les principales différences entre les stratégies de fermeture totale du cycle fondées sur les rapides et sur les hybrides. Deux cycles partiellement fermés sont également étudiés : l'intérêt du cycle 2 tient à l'importance de la question de l'incinération du plutonium et au fait que la transmutation est toujours précédée ou accompagnée de l'incinération du plutonium. Le cycle avec recyclage hétérogène H2 constitue une alternative possible au cycle fermé bien qu'offrant de moindres possibilités de transmutation. Enfin, la stratégie « tout rapide » représente l'objectif à long terme du développement de l'énergie nucléaire. Seuls sont étudiés les réacteurs incinérateurs fonctionnant avec des combustibles solides et optimisés pour obtenir un taux d'incinération élevé de façon à pouvoir desservir un parc fortement doté en REO. L'originalité de cette comparaison tient à l'harmonisation des méthodes de calcul et l'utilisation, pour les réacteurs et les cycles, de paramètres évalués spécialement pour cette étude.

Critères de durabilité

La comparaison porte sur trois dimensions de la durabilité, l'efficacité d'utilisation des ressources, la protection de l'environnement et la rentabilité. Les principaux critères portés sur le second axe sont les quantités de métal lourd et de TRU évacués dans le dépôt (pertes) ainsi que leur radiotoxicité. Les principaux résultats que l'on trouvera présentés sur la figure 2 peuvent être résumés comme suit :

- toutes les stratégies de transmutation dans des cycles entièrement fermés permettent, en principe, des réductions identiques de l'inventaire des actinides et de la radiotoxicité à long terme des déchets de haute activité, et en tout cas comparables à celles obtenues avec une stratégie purement RNR. Il n'existe par conséquent pas de différences marquées entre les possibilités offertes par les RNR et par les systèmes hybrides ;

Figure 1. **Récapitulation des cycles du combustible analysé**

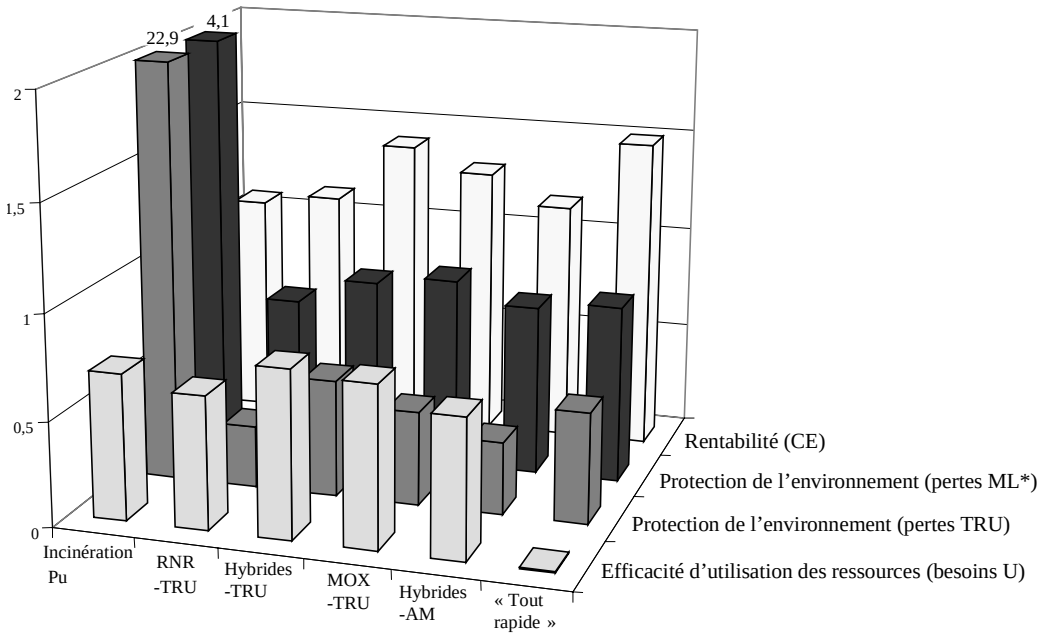


Note :
Les « principaux » cycles du combustible correspondent à des stratégies fondamentales. Il est possible de les combiner. Par exemple, le cycle MOX-TRU associe des éléments du cycle « hybrides-TRU » et des cycles d'incinération du plutonium. L'analyse de ces cycles a été effectuée à l'aide d'une même bibliothèque de données nucléaires et d'un même système de codes de réacteur.

- avec les paramètres adoptés pour les réacteurs et les cycles du combustible, ces stratégies permettent une division par plus de cent de la radiotoxicité des déchets, et une réduction plus forte des quantités de métal lourd et de TRU mises en dépôt (pertes), par rapport au cycle ouvert. Ceci suppose le multirecyclage des combustibles, des taux de combustion élevés et des pertes très limitées lors du retraitement et de la fabrication du combustible. En ce qui concerne ce dernier paramètre, le pourcentage de 0,1 %, valable actuellement pour l'uranium et le plutonium, a été appliqué à l'ensemble des actinides, bien que cela représente un objectif ambitieux ;

- du point de vue de la production de déchets de haute activité et de la technologie, les cycles RNR-TRU et hybrides-AM sont équivalents. Le premier peut évoluer progressivement vers une stratégie tout rapide, mais exige un investissement initial plus lourd dans les technologies des réacteurs rapides et des cycles du combustible avancés. Le second restreint les actinides mineurs à un petit flux secondaire du cycle pour lequel on a néanmoins besoin d'une technologie très innovante. Les systèmes hybrides présentent l'avantage d'incinérer des actinides mineurs purs sans dégrader les paramètres de sûreté du cœur du réacteur ;
- l'analyse économique montre que la compétitivité de la technologie de la transmutation dans des systèmes hybrides peut être accrue si l'on incinère le maximum de plutonium dans des réacteurs classiques, c'est-à-dire des REO alimentés en MOX et des rapides. Cela favorise le cycle hybrides-AM qui, avec le cycle RNR-TRU, présente également les plus faibles coûts de production d'électricité de tous les cycles avec transmutation. Dans ces cas, la hausse du coût de l'électricité de la stratégie de séparation-transmutation par rapport au cycle ouvert est relativement faible, de 10-20 %. Bien qu'inacceptable dans la conjoncture actuelle, une pareille hausse de coût reste limitée et pourrait se révéler abordable si les prix des combustibles fossiles devenaient moins intéressants ou si la société décidait de valoriser davantage la diminution de la radiotoxicité des déchets.

Figure 2. **Comparaison en fonction des critères de durabilité**



Cycles du combustible

*ML = Métaux lourds.

Besoins U : besoins en uranium naturel par rapport à un cycle ouvert.
Pertes ML : pourcentage de transuraniens mis en dépôt (% du cycle ouvert).
Pertes ML : pourcentage de métal lourd évacué (% du cycle ouvert).
CE : coût de l'électricité par rapport au cycle ouvert (cas nominal).

Note : dans le cas du cycle d'incinération du Pu, les pertes de TRU et de ML ne sont pas à l'échelle.

Cette étude montre également que, en soi, l'incinération du plutonium est utile dans une optique de gestion du plutonium mais qu'elle ne remplit pas les conditions requises pour les stratégies de transmutation étant donné que la radiotoxicité des déchets de haute activité n'est divisée que par cinq.

Le recyclage hétérogène de l'américium et du curium dans des aiguilles cibles spéciales évacuées après irradiation, représenté par le cycle de recyclage hétérogène des actinides mineurs, est techniquement moins contraignant qu'un cycle du combustible fermé mais aussi deux fois moins efficace pour atténuer la radiotoxicité. Cette solution est étudiée pour la transmutation à court terme sans système hybride.

S'agissant des périodes de transition, l'étude confirme que les contraintes physiques associées à la production et la destruction en pile et hors pile des inventaires de combustible imposent des constantes de temps très longues pour l'introduction et l'abandon définitif de tout système électro-nucléaire avancé quel qu'il soit, et que la technologie de la séparation et de la transmutation ne pourra tenir ses promesses que si elle est introduite avec l'intention de l'exploiter au moins un siècle. En particulier, on ne pourra en tirer tout le bénéfice attendu que si l'inventaire de TRU du système est finalement incinéré et non évacué avec les déchets. À cet égard, il convient de mentionner que la stratégie hybrides-TRU se caractérise par un inventaire en TRU moindre en régime permanent et, dans l'hypothèse d'une sortie du nucléaire, par le fait que cet inventaire pourrait être incinéré plus rapidement qu'avec les autres stratégies.

Enfin, on retiendra que toutes les stratégies de transmutation reposant sur un parc de réacteurs comportant des REO exigent des ressources en uranium équivalentes et produisent des quantités d'uranium résiduel aussi importantes qu'un parc de REO en cycle ouvert. Si l'uranium résiduel n'est pas considéré comme une ressource pour les futurs réacteurs rapides, il faudra aussi évaluer ses répercussions radiologiques à long terme.

Technologie et sûreté des systèmes hybrides

Si les réacteurs rapides et les systèmes hybrides affichent des résultats équivalents pour ce qui est de leur impact sur l'environnement, en revanche leurs caractéristiques technologiques, leur fonctionnement et leurs niveaux de sûreté sont très différents.

Deux avantages des systèmes hybrides nous intéressent tout particulièrement ici :

- le système hybride sous-critique permet de concevoir des cœurs de réacteurs dont les caractéristiques de fonctionnement ne seraient pas satisfaisantes dans d'autres circonstances. En particulier, la possibilité d'exploiter un incinérateur d'actinides sous-critique avec un combustible exempt d'uranium (ou de thorium) permet de maximiser l'efficacité de l'incinérateur et, par conséquent, d'utiliser un minimum d'installations de transmutation spécialisées dans le parc de réacteurs ;
- en outre, ce concept permet d'ajuster, c'est-à-dire d'élargir, la marge de réactivité à la criticité prompte, ce qui limite les possibilités d'excursion de puissance du cœur. Cette propriété s'avère particulièrement intéressante dans les incinérateurs d'actinides mineurs où la marge est seulement de moitié de celle d'un réacteur rapide normal si le cœur fonctionne en mode critique. Les cœurs d'incinérateurs de TRU sont donc moins dégradés de ce point de vue.

Il faut toutefois mettre en balance les avantages des systèmes hybrides avec les défis techniques que représente le couplage d'un réacteur et d'un accélérateur et la nécessité de faire face à de nouveaux types de transitoires d'exploitation et d'accidents.

Les premiers appellent une analyse des problèmes suivants :

- bien que la mise au point des accélérateurs ait progressé, puisqu'il paraît désormais possible d'utiliser des faisceaux de 10 MW, dans le cas des cyclotrons, et de 100 MW, dans celui des accélérateurs linéaires, les pertes de faisceau et, plus important, la fréquence des instabilités de faisceau doit être encore réduite si l'on veut remplir les critères d'activation, de fluctuation rapide de la température et de contraintes mécaniques pour les structures sensibles.

- il faut encore approfondir divers problèmes liés au couplage de l'accélérateur au réacteur. En particulier, la cible, et plus précisément la fenêtre, méritent qu'on leur accorde une attention particulière car ce sont des composants soumis à des conditions de contrainte, de corrosion et d'irradiation complexes que l'on ne rencontre pas dans des réacteurs normaux.

S'agissant du pilotage et du comportement dynamique de ces systèmes, les questions suivantes doivent être étudiées :

- le pilotage d'un système hybride par un faisceau plutôt que par un système de compensation de la réactivité utilisant un absorbeur limite les possibilités de transitoires de réactivité. Dans un incinérateur de transuraniens sous-critique, cependant, cet avantage est compensé par la pénalité économique que constitue la perte de réactivité aux taux de combustion élevés, ce qui exige d'augmenter l'intensité du faisceau pour maintenir la puissance à la fin du cycle en réacteur. La comparaison est compliquée car elle suppose aussi que l'on puisse comparer les exigences de sûreté des deux systèmes de pilotage ;

- contrairement à leur comportement statique, on connaît mal le comportement des cœurs sous-critiques lors de transitoires de réactivité ou de transitoires touchant les sources. L'utilisation d'une source externe de neutrons qui peut varier très rapidement, associée au fait que la contre-réaction de la réactivité soit très faible, impliquent des réactions très brusques et violentes (suivant le niveau de sous-criticité) à des instabilités de l'accélérateur et à des contrôles-commandes d'où des contraintes supplémentaires pour les mécanismes de commande, le combustible et les procédés d'évacuation de la chaleur. Le combustible en particulier doit être capable d'amortir les différentes perturbations de l'équilibre thermique.

- si la possibilité d'un accident provoquant la dislocation du cœur ne peut pas être exclue de manière déterministe, il faudra mettre au point un mécanisme de contre-réaction de réactivité prompte.

Spécifications du cycle du combustible

Le cycle du combustible d'un système de transmutation pose aussi d'importants problèmes technologiques qui découlent directement de l'objectif même de la transmutation qui suppose la contamination du cycle du combustible par de fortes concentrations d'actinides mineurs. L'un des principaux est le retraitement du combustible, mais la fabrication, la manutention et le transport soulèvent également de nouvelles difficultés. Les conclusions concernant ces différent aspects peuvent être résumées comme suit :

- les systèmes de transmutation font en général appel à des combustibles inhabituels caractérisés par de fortes chaleurs de décroissance et émission de neutrons. La démonstration de la capacité de fabriquer et de retraiter ces combustibles et de leur comportement à des taux de combustion élevés nécessitera d'importants travaux. Les combustibles utilisés dans les systèmes hybrides sont particulièrement enrichis en actinides mineurs et ne pourront

probablement être retraités que par les méthodes pyrochimiques. Or ces méthodes doivent encore être perfectionnées pour pouvoir supporter des chaleurs de décroissance dix à plus de vingt fois supérieures aux niveaux rencontrés lors du retraitement pyrochimique des combustibles de réacteurs rapides.

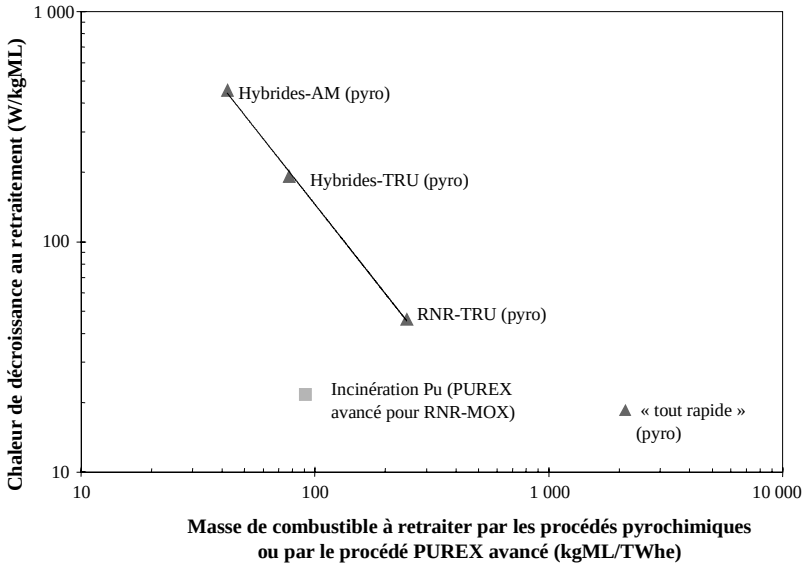
- l'introduction à échelle industrielle des technologies de retraitement pyrochimiques passe par la mise au point de nouveaux schémas de procédé et le recours à des réactifs éventuellement très corrosifs dans des environnements hostiles. D'où de nouveaux dangers chimiques et radiologiques contre lesquels il faudra prendre des mesures ;

- le procédé aqueux de retraitement PUREX peut être considéré comme valable pour le combustible RNR-MOX dans les cycles d'incinération du plutonium et le cycle à double strate. Le retraitement de ce combustible après un court temps de refroidissement avec le rendement de séparation exigé nécessite cependant une amélioration du rendement de dissolution du plutonium et une modification du procédé PUREX ;

- en raison de la forte radioactivité du RNR-MOX, des mesures s'imposent pour limiter les doses de rayonnement dans l'usine de fabrication et durant le transport des assemblages combustibles. Les contraintes de blindage et la nécessité d'écourter les transports de combustibles multirecyclés plaident en faveur du retraitement pyrochimique sur le site même du réacteur.

Ces dernières années, une bonne partie de ces problèmes, notamment ceux posés par la séparation, ont été résolus à l'échelle du laboratoire. Pourtant, la mise en œuvre des procédés à l'échelle industrielle exige aujourd'hui encore d'importantes extrapolations.

Figure 3. **Besoins de retraitement avancé**



Notes :

1. Le cycle hybrides-AM exige tant un retraitement pyrochimique qu'un procédé PUREX avancé. Pour ce dernier, les besoins sont très proches de ceux d'un cycle d'incinération du Pu.
2. Les chaleurs de décroissance sont à comparer à une valeur « normale » de 6 W/kgML dans le cas du combustible REO-MOX.

La figure 3 récapitule les exigences des différents cycles du combustible en matière de procédé avancé de retraitement aqueux, dans le cas combustible RNR-MOX multirecyclé, et de retraitement pyrochimique. S'agissant du retraitement pyrochimique dans les stratégies de transmutation, la forte chaleur de décroissance est légèrement compensée par les petites quantités produites, si bien que les difficultés de retraitement restent globalement du même ordre que pour les autres solutions (le produit des deux valeurs étant à peu près constant, comme le montre la figure 3).

Fait remarquable, les besoins de traitement pyrochimique du cycle tout rapide sont nettement supérieurs à ceux des cycles de transmutation ce qui s'explique par la nécessité d'installer dans la même aiguille le combustible nourricier et le combustible de couverture et de mélanger les deux composants avant le traitement. Le mélange présente l'avantage d'atténuer la chaleur de décroissance du combustible à retraiter et de renforcer la résistance à la prolifération du système, mais, en revanche, il impose une forte production de combustible avec les conséquences économiques que cela suppose. Cette pénalité économique pourrait être allégée en séparant la couverture du combustible nourricier et en la retraitant par les procédés PUREX ou UREX. Du seul point de vue de la chaleur de décroissance, il serait évidemment préférable d'éviter la stratégie de transmutation et de passer directement à la stratégie des réacteurs rapides.

Transmutation des produits de fission

La transmutation des produits de fission avait été étudiée dans le rapport de synthèse publié en 1999. La présente étude montre que en dehors de son intérêt potentiel, comme puissante source de neutrons, le système hybride n'ouvre pas de perspectives très nouvelles dans ce domaine. On retiendra cependant que :

- l'excès de neutrons produits dans les incinérateurs critiques et sous-critiques peut en principe servir à transmuter les produits de fission. Avec les flux de neutrons qui existent dans ces systèmes, il est théoriquement possible de transmuter les produits de fission à vie longue. La transmutation des produits de fission à vie courte, plus abondants, n'y est cependant pas réalisable, les taux de consommation étant insuffisants. En d'autres termes, la transmutation permet en principe d'atténuer le risque que présentent à long terme les produits de fission évacués dans un dépôt géologique, mais elle n'est pas capable de réduire dans de fortes proportions la production de chaleur ni la masse des produits de fission évacués ;

- maximiser le ratio du nombre de réacteurs classiques au nombre de réacteurs incinérateurs peut se solder par un bilan neutronique insuffisant pour pouvoir transmuter les produits de fission à vie longue de tout un parc nucléaire. Cela vaut en particulier pour les incinérateurs d'actinides mineurs dans le cycle de séparation-transmutation d'une stratégie à double strate. Les stratégies d'incinération des TRU, et plus précisément la stratégie RNR-TRU, présentent de bonnes capacités de transmutation des produits de fission. Si l'on se contente de transmuter l'^{129}I et le ^{99}Tc, toutes les stratégies d'incinération peuvent en théorie s'acquitter de cette mission ;

- dans la pratique, la nécessité d'une séparation isotopique de même que les difficultés de réalisation des cibles constituent d'importants obstacles à la transmutation des produits de fission si bien que l'on ne compte aujourd'hui qu'un ou deux éléments transmutables (à ce jour, la faisabilité de la transmutation a été démontrée pour le ^{99}Tc seulement). De ce fait, la séparation, suivie de l'immobilisation dans une matrice particulièrement stable, pourrait bien être la seule méthode réaliste pour réduire l'impact radiologique des autres produits de fission à vie longue.

Besoins de R&D

La mise au point de réacteurs et de cycles du combustible avancés jusqu'au stade où leur exploitation peut s'effectuer dans des conditions techniques et économiques satisfaisantes est une entreprise de longue haleine. L'étude conclut que, pour maintenir ouverte la voie de la séparation et de la transmutation, il faudra poursuivre des études et recherches ciblées sur les réacteurs rapides critiques et sous-critiques, les combustibles avancés et les cibles d'irradiation contenant des éléments transmutables. L'accent devrait être mis par conséquent sur :

- l'expérimentation des combustibles, dans la mesure où il sera impensable d'envisager sérieusement l'un ou l'autre concept tant que les combustibles pertinents n'auront pas été définis et démontrés, c'est-à-dire caractérisés, fabriqués, irradiés et retraités ;

- la disponibilité d'installations d'irradiation à spectres rapides, sachant qu'il n'est pas nécessaire que ces installations soient des systèmes hybrides ;

- la démonstration à une échelle appropriée des performances des procédés pyrochimiques de manière à affiner l'évaluation de la viabilité technico-économique des diverses options du cycle du combustible ;

- la mise en évidence des avantages et inconvénients des différents caloporteurs utilisables dans les systèmes à spectres rapides.

- des outils de modélisation améliorés permettant de simuler le comportement des matériaux dans diverses conditions d'irradiation et, éventuellement, à haute température ;

- des analyses de sûreté des systèmes hybrides afin d'étudier les mécanismes pouvant provoquer la dislocation du cœur, si l'éventualité de tels accidents ne peut être exclue de manière déterministe.

Enfin, il convient de souligner que, indépendamment des travaux de R&D mentionnés ci-dessus, on ne saura si les mérites de la séparation et de la transmutation compensent les investissements technologiques et financiers nécessaires qu'au prix d'un renforcement substantiel des évaluations des performances de dépôts géologiques pour un terme source représentatif de la séparation et la transmutation.

1. INTRODUCTION

1.1 Nuclear energy development in the past and objectives for the future

Today's nuclear energy system is the result of a fifty-year development during which this technology has reached industrial maturity and became a reliable resource for our electricity needs. Most of this development has been concentrated on light water reactor (LWR) concepts (pressurised water reactors and boiling water reactors) and their fuel cycle. In the OECD, several other reactor concepts have also been studied (and some prototypes have been constructed and operated) but, with the exception of the CANDU reactor,[1] were not developed to internationally commercial systems.

The success of the LWR is based on the early recognition that natural fissile material were considered scarce and that nuclear energy could develop only if systems with low fissile inventories per unit power would be built in the start phase. LWRs, as initially developed for naval applications, fulfilled this criterion and used simple and relatively cheap technology that enabled a first generation of power stations to be constructed rapidly. The necessary uranium enrichment technology was available from the military development. The significant plutonium generation in LWR fuels was considered to be an asset because plutonium is an excellent fuel for fast reactors and the anticipated deployment of fast reactors around the turn of the century would have required large fissile inventories.[2] In the early days of nuclear energy, however, the back-end of the fuel cycle was not given the same attention as the reactors, and the concept of geologic disposal of radioactive waste was not yet questioned by the public.

Because the known uranium resources increased with prospecting and the growth of nuclear energy did not meet the early expectations, uranium became cheap and the envisaged rapid introduction of fast reactors did not come to pass. In many countries, a once-through fuel cycle developed where spent fuel is accumulating in spent fuel storage pools and intermediate storage facilities. Other countries embarked on a reprocessing fuel cycle, taking advantage of the PUREX technology,[3] which was also available from the military application, to separate plutonium and uranium. Whereas some of the recovered plutonium is recycled in the form of uranium-plutonium mixed-oxide (MOX) fuel in LWRs, the remaining mix of minor actinides and fission products is conditioned for final waste disposal.[4] Today, after some forty years of nuclear energy deployment, most countries with a nuclear energy programme have a growing stock of spent fuel, or separated plutonium and vitrified high-level waste (HLW), where the further management of this material is uncertain.

This situation is particularly uncomfortable since, in the meantime, the back-end of the fuel cycle has become the main focus of much of the criticism against nuclear energy, mostly oriented towards the final storage of spent fuel or HLW. There is a consensus within the OECD Member countries that geologic disposal, in one or another form, is an appropriate solution to protect humans and their

1. CANDU: a heavy-water-moderated and -cooled reactor developed and widely used in Canada.
2. Fast reactor: a nuclear reactor in which most of the fissions are caused by fast neutrons. It contains no moderator and is capable of generating more fissile material than it consumes.
3. PUREX: generic name for solvent-extraction processes using TBP as the extractant.
4. Minor actinides (MA): neptunium (Np), americium (Am), curium (Cm) and higher-Z actinides.

environment in the far-reaching future. However, difficulties encountered in siting, constructing and licensing repositories, not to mention public opposition against nuclear waste, have caused delays in the construction of these facilities.

On the other hand, the growing awareness that the contribution from nuclear energy to sustainable development cannot be ignored calls for a re-evaluation of nuclear development strategies for the coming decades. Environmental friendliness, cost-effectiveness and resource-efficiency will be essential axes in such evaluations, and nuclear holds development potential along all these axes. It is clear that nuclear energy could play an important role more easily if it responds also to the concerns of the society. Considering this situation, three objectives can be put forward for future nuclear energy systems:

- While the known uranium resources give us some hundred more years of supply with today's nuclear power park, we should not forget that LWRs use less than one per cent of the energy content of the mined uranium, the rest of the energy content being stored in spent fuel and depleted uranium from the enrichment process. The requirement of resource-efficiency will increase pressure to move to fuel cycles which can exploit a higher fraction of the energy content of the mined uranium. The growing stockpiles of spent fuel, reprocessed plutonium and uranium, and depleted uranium hold in that respect a large amount of energy for the next millennia, sufficient to meet most of mankind's energy needs.

- Emission of greenhouse gases as well as generation of waste in general has become a major public concern. Nuclear energy does not produce greenhouse gases, but the generated highly radiotoxic waste has animated the public debate during the past years. More environmentally friendly ways of producing nuclear energy by reducing the amount of waste, and especially HLW, would enhance the potential of nuclear energy for the future.

- While LWRs will continue to cover a large fraction of the nuclear energy demand, some advanced reactors utilising the remaining energy content of spent fuel and dealing with the actinide waste should be integrated into the system to assist nuclear energy fulfilling its long-term promises. Since the economic viability of advanced reactors has not yet been proven, the ratio of conventional to advanced reactors should remain high in the short- to medium-term in order to reduce the incremental cost per unit power produced.

1.2 Fuel cycle options and paths to the future

Bearing these objectives in mind, energy policy makers will have to decide on a path forward while taking fuel cycle constraints dictated by national policies into account. In principle, the following strategies can be envisaged:

- A first strategy is to remain with the once-through fuel cycle. This could be the choice, for instance, for countries with a modest nuclear energy programme and no recycling infrastructure. If the spent fuel cannot be sent to an international fuel cycle centre, a national plan for direct disposal of the fuel, including the demonstration of the long-term safety of the geologic repository, will have to be implemented.

- A second strategy, the plutonium burning strategy, is to close the fuel cycle for plutonium with the principal motivation to utilise the plutonium and not dispose it with the spent fuel. The plutonium can be recycled, first in LWRs and later in fast reactors. Minor actinides and fission products are vitrified and disposed in geologic repositories. Since the uranium and the plutonium are separated, the volume of the HLW is reduced. However, in terms of the

uranium requirement and the overall radiotoxicity of the HLW,[5] the benefit of the plutonium burning strategy is small and the long-term safety of the repository will still have to be demonstrated.

- A third strategy, the transmutation strategy, is to close the fuel cycle of conventional, i.e. LWR-dominated, reactor parks also for the minor actinides by recycling all actinides homogeneously or heterogeneously in existing and innovative reactor types. If the fuel reprocessing losses are sufficiently small, complete closure of the fuel cycle would result in a considerable reduction of the actinide content, and hence the long-term radiotoxicity, of the HLW. Innovative reactor types and new recycle infrastructures, including the pyrochemical or "dry" reprocessing technique, would be necessary, especially for burning highly concentrated transuranics (TRU) and minor actinides.[6] If some of the long-lived fission products could also be transmuted or separately conditioned, the radiotoxicity of the remaining HLW would decay within a few hundred years, meaning that repository designs for such HLW may meet licensing requirements more easily.

- Lastly, the fast reactor strategy, in contrast to the first three strategies, aims primarily at improving the uranium utilisation and, to this end, substitutes LWRs by fast reactors at a large scale.[7] Conventional fast reactors have a fuel cycle which is closed for plutonium, but leaves the minor actinides in the HLW stream and hence still generates a non-negligible amount of actinide waste. However, this actinide waste can be much reduced, if all actinides are recycled as this is realised in the Integral Fast Reactor (IFR) concept [3]. The pyrochemical reprocessing method is inherently suited for such a fully closed fuel cycle because it combines the fission product extraction with the co-processing of the actinides.

The achievements of these strategies with regard to the two axes "radiological cleanliness" (one of the most important features of an environmentally friendly nuclear energy system), measured in terms of actinide radiotoxicity reduction, and "uranium utilisation" are visualised in Figure 1.1. In Chapter 2, it will be shown that an actinide toxicity reduction by a factor of 100 relative to the once-through fuel cycle is feasible for high burn-up fuels and a fuel cycle which is optimised for small reprocessing and fabrication losses. A resource utilisation of 100% means that the mined uranium (or thorium) is completely fissioned. In particular, the figure illustrates that a future advanced nuclear energy system could deliver hundred times more energy than today's conventional nuclear energy system without any increase in the uranium consumption and the actinide waste production.

Figure 1.1 also shows paths to the future. In the early days of nuclear energy, when the uranium resource was assumed to be the limiting parameter for the nuclear development, a rapid introduction of fast reactors based on the MOX fuel and PUREX reprocessing technology (Path A in the figure) appeared imperative. However, the convential fast reactor development came to a halt in the 80s in the wake of the discovery of additional cheap uranium resources, unexpected technical difficulties with the sodium technology, and considerable cost increases in the demonstration programmes (see Chapter 4). The transition from the conventional fast reactor strategy to a fast reactor strategy with a fully closed fuel cycle was envisaged already at that time, but not given much attention because the burning of minor actinides was judged to be of a lesser priority and economically unattractive.

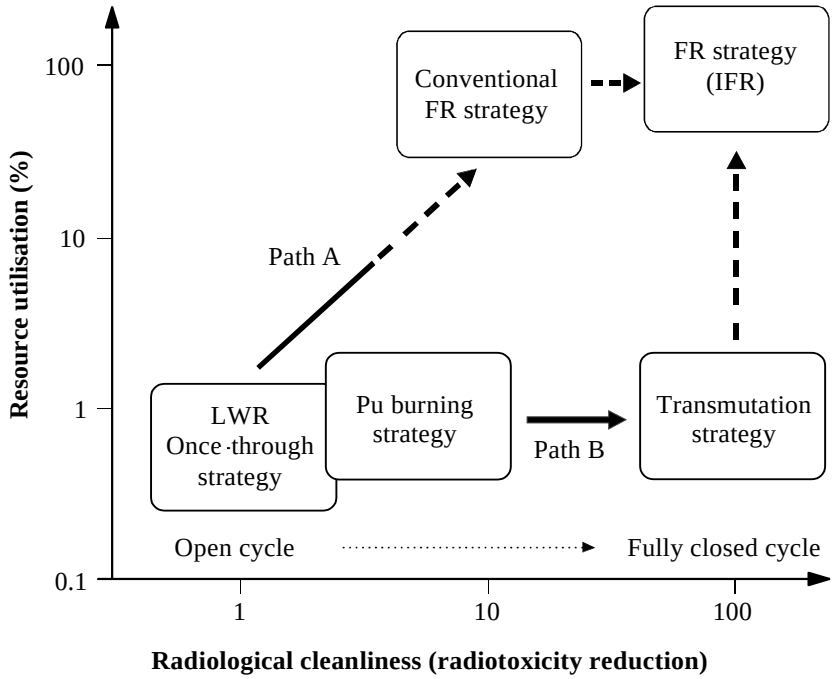
5. In the context of the present study, the term radiotoxicity is used to quantify the radiation dose to which a human would be subjected, if he would incorporate the radioactive material in drinking water. The quantity does not take any protective barriers between the material and the human into account.
6. Transuranics: actinides with a higher Z than that of uranium.
7. High-convertor LWRs (HCLWR) were not part of this study but a reactor park consisting of a suitable mix of fast reactors and such HCLWRs could also achieve this goal.

Today, while cheap uranium is still available for the coming decades, the nuclear scene is preoccupied with the nuclear waste problem as a result of the world-wide difficulties with the implementation of HLW repositories. Under these boundary conditions, which can be expected to prevail for some decades, Path B in Figure 1.1 may become the favoured path to the future. Path B attempts to reach the long-term goal of the fast reactor strategy with fully closed fuel cycle via the transmutation strategy, with or without a preceding plutonium burning phase.

The clarification of the role and added value of the accelerator-driven system (ADS) with regard to Path B, to be called the *transmutation path*, is the principal purpose of the present study.[8] The study examines and elucidates this role with the help of different transmutation "schemes", with separation or co-processing of the actinides, and assesses the technology development status with emphasis on the sub-critical system, the target, the accelerator, new fuels, alternative reprocessing methods, safety, cost/benefit aspects, and R&D requirements. ADS applications with objectives other than transmutation, such as the development of "ultra-safe" fast reactors are not in the scope of this study.

While the potential of the ADS for breeding fissile material and transmuting nuclear waste was recognised already at the beginning of the development of nuclear energy, technological limits for a long time did not permit the application of this technology at a commercial scale. Recent progress, especially in accelerator technology, has led to a renewed interest in the concept and promoted increasing international collaboration in this field.

Figure 1.1. **Nuclear energy strategies and paths to the future**



8. In the context of the present study, the accelerator-driven system is a hybrid facility consisting of a high-current proton accelerator and a sub-critical reactor.

1.3 Transmutation and role of ADS

1.3.1 Principle and benefit of transmutation

Transmutation aims at reducing the radiological impact of actinides and fission products in the HLW by nuclear transformation of troublesome long-lived nuclides in strong radiation fields.[9] Assuming that the HLW can be safely enclosed in waste containers for about a millennium, the period of concern begins about 1 000 years after the irradiation of the fuel, i.e. at a time when the majority of the fission products have decayed and the radiotoxicity of the HLW is strongly dominated by actinides. However, long-lived fission products must also be considered since they are more mobile than the actinides and, therefore, dominate the long-term risk of geologic repositories.[10]

Regarding suitable radiation sources for transmutation, the requirement for high intensity and energy efficiency means that, in the medium term, only nuclear fission reactors and spallation sources can be utilised. These sources deliver neutrons which induce transmutation reactions in the energy range from thermal up to about 20 MeV. It should be noticed that, in realistic accelerator-driven systems, high-energy spallation neutrons do not significantly contribute to the transmutation reactions, but can influence the activation of components.

The primary benefit of transmutation is a reduction in the minor actinide and long-lived fission product content of the HLW. The first and most effective step to reduce the total mass of the HLW is the transition from an LWR once-through strategy with direct disposal of the fuel elements to a plutonium burning strategy with HLW vitrification. Compared with the latter, transmutation strategies have only a modest mass reduction potential. Since the radiotoxic nuclides in the HLW can only partially be eliminated, transmutation does not make geologic disposal concepts superfluous, but must be considered as a complementary waste management method which may ease the design and licensing requirements for geologic repositories because the geosphere barrier would no longer have an important safety function.

With regard to alternative advanced waste management methods, the partitioning of individual actinides and fission products in combination with their immobilisation in special matrices, i.e. their conditioning and confinement in matrices which are more stable than glass, is worth mentioning. An advantage of such methods is their applicability to short-lived fission products which cannot be transmuted (see Chapters 2 and 8). Partitioning/immobilisation could be employed, for example, to reduce the heat load of HLW which is also an important issue for geologic repositories. However, it should be emphasised that, whereas partitioning/immobilisation is a possible alternative for reducing the long-term risk of a repository, the method is not suited for mitigating the hazard in the case of human intrusion scenarios because the total inventory of the radiotoxic nuclides remains unchanged.

1.3.2 Actinide transmutation

For the transmutation of actinides, the key reaction is the *fission reaction* which transforms long-lived, highly radiotoxic actinides into mostly short-lived, less toxic fission products. Other reactions such as capture and (n,2n) reactions just transform actinide species into other actinide species without

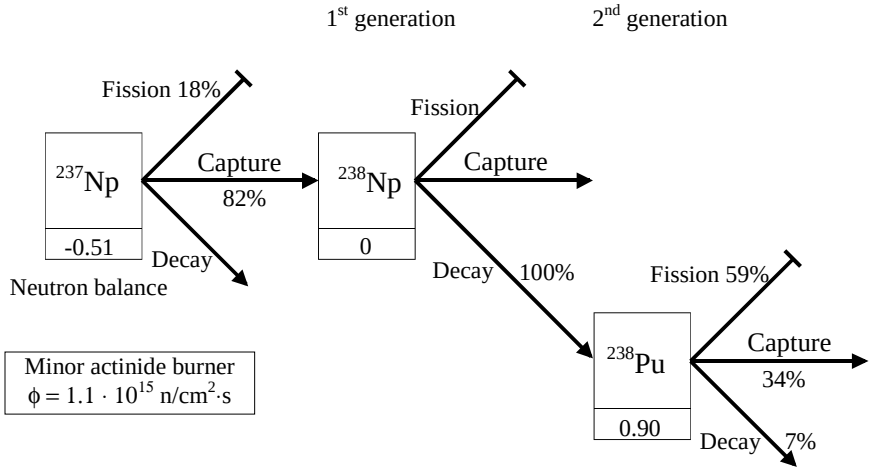
9. It should be noted that transmutation addresses only one aspect of the impact of the fuel cycle on the environment and that, in a general risk assessment study, the environmental effects arising from the fuel cycle front-end activities, secondary wastes, etc., would also have to be considered.
10. The long-term risk of a geologic repository is usually evaluated in terms of an annual individual dose for the concerned population (Sievert per year) taking the mitigation of the radiotoxic releases by the multiple barriers (waste container, engineered repository near-field, host rock) and the biosphere behaviour of the nuclides into account (see Chapter 2).

a significant effect on long-term radiotoxicity. However, these reactions are useful insofar as they transform fertile actinides with a low fission probability into fissile actinides with a high fission probability.[11] The release of additional fission energy is a small "extra gain" from the transmutation of minor actinides.

The transmutation of an actinide is completed, when the transformation chain, which involves "generations" of neutron reactions and radioactive decays, terminates with a fission. Figure 1.2 illustrates the first three steps of the transmutation process for ^{237}Np. The first transformation step consumes on average 0.51 neutrons because it is dominated by capture reactions, the second step is a simple decay, and the third step produces on average 0.90 neutrons because it is dominated by fission reactions. Up to the second generation, the neutron balance (excess) is +0.39, and 66% of the original ^{237}Np atoms are transmuted.[12] The *"overall neutron balance"*, i.e. the neutron balance over all generations until all original atoms are fissioned, is an important basic parameter which depends on the type of transmutation system. Interesting general observations regarding this parameter are that:

- Fast systems feature a significantly better overall neutron balance for the transmutation of actinides than thermal systems and provide many excess neutrons which could be utilised for other applications, e.g. the transmutation of fission products.
- The overall neutron balance of thermal systems does not constrain the transmutation of typical LWR discharged transuranic mixtures.
- A deficit in the overall neutron balance of thermal systems does not allow a complete transmutation of pure minor actinides.

Figure 1.2. **Transformation chain for the transmutation of Neptunium-237**



Points 2 and 3 mean that, from a neutron balance viewpoint, TRU burners can be designed as critical or sub-critical systems with any type of neutron spectrum, but dedicated minor actinide burners must be designed as critical or sub-critical fast reactors. It will be shown in Chapter 2 that critical and sub-critical fast reactors feature similar overall neutron balances, and that the advantages of the sub-criticality are primarily a gain in core design and operation flexibility due to the removal of the

11. The fertile actinides have a fission threshold which makes the fission probability strongly dependent on the neutron spectrum.
12. The numerical example applies to the accelerator-driven minor actinide burner described in Chapter 3.

criticality constraint and its potential to compensate degraded safety characteristics of actinide burners with a fast neutron spectrum.[13]

The equivalence between the actinide transmutation and the fission process implies that the transmutation rate in an actinide burner is limited by the thermal power. Moreover, the fuel fraction which can be transmuted in a single pass of the fuel through the burner cannot exceed the fuel burn-up. Since the burn-up for solid fast reactor fuel is limited to about 25%,[14] it is clear that an effective actinide burner cannot operate in a once-through mode, but requires a fuel cycle which allows the fuel to be recycled many times. For the maximum burn-up of 25% and recycle intervals of 6 years,[15] it takes 96 years to achieve a hundredfold waste mass reduction. This means that the feasibility of transmutation depends not only on the establishment of a suitable fuel cycle, but also on the assumption that the technology can be sustained during a period of at least hundred years. High development costs and long lead times speak also for a minimum utilisation period of this length.

Important conclusions from this discussion are that:

- An effective transmutation system calls for a fully closed fuel cycle in which all actinides are recovered with a nearly 100% efficiency and recycled.
- To realise the potential of this system, it must be operated for an extended period of at least hundred years.

Under the present market forces, thermal, and especially fast, advanced systems with closed fuel cycles are not competitive with LWRs. Therefore, there exists a strong incentive to operate actinide burners in symbioses with LWRs and to optimise the burner efficiency for a high LWR-to-burner support ratio. Proposals for implementing such LWR-burner symbioses follow two basically different approaches:

- An evolutionary approach, where the plutonium and the minor actinides in LWR-discharged fuel are separated and recycled individually in different conventional and advanced reactor types using predominantly basically proven aqueous reprocessing technology.
- An *innovative approach*, where the transuranics in LWR-discharged fuel are recovered together and transferred to a closed TRU burner fuel cycle using pyrochemical reprocessing technology which is well suited for handling the high activity of multi-recycled fuels in closed fuel cycles.

The evolutionary approach has the advantage that it can be implemented in successive steps. In particular, the first step could be the establishment of a plutonium burning strategy, and this could later be upgraded to a transmutation strategy by complementing the reactor park with dedicated minor actinide burners whose fuel cycle is optimised for this task. An early introduction of a plutonium burning strategy could be motivated by the need to reduce the stocks of separated plutonium which have accumulated in some countries due to the delay in the commercialisation of fast reactors.

Since plutonium burning represents only a small first step towards a transmutation strategy (see Figure 1.1), it is not in the focus of the present study. It may, nevertheless, be recalled that plutonium can be managed effectively with LWRs and fast reactors; innovative reactor and reprocessing technologies are not required, and respective development issues have been dealt with extensively in

13. The increased design flexibility allows, for instance, improving the "burner effectiveness" (see Chapter 2).
14. 25% is the highest fuel burn-up considered by the expert group (see comparative assessment in Chapter 3).
15. The 6 years comprise 3 years of in-pile and 3 years of out-of-pile time and are typical for an accelerator-driven TRU burner (see Chapter 3).

the framework of working parties and workshops of OECD/NEA [4,5] as well as in many international conferences. The recycling of plutonium in the form of MOX in LWRs has already reached industrial maturity. It should, however, be noticed that LWRs alone cannot burn plutonium in the longer term because the buildup of the even, non-fissile plutonium isotopes in a thermal neutron spectrum constrains the number of recycles to two or three at most; the remaining degraded plutonium has to be disposed or transferred to a fast reactor fuel cycle. From the viewpoint of transmutation, the build-up of minor actinides, especially the highly radioactive curium, is also a drawback of a "thermal" recycling strategy.

As indicated before, fast-spectrum systems are required to handle the minor actinides. Conventional fast reactors can burn self-generated minor actinides in a closed cycle, but they are not suited for burning pure minor actinides. Compared with conventional MOX-fuelled fast reactor cores, dedicated minor actinide burner cores have significant safety disadvantages due to an increased coolant void reactivity effect in liquid-metal cooled systems, a generally smaller fuel Doppler reactivity coefficient and a considerably reduced fraction of delayed neutrons, β_{eff}.[16] The coolant void reactivity effect can be mitigated by reducing the size and optimising the geometry of the core, or eliminated completely by replacing the liquid-metal by a gas coolant. The application of the ADS concept to a minor actinide burner core is an interesting possibility to compensate the safety disadvantages arising from the small Doppler coefficient and the small β_{eff} value which cannot be otherwise compensated. To cope with the high activity of the fuel, dedicated minor actinide burners require a fuel cycle with pyrochemical reprocessing. Thanks to a very favourable support ratio, however, the investments into advanced reactor and reprocessing technology remain small.

The innovative approach to LWR-burner symbioses aims at co-processing plutonium and minor actinides to avoid the use of technologies with a potentially high proliferation risk. After initial separation of the uranium from the LWR spent fuel, the actinides are recycled in a TRU burner with a closed fuel cycle using pyrochemical reprocessing without further actinide separation. The core characteristics of TRU burners are less degraded than those of minor actinide burners and allow the burner to be operated in a critical state. The ADS concept, however, offers additional design flexibility which can be utilised to increase the LWR-to-burner support ratio. Compared with the evolutionary approach, the innovative approach requires a larger investment in dedicated actinide burners.

In Chapter 3, the essential features of the different actinide transmutation approaches will be discussed and compared with the help of six *principal fuel cycle schemes*, including three transmutation schemes, an LWR once-through reference case, a plutonium burning scheme in which the fuel cycle is closed for plutonium only, and a pure fast reactor scheme. It will be shown in Chapter 2 that, for closed fuel cycles, the transmutation performance in terms of actinide waste mass and radiotoxicity reduction depends primarily on two parameters, the fuel burn-up and the fuel losses in the reprocessing which have to be maximised and minimised, respectively. Goals for waste mass and radiotoxicity reduction will be derived; and it will be shown that these goals can be met with both evolutionary and innovative transmutation approaches, but that the consequences on the fuel cycle can vary considerably between the different approaches.

With regard to consequences for the fuel cycle, it is important to note that the multi-recycling of fuels, especially in minor actinide and TRU burners, results in very high activities, decay heats and neutron source strengths of the fuels which complicate the reprocessing, handling, and shielding of the fuels. To reduce these problems, it has been proposed to restrict the multi-recycling to neptunium and to recycle americium and curium in separate targets which are disposed of after a single irradiation in

16. A sufficiently negative fuel Doppler coefficient restricts the energy release in an accidental prompt-critical power excursion and β_{eff} determines the reactivity margin within which the chain reaction can be controlled. By substituting minor actinide fuel for normal MOX fuel, β_{eff} is about halved.

a fast reactor. Advantages of such "heterogeneous" recycling concepts are that the fuel cycle – a normal MOX fast reactor fuel cycle with standard aqueous reprocessing – is much less contaminated with minor actinides, the mass flows of the latter are generally reduced, and the safety of the burner is not significantly impeded. Disadvantages of the heterogeneous concepts are the limited number of target assemblies which can be accommodated in the reactor and the limited burn-up of the targets which allows only modest waste mass reductions to be achieved.

1.3.3 Fission product transmutation

For the long-lived fission products, the goal is to transform them into shorter-lived or stable species by means of *neutron capture reactions*. Other nuclear reactions have also been considered, but, presently, do not fulfil the criteria for a practical application. The fission product transmutation involves different problems than the actinide transmutation: the fission products have to be separated individually from HLW with a high decontamination factor and processed to stable targets for irradiation, the necessity of isotopic separations and small reaction cross sections constrain the practical implementation of the processes and, not least, the lack of producing net amounts of energy jeopardises the economic viability of the processes.

The traditional fission product transmutation method is the irradiation of targets in a strong flux of neutrons produced by a fission reactor or a spallation neutron source. Neutron economy considerations favour concepts where the fission product targets are irradiated in moderated sub-assemblies of fast reactors. More recently, it has been suggested to use lead or lead-bismuth as coolant for fast-spectrum systems because the small energy loss of neutrons in collisions with the lead atoms enhances the probability for the slowing down neutrons to be captured in the resonances of the nuclide to be transmuted. Based on this principle, an accelerator-driven fission product transmuter, in which nearly every source neutron induces a transmutation reaction, is theoretically feasible.

It appears that, in principle, the transmutation of long-lived fission products would be a useful method to mitigate the long-term risk of geologic repositories; however, the practical feasibility of the required processes is less obvious than in the case of the actinides and, so far, has been established only for ^{99}Tc. This means that, for most potentially troublesome long-lived fission products, including ^{135}Cs, ^{126}Sn, ^{79}Se and possibly also ^{129}I, partitioning followed by special conditioning and confinement in a very stable matrix may remain the only realistic method for reducing their radiological impact.

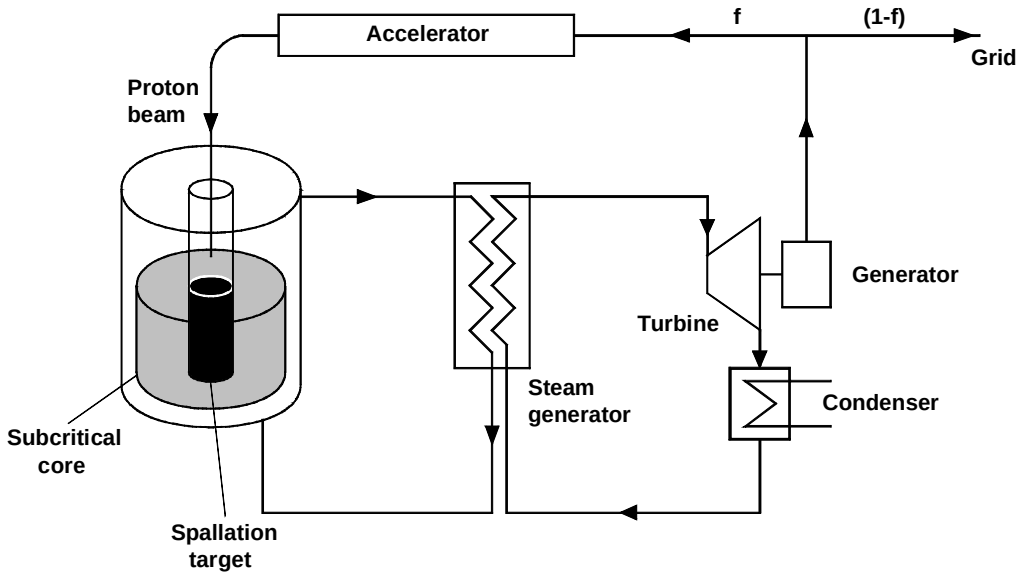
1.4 The ADS concept

The concept of accelerator-driven systems (frequently called hybrid systems) combines a particle accelerator with a sub-critical core (see Figure 1.3). Most proposals assume proton accelerators, delivering continuous-wave beams with an energy around 1 GeV. The accelerator is either a linear accelerator (linac) or a circular accelerator (cyclotron). High-power accelerators have been under continuous development, and the construction of machines with the required specifications, i.e. electrical efficiencies in the vicinity of 50% and beam powers up to 10 MW for cyclotrons and up to 100 MW for linacs, now appears to be feasible.

The protons are injected onto a spallation target to produce source neutrons for driving the sub-critical core. The target is made of heavy metal in solid or liquid state. Spallation reactions in the target emit a few tens of neutrons per incident proton, which are introduced into the sub-critical core to induce further nuclear reactions. Except for the sub-critical state, the core is very similar to that of a critical reactor. It can be designed to operate either with a thermal or fast neutron spectrum.

The energy conversion part of an accelerator-driven nuclear power system is similar to that of a normal power plant. However, in the accelerator-driven system, the electrical energy which is recycled to the accelerator reduces the net electrical efficiency of the system. For an ADS with a neutron multiplication factor of 0.95, the reduction amounts to about 12%. This means that the accelerator-driven system produces about 14% more high-level waste and rejects about 20% more heat to the atmosphere than a normal power plant with the same net electrical output.

Figure 1.3. **Concept of an accelerator-driven system**



The principal advantages and disadvantages of accelerator-driven systems as compared with the corresponding critical reactor systems are summarised in Table 1.1. The comparison applies not only to transmutation applications on which the present study is focussed, but also to other applications such as the breeding of fissile material (electro-breeding), the development of the thorium-^{233}U fuel cycle, and the development of ultra-safe energy producers. For instance, the potential for improving the neutron economy, which is related to the neutron abundance of the spallation process, is more relevant for breeding than for transmutation applications.

In the context of transmutation, the principal non safety-related advantage of the ADS is the increased core design and fuel management flexibility resulting from the removal of the criticality condition. However, this advantage has to be weighted against several technical and operational disadvantages. For example, the benefit from lengthening the reactor cycle has to be balanced against the investment in the more powerful accelerator required for coping with the lower end-of-cycle neutron multiplication factor.

Table 1.1. **Comparison of accelerator-driven sub-critical and critical reactor systems**

	Advantages of accelerator-driven systems	**Disadvantages of accelerator-driven systems**
Design and operation	♦ The possibility to operate a reactor core at a *neutron multiplication factor below 1* opens opportunities for new reactor concepts, including concepts which are otherwise ruled out by an insufficient neutron economy ♦ In particular, this allows transmuters to be designed as pure TRU or MA burners and hence the fraction of specialised transmuters in the reactor park to be minimised ♦ The proportionality of the reactor power to the accelerator current simplifies the reactor control	♦ *Accelerator:* Very high reliability required to protect structures from thermal shocks ♦ *Beam window and target* subjected to unusual stress, corrosion and irradiation conditions ♦ *Sub-critical core:* Increased power peaking effects due to external neutron source ♦ Compromises between neutron multiplication factor and accelerator power required ♦ Increased overall complexity of the plant ♦ Reduction in net plant electrical efficiency due to power consumption of accelerator
Safety	♦ The reactivity margin to prompt criticality can be increased by an extra margin which does *not depend on the delayed neutrons* ♦ This enables the safe operation of cores with degraded characteristics as they are typical e.g. for pure MA burners ♦ *Excess reactivity can be eliminated*, allowing the design of cores with a reduced potential for reactivity-induced accidents	♦ New types of reactivity and source transients have to be dealt with (external neutron source can vary rapidly and reactivity feedbacks in TRU- and MA-dominated cores are weak)

Note: Issues of particular relevance for the transmutation of TRU and minor actinides (MA) are underlined.

Important design and material problems arise from the installation of a target in the centre of a reactor: the interfacing of an accelerator with a reactor rises containment questions, and the target and surrounding structure materials are subjected to complex degradation phenomena due to combined thermo-mechanical loads, high-energy particle irradiation and, in contact with liquid heavy metals, corrosion effects which are much more severe than those encountered in normal reactors. This applies particularly to the beam window which may, therefore, require frequent replacement.

High-power accelerators will have to be improved with respect to the beam losses which cause radiation damage and activation in the accelerator components and the frequency of beam trips. In an ADS, beam trips cause similar temperature and mechanical stress transients as fast control rod insertions (scrams) in critical reactors. Current accelerators feature beam trip frequencies which lie orders of magnitude above the current criteria for such transients.

Regarding safety aspects, the prominent feature of the ADS is its reduced potential for reactivity-induced accidents. This is particularly relevant for actinide burners which suffer from a general degradation of the safety parameters of the core. From the viewpoint of transmutation, a general conclusion from Table 1.1 is that an ADS has interesting design and safety advantages, but that these must be weighted against non-trivial technical and operational disadvantages which will also have economic consequences.

The diverse technical aspects of the ADS have been studied in many OECD Member countries. However, there is still a need for assessing more thoroughly the added value the ADS in the context of complete fuel cycles.

1.5 Framework for the present study

A first series of comprehensive studies investigating the role and feasibility of partitioning and transmutation as an alternative waste management option was conducted in the 1970s predominantly in Europe. On the whole, these studies denied the existence of a cost, safety, or any other incentive for developing this technology. A renewed interest in P&T arose in the 1980s in response to an increasing public opposition against the geologic disposal of radioactive waste. More recently, the difficulties in the commercialisation of the large, sodium-cooled fast reactor and the progress in the development of high-power accelerators strengthened the interest in transmutation technologies in general and accelerator-based technologies in particular. A detailed history of this development is given in Annex C.

In response to the increased interest in P&T and ADS technologies, OECD Member countries launched several R&D programmes, such as the OMEGA programme in Japan [6] and the SPIN programme in France [7], and, more recently, developed "roadmaps" for the demonstration of these technologies as e.g. the US Roadmap for Developing Accelerator Transmutation of Waste Technology [8] and the European Roadmap for Developing Accelerator-driven Systems for Nuclear Waste Incineration [9]. The Nuclear Energy Agency of the OECD initiated a long-term programme on P&T in 1989. The respective projects address a wide range of issues ranging from basic nuclear data questions to systems studies. In parallel, an International Information Exchange Programme has been established to strengthen the international collaboration.

A first P&T systems study, conducted by OECD/NEA from 1996 to 1998 [2], focused on a review of the progress in the separation of long-lived actinides and fission products, the options for their transmutation, and the benefit for the waste management. Specific fuel cycle schemes were discussed, covering plutonium-recycling and the additional burning of minor actinides in dedicated systems. However, the study did not address the more performant transmutation strategies with fully closed fuel cycles, nor the technology of the ADS and the specific role of the latter in such closed fuel cycles.

The present, second P&T systems study is complementary to the first study. It aims at clarifying the roles and relative merits of critical and sub-critical fast-spectrum systems in closed fuel cycles with the help of a set of representative "fuel cycle schemes" and assesses the development status of the ADS with emphasis on reactor technology and safety, fuel cycle technology, cost/benefit issues, and general feasibility.

Chapter 2 of the report defines target values for the waste mass and radiotoxicity reduction to be achieved by an effective transmutation strategy, discusses the incentive for closed fuel cycles in general, including the role of fast-spectrum systems in these fuel cycles, and summarises the principal results of the comparative analysis of the fuel cycle schemes. The fuel cycle schemes, the results of the comparative analyses and the associated fuel fabrication and reprocessing issues are described in detail in Chapter 3.

The technological issues and differences between fast reactors and accelerator-driven fast reactors are covered in Chapter 4. ADS safety, with emphasis on uranium-free systems, is handled in Chapter 5; Chapter 6 is devoted to a preliminary cost analysis, and Chapter 7 overviews the perceived R&D needs. Chapter 8 focuses on the transmutation of long-lived fission products where Chapter 9 is introducing alternative actinide transmutation approaches.

2. TRANSMUTATION STRATEGIES

2.1 Introduction

The transmutation strategy as defined in Chapter 1 involves the recycling of both plutonium and minor actinides with the goal of converting all actinides to fission products. Immediate benefits of a reprocessing strategy, with or without minor actinide transmutation, are the elimination of plutonium from the HLW and a reduction in the total mass of the HLW in comparison with a (spent fuel) direct disposal strategy. The closure of the fuel cycle for plutonium reduces the natural uranium requirement by 30%, and the additional fissioning of the minor actinides reduces the natural uranium requirement by another 5%.[17] The latter is an "extra gain" from transmutation which, alone, would not justify the development of new reactor and fuel cycle technology.

From a discussion of the contribution of actinides and fission products to the radiotoxicity and long-term risk of HLW, Chapter 2 first derives target values for the reduction of the actinide waste mass and the fuel reprocessing losses which have to be set for an effective actinide transmutation strategy. A second part of the chapter deals with the implications of a fully closed fuel cycle for the overall neutron economy and transmutation performance of an actinide burner, compares different actinide transmutation strategies, and summarises the results of a consistent analysis of "principal fuel cycle schemes", carried out by the Expert Group. Finally, transient aspects in nuclear energy scenarios and the feasibility of transmuting long-lived fission products are briefly discussed.

2.2 Radiotoxicity and long-term risk of high-level waste

Figure 2.1 shows the radiotoxicity of uranium-oxide fuel with an average burn-up of 50 GWd/tHM as discharged from the reference LWR considered in the present study. In the figure, this radiotoxicity is compared with the radiotoxicity of the remaining HLW after separation of 99.9% of the uranium and plutonium, assuming a cooling time of 4 years between fuel discharge and reprocessing. A decomposition of the latter into nuclide contributions is also shown.

It can be seen that the radiotoxicity is dominated, first, by short-lived fission products, and later, by actinides. A few hundred-thousand years after the discharge, the radiotoxicity of the unprocessed fuel drops to the natural toxicity level for LWRs, i.e. the equilibrium radiotoxicity of the natural uranium required to fabricate the fuel.[18] The separation (and intermediate storage) of uranium and plutonium would reduce the radiotoxicity of the remaining HLW in the time frame from 10^3 to 10^5 years by an order of magnitude, but it would still take some twenty-thousand years for the radiotoxicity of this waste to reach the LWR natural toxicity level. Moreover, when defining a HLW radiotoxicity reduction goal for a fuel cycle strategy involving reactors with increased resource efficiency, it should be borne in mind that the natural toxicity level decreases proportionally with the

17. Evaluated for the plutonium burning and the "double strata" strategies discussed in Section 2.7.1.
18. The LWR natural toxicity level is calculated as the product of the natural uranium requirement for an LWR once-through strategy (20.5 t/TWhe for a burn-up of 50 GWd/tHM) and the radiotoxicity of natural uranium, including daughter products (20 Sv/kg).

natural uranium requirement. The figure indicates that, for a pure fast reactor strategy, the natural toxicity level corresponds about to the radiotoxicity of the long-lived fission products.

Figure 2.1. **Radiotoxicity of LWR spent fuel**

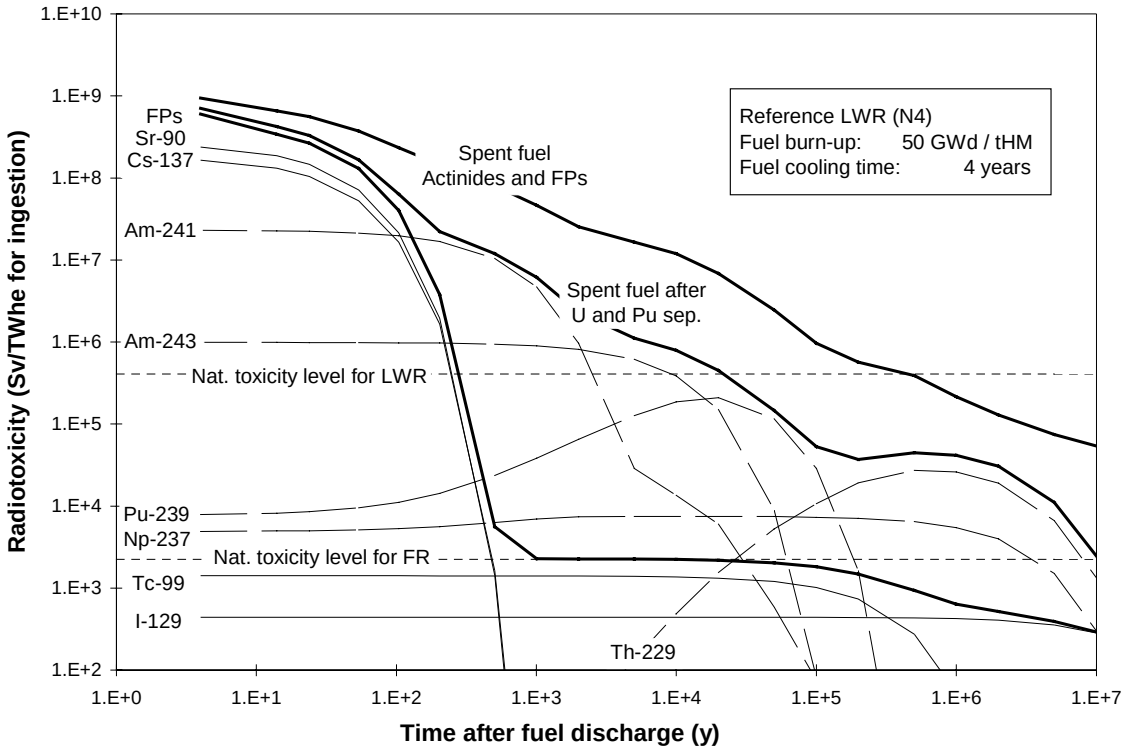


Figure 2.2 compares risks in terms of annual individual doses to the population for different concepts of geologic repositories, the four examples representing the direct storage of spent fuel in chemically reducing or oxidising environments [10,11], and the emplacement of vitrified HLW in cristalline host rock or clay [12,13]. It should be noted that the curves apply to nuclear energy scenarios with different energy production and different amounts of spent fuel. The figure shows that, with the exception of the Yucca Mountain repository, the doses to the population lie at least two orders of magnitude below the natural radiation exposure.

Conclusions of direct relevance for the present study are:

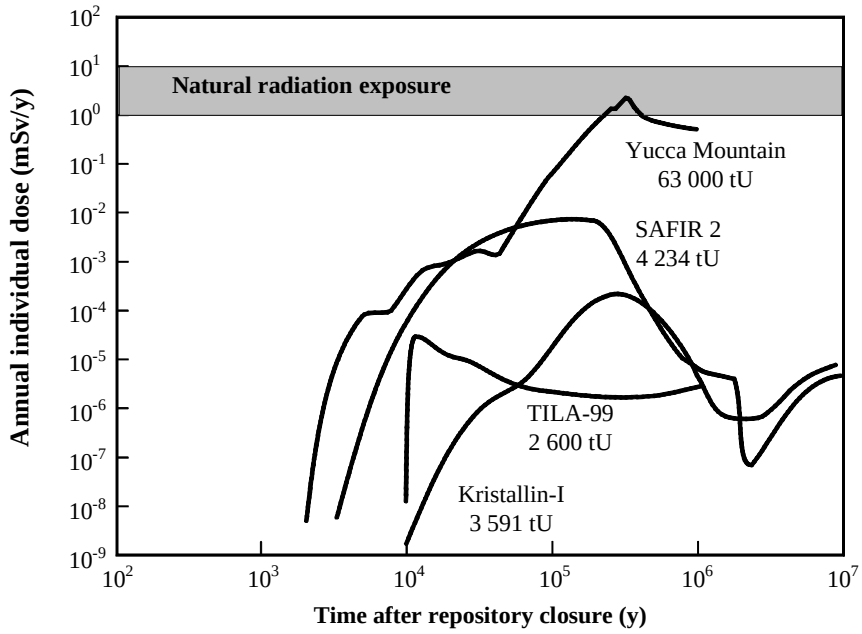
- From the viewpoint of the *radiotoxicity*, which plays a role mainly in accidental intrusion scenarios, P&T must first be concerned with the actinides, particularly the minor actinides americium and neptunium,[19] the toxicity of the fission products lying at least two orders of magnitude below that of the actinides after a few hundred years.
- The *long-term risk* of a geologic repository is usually dominated by fission products which are generally more mobile than actinides. Dose contributions arise primarily from ^{129}I, ^{135}Cs,

19. Two other heavy nuclides appearing in Figure 2.1, ^{239}Pu and ^{229}Th, are decay products of ^{243}Am and ^{237}Np, respectively. In the time frame of interest to P&T, curium is not a dominant contributor to the waste radiotoxicity.

^{99}Tc, ^{126}Sn and ^{79}Se, their order of importance depending on the repository concept.[20] The fission product risk peaks in the time range 10^4 to 10^6 years after the closure of a repository, whereas the (smaller) actinide risk arises "only" after one million years).[21]

- Actinide transmutation strategies address primarily the radiotoxicity (hazard) of the HLW. To reduce the long-term risk (dose to the population), the long-lived fission products would also have to be transmuted or specially conditioned.

Figure 2.2. **Annual individual dose for different repository concepts**



Dominant nuclides:

TILA-99 spent UOX, reducing env.	^{129}I / ^{126}Sn / ^{129}I / ^{229}Th
Yucca Mountain spent UOX, oxidising env.	^{99}Tc / ^{237}Np (^{229}Th)
Kristallin-I vitrified HLW in granite	^{79}Se / ^{135}Cs / ^{99}Tc / ^{231}Pa
SAFIR 2, provisional vitrified HLW in clay	^{79}Se / ^{129}I / ^{229}Th

2.3 Goals for actinide mass reduction and fuel losses

2.3.1 Actinide mass reduction

As discussed in Section 2.2, the radiotoxicity of LWR-discharged fuel requires a few hundred-thousand years to decay to the LWR natural toxicity level. With a hundred-fold reduction in the

20. Vitrified HLW does not contain a significant amount of ^{129}I because iodine is released during reprocessing and (currently) discharged to the sea. Since sea disposal may no longer be practicable for advanced nuclear fuel cycles, ^{129}I is also a candidate for P&T in vitrification scenarios.
21. For the Yucca Mountain repository, the actinide risk becomes dominant after 50 000 years (see Figure 2.2).

actinide content of the HLW, this goal could be reached after about thousand years, i.e. within the time span during which the HLW container can be expected to fulfil its safety function. For transmutation strategies involving fast reactors, an even higher actinide reduction factor would be desirable. For such strategies, a reduction in the actinide content of the HLW by a *factor of 100* must therefore be set as a minimum goal.

It is obvious that a hundred-fold reduction of the actinide mass cannot be achieved in a single pass of the fuel through a reactor. Hence, *multi-recycling* of the fuel will be essential.[22] In fact, the ideal P&T system has a fuel cycle which is fully closed for the actinides, meaning that only fission products are separated from the spent fuel and all actinides are returned to the reactor, together with a "top-up" of new fuel replacing the fuel which was fissioned. It is also clear that such a system must be operated for many decades before the composition of the discharged fuel, which determines the specific waste radiotoxicity, reaches an equilibrium.

2.3.2 Fuel losses in the reprocessing

In practice, the actinides cannot be recovered completely from the spent fuel, and the remainder will go to waste. For the fully closed system illustrated in Figure 2.3, the mass of actinides going to waste is:

$$M^W = \delta \; L \; M^F,$$

where M^F is the total mass of actinides fissioned, L is the actinide loss fraction during reprocessing and fuel fabrication, and the burn-up factor, δ, can be evaluated from the fraction of heavy metal fissioned, B, as (1 - B)/B. For equilibrium conditions and small actinide losses, M^F equals the top-up fuel mass, M^T, which, in general, can be divided into the mass, M^B, of transuranic or minor actinides to be burnt, and a diluent, usually consisting of uranium (see Figure 2.3, TRU/MA and diluent supply). The diluent allows to optimise the core characteristics of the actinide burner (see Section 2.3.4).

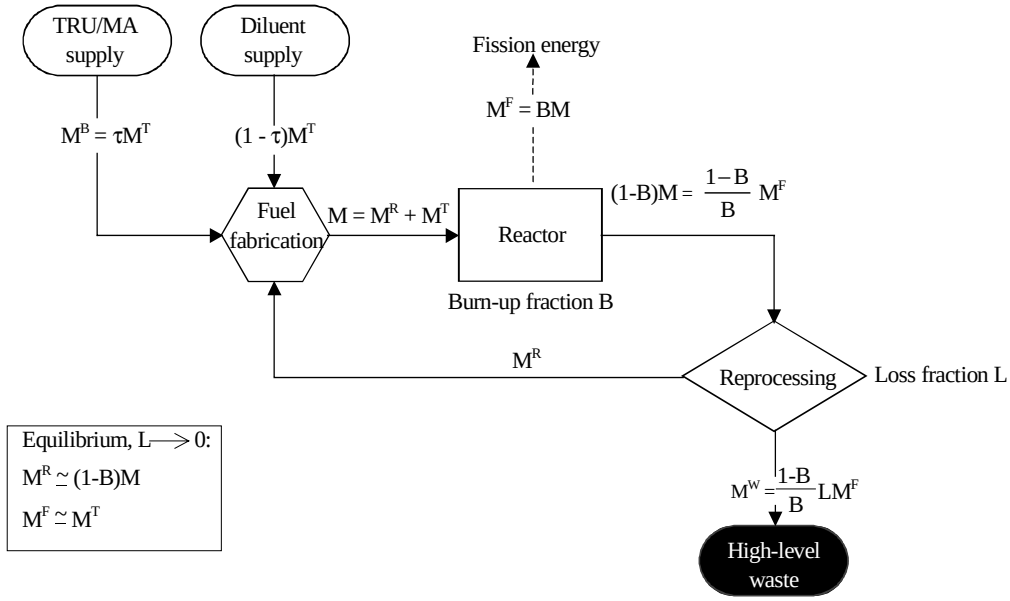
Denoting the transuranic or minor actinide fraction of the top-up fuel, M^B/M^T, by τ and the "waste mass reduction factor", M^B/M^W, by R^M, one obtains the simple expression

$$L = \tau/(\delta \; R^M),$$

which gives the allowable losses as a function of the waste mass reduction factor. For the desired reduction factor of 100, an achievable average fuel burn-up of 15%,[23] and a top-up fuel without a diluent ($\tau = 1$), the expression yields L = 0.18%. Since average burn-ups beyond 15% have not yet been proven with known fuel technologies, a *target value of 99.9%* for the actinide recovery yield must consequently be set for an effective actinide transmutation system.

22. The highest average fuel burn-up for systems with solid fuels considered by the Expert Group is 25% (ATW project goal). The burn-up can be higher, if the actinides are recycled heterogeneously in separate target pins (see Chapter 3, Section 3.3.2) or if alternative fuels, consisting e.g. of coated particles, are introduced (see Chapter 9), but would still be far from the goal of 99%.
23. In agreement with the reference burn-up of 140 GWd/tHM for TRU and MA burners (see Table 2.3).

Figure 2.3. **Actinide mass flow in fully closed fuel cycle in equilibrium**



2.4 Reactor requirements in fully closed fuel cycles

2.4.1 Neutron balance of equilibrium core

For neutronic reasons, not all reactors can operate with a fully closed fuel cycle. To assess the suitability of an equilibrium core in terms of neutron multiplication,[24] the production-to-absorption ratio of the actinides in the equilibrium core, η_{ec}, is a useful parameter. Alternatively, the overall neutron balance for the complete fissioning of an actinide or an actinide mixture can be measured in terms of the neutron excess parameter -D (see Chapter 1, Section 1.3.2, and [14]). An η_{ec} value smaller than 1 means that the equilibrium core cannot maintain a chain reaction; a negative -D value indicates that an actinide or an actinide mixture cannot be completely transmuted. The parameters are mainly influenced by the top-up fuel composition, the neutron spectrum, and the flux level. It can be shown that both approaches lead to the same conclusions.

The η_{ec} and -D values in Table 2.1 refer to different ADS concepts which are designed to burn pure transuranics or minor actinides as well as an ALMR-type fast reactor which is fed with pure uranium. The fast-spectrum systems are those described in Table 2.3, the thermal ADS is the graphite-moderated molten-salt system proposed in [15]. Different top-up fuels are considered: the plutonium and transuranic mixtures correspond to PWR spent fuel with a burn-up of 50 GWd/tHM; the MA mixture is that produced by the first stratum of the "double strata strategy" described in Section 2.7.1.

It can be seen that minor actinides cannot be completely transmuted in thermal systems and that fast systems offer more excess neutrons than thermal systems. Interestingly, the accelerator-driven fast systems have smaller neutron excesses than the critical fast reactor. This is due to the moderation effect of the high zirconium content of the uranium-free fuels on the neutron spectrum of the fast

24. The equilibrium core is the core which is established asymptotically by recycling the discharged fuel indefinitely in the same reactor as illustrated in Figure 2.3. Its neutronic parameters can differ considerably from those of a start-up core.

burners. Large neutron excesses are advantageous, if the systems are also utilised as fission product transmuters (see Chapter 8, Section 8.2).

Table 2.1. **Overall neutron balance parameters of different equilibrium cores**

Top-up fuel	Thermal TRU burner (ADS)		Fast TRU burner (ADS)		MA burner (ADS)		Critical fast reactor	
	η_{ec}	-D	η_{ec}	-D	η_{ec}	-D	η_{ec}	-D
Uranium-238	0.92	-0.24	1.28	0.64	1.28	0.64	<u>1.41</u>	<u>0.85</u>
Plutonium	1.15	0.40	1.80	1.34	1.74	1.28	2.03	1.53
Minor actinides	0.89	-0.37	1.37	0.86	<u>1.33</u>	<u>0.79</u>	1.52	1.10
Transuranics	<u>1.11</u>	<u>0.30</u>	<u>1.75</u>	<u>1.29</u>	1.69	1.23	1.96	1.48

Notes:
1. Underlined values indicate that the concept has been optimised for the indicated top-up fuel.
2. A PWR loaded with 30% MOX has a similar neutron economy as the thermal ADS.

2.4.2 Core design constraints

In practice, the design of a TRU or MA burner core, like that of any reactor core, is also constrained by performance and safety parameters, such as the reactivity swing during burn-up, the coolant void reactivity effect, the fuel Doppler coefficient, and the effective delayed-neutron fraction. Unfortunately, for a sodium-cooled fast reactor, the substitution of normal MOX fuel by TRU- or MA-dominated fuel has an unfavourable influence on several of these parameters. This shortcoming of the conventional fast reactor has led to a renewed interest not only in the ADS, but also in various alternative fast and thermal reactor concepts which had been studied in the past, but have not been developed to commercial systems.[25]

To ensure that a critical burner core performs satisfactorily and has acceptable safety parameters, it is usually necessary to blend the TRU or minor actinides with the fertile materials uranium or thorium. However, blending reduces the transmutation effectiveness of the system. Accelerator-driven systems do not require blending and offer the possibility to increase the safety margin to prompt criticality. The latter feature is particularly important for MA burners, which are difficult to control as critical systems because the effective delayed-neutron fraction is only about half of that of a normal fast reactor.

In response to the new core design issues raised by actinide burners and the increased interest in advanced reactor technology in general, government and industry funded design teams in many countries with nuclear programmes are currently spending a considerable effort on the optimisation of a broad range of advanced reactor designs featuring both critical and accelerator-driven cores.

25. For example, the (positive) coolant void effect in sodium-cooled fast reactors could be mitigated by substituting the sodium by lead, or even eliminated by substituting the liquid metal by a gas coolant.

2.5 Transmutation performance in fully closed fuel cycles

2.5.1 Transmutation effectiveness

Various definitions for the transmutation effectiveness of actinide burner cores, usually based on the minor actinide balance of the core, are given in the literature [2]. However, since these definitions do not account for the recycling of the fuel, they do not give meaningful results for an equilibrium core. For measuring the overall transmutation effectiveness of a system with a fully closed fuel cycle, the most appropriate parameter is the "burner effectiveness", defined as the relative content of the top-up fuel in transuranic and minor actinides, M^B/M^T, i.e. the already discussed parameter τ. This parameter is directly related to the supplier-to-burner support ratio which indicates how many supplier reactors (usually LWRs) can be supported by a burner reactor.

It is important to note that the thus defined burner effectiveness does not depend directly on the choice of the neutron spectrum, the fuel type and the coolant, but is governed by the above-mentioned performance and safety constraints of the core. For a critical TRU burner based on liquid metal technology, τ is smaller than about 0.5, and in the case of homogeneous MA recycling in a EFR-type fast reactor τ is less than 0.1. The possibility to operate sub-critical MA and TRU burner cores with a uranium-free top-up fuel and hence 100% burner effectiveness, i.e. $\tau = 1$, is probably the *most important advantage of accelerator-driven systems*; it allows to maximise the supplier-to-burner support ratio and the waste mass reduction factor.[26]

2.5.2 Radiotoxicity reduction

The actinide waste radiotoxicity reduction, i.e. the radiotoxicity of the top-up fuel divided by the radiotoxicity of the actinide losses in the fuel cycle, can be separated into a constant mass reduction factor and a time-dependent neutronic transmutation factor, $R^N(t)$, sometimes called "neutronic toxicity reduction" [16]. The latter depends on the characteristics of the core and the composition of the top-up fuel. Using the same notation as before, the radiotoxicity reduction relative to the top-up fuel, $R^T(t)$, is:

$$R^T(t) = R^N(t)\ M^T/M^W$$

or, in terms of the fuel burn-up and the fuel loss,

$$R^T(t) = R^N(t)/(\delta\ L)$$

For the TRU and MA burners studied by the Expert Group, the factor $R^N(t)$ assumes values between 0.7 and 2.4, meaning that the neutronic contribution to the radiotoxicity reduction is small compared with the goal for the total toxicity reduction by a factor of 100. The analysis in [17] shows that the addition of uranium to a fertile-free top-up fuel has a small positive effect on $R^N(t)$, i.e. increases $R^N(t)$ by a factor of about two. This applies to both critical and sub-critical cores.

Important conclusions to be drawn from the discussion in Section 2.5 are that:

- Regarding the neutronic transmutation factor, no single actinide burner design has a significant advantage over other designs and this factor is close to 1.
- The ADS has the advantage that it can burn pure transuranics and minor actinides and thus support a large number of supplier reactors.

26. Since the waste mass reduction is proportional to τ/L, an actinide burner with a higher burner effectiveness allows the same waste mass reduction goal to be achieved with higher fuel losses (see Section 2.3.2).

- Radiotoxicity reduction has to be achieved primarily by an *actinide mass reduction* which implies the maximisation of the fuel burn-up and the minimisation of the reprocessing and fuel fabrication losses.

The importance of advanced reprocessing and fuel technologies for P&T is thus confirmed.

2.6 Actinide transmutation strategies

From the neutron economy considerations in Section 2.4.1 it follows that the complete closure of the fuel cycle of a fission-based nuclear energy system is eased by integrating (critical or sub-critical) fast reactors into the system. From the viewpoint of both the resource utilisation and the actinide waste production, the ideal system is a single component system based only on fast reactors with a fully closed fuel cycle. It is well known that such a system has the potential of fissioning the natural uranium and thorium resources with a close to 100% efficiency while producing only a very small amount of actinide waste.[27]

In practice, the rapid substitution of the existing LWRs by fast reactors would require considerable technical and economic investments which are currently not justified because the long-term future of nuclear energy is still unclear and the operation of the current LWRs is not constrained by a uranium shortage. This has led to the development of various multi-component approaches to actinide transmutation which take account of different regional boundary conditions as well as political factors. A common feature of the currently discussed approaches is the incorporation of a relatively high fraction of conventional LWRs. Table 2.2 provides an overview of the principal approaches and indicates respective driving forces. In view of the historic development, the table distinguishes between evolutionary and innovative approaches.

The *evolutionary approach*, adopted mainly in Europe and Japan, aims at closing the fuel cycle in successive steps, starting with the recycling of plutonium in LWRs and later in fast reactors using conventional reprocessing and MOX fuel technology, and finally eliminating the minor actinides partially or completely by either burning them in a dedicated fast-spectrum burner with a fully closed fuel cycle operating in the second stratum of a double strata fuel cycle [18], or recycling them heterogeneously as targets in conventional reactors.[28] The evolutionary approach has the advantage that it can respond flexibly to changes in the priorities for plutonium and minor actinide management, and that new technologies have to be developed only for a comparatively small number of minor actinide burners which support a large park of conventional LWRs and fast reactors.

The *innovative approach*, first suggested in the USA, aims at co-processing plutonium and minor actinides to avoid the use of technologies with a potentially high proliferation risk. After initial separation of the uranium from the LWR spent fuel, the actinides are recycled in a transuranic burner with a closed fuel cycle using pyrochemical reprocessing without further actinide separation. For a TRU burning strategy, the number of burners is four to six times larger than the number of minor actinide burners in an equivalent double strata strategy, but the burners are not subjected to a (fast) neutron-spectrum condition. Nevertheless, most of the currently evaluated critical and sub-critical transuranic burners feature a fast neutron spectrum. Notable exceptions are, for instance, the AMSTER [19] and the thermal ATW concepts [15].

27. The actinide waste mass is equal to the uranium or thorium mass that was not fissioned.
28. For the heterogeneous recycling of minor actinides, which is technologically more conventional but less effective in reducing the radiotoxicity, see Chapter 3, Section 3.3.2.

Table 2.2. **Principal actinide transmutation strategies**

Innovative: co-processing of Pu and MA	**Evolutionary: Pu and MA handled separately**
Principal driving force: non-proliferation *TRU burnt in fully closed fuel cycle:* • New fuel technology required (metal fuel, molten salts, etc.) • Dry reprocessing particularly suited for closed fuel cycles and very active fuels • Technology for uranium-free fuels not yet demonstrated *Different TRU burner options:* • <u>TRU burning in FR</u> Requires fuel with a fertile component, limiting the LWR-to-FR support ratio to about 2 (transition to pure IFR strategy possible) • <u>TRU burning in fast-spectrum ADS</u> Possibility to utilise uranium-free fuel and hence increase the LWR-to-ADS support ratio to about 3 (transition to pure ADS, i.e. Energy Amplifier, strategy possible) • TRU burning in thermal reactor Requires fuel with a fertile component (e.g. AMSTER molten-salt reactor) • TRU burning in thermal-spectrum ADS Possibility to utilise uranium-free fuel and hence achieve a very low HM inventory (e.g. thermal ATW concept)	Principal driving forces: Plutonium utilisation and waste management *Plutonium burnt in semi-closed fuel cycle:* • After two to three "thermal" recycles, plutonium must be transferred to a fast reactor fuel cycle • MOX fast reactor requires fuel with a fertile component, limiting the LWR-to-FR support ratio to about 4 • Plutonium burning does not require an ADS • Transition to pure FR-MOX system possible • Existing MOX fuel technology and PUREX-type reprocessing appropriate *MAs burnt in dedicated fully closed fuel cycle:* • <u>Double strata strategy</u>, requires new reactor type with a fast neutron spectrum • ADS can utilise pure MA fuel and hence support about 15 conventional reactors • ADS has safety advantages • New fuel technology and dry reprocessing for very active fuels has to be developed *Alternative MA handling options:* • Heterogeneous recycling of Am and Cm in targets (Np can be burnt together with Pu) • MA immobilisation in a very stable matrix

Notes:
1. Transmutation strategies involving critical and sub-critical fast-spectrum systems, on which the present study is focussed, are underlined.
2. Some alternative or "mixed" transmutation strategies which cannot be assigned to a single category of the table are described in Chapter 9, Section 9.3.

2.7 Comparison of nuclear fuel cycle schemes

2.7.1 Characteristics of the schemes

Six "principal fuel cycle schemes", which are representative for and encompass most of the currently proposed schemes, have been selected by the Expert Group and compared with respect to different sustainability parameters.

The schemes represent:

1) The *LWR once-through fuel cycle* with direct disposal of the spent fuel (reference case).

2) *Plutonium burning in LWRs and fast reactors*, where the fast reactor is optimised for a high plutonium consumption to minimise the number of fast reactors in the reactor park.

3a) *TRU burning in ALMR-type critical fast reactors* optimised for a low conversion ratio (CR = 0.5).

3b) *TRU burning in ATW-type sub-critical fast reactors* (Same strategy as 3a, but substituting the FR by an ADS to increase the fraction of LWRs in the reactor park).

4) The *double strata strategy* with LWRs and fast reactors in the first stratum as in scheme 2 and accelerator-driven dedicated MA burners in the second (P&T) stratum.

5) The *pure fast reactor strategy* based on the IFR concept where the fuel cycle is closed for all actinides, representing the long-term goal for the nuclear development.

The schemes are illustrated in Figure 2.4, and information on the assumed reactor and fuel cycle characteristics is given in Table 2.3. The plutonium burning scheme, scheme 2, is not a transmutation scheme. It is included in the analyses because, in combination with the double strata scheme, it allows to assess the "extra gains" from burning the minor actinides. LWR-MOX reactors are incorporated in this scheme, because it is representative for the evolutionary path and MOX recycling in LWRs is already a standard practice. The TRU burning schemes, 3a and 3b, implement a pure co-processing strategy. For the fast-spectrum systems in schemes 3a, 3b and 5, IFR-type fuel cycles [3] are appropriate. A lead-bismuth cooled ADS with nitride fuel as proposed by Japan [20] is chosen for the MA burner in scheme 4. European alternatives for accelerator-driven MA burners described in [21] can be expected to have a similar transmutation performance.

To ensure direct comparability of the results, the six principal fuel cycle schemes were analysed using a single nuclear data library, a single reactor code system, and consistent input data for reactor and fuel cycle parameters based on recommendations of the Expert Group. Two additional schemes were analysed independently, but using compatible reactor and fuel cycle assumptions. The additional schemes represent:

3c) *TRU burning in ADS with preceding MOX recycling* (MOX-TRU burning) to maximise the fraction of LWRs in the reactor park.

H2) *Heterogeneous recycling of americium and curium* in special "target" pins which are disposed of after irradiation in special fast reactor subassemblies (This is a technologically more conventional, but less effective transmutation method which does not depend on an ADS and is being explored as a near term option).

Figure 2.5 shows the percentage contributions of the reactor components to the total electricity production (usually called electrical support ratio) as derived from the calculated mass flows for the different schemes. It is interesting to note that, among the transmutation schemes, the MOX-TRU burning scheme and the double strata scheme feature maximum electricity productions in LWRs, and in conventional reactors (LWR and FR-MOX), respectively. The heterogeneous recycling scheme is unique in that it produces less electricity in LWRs than any of the other schemes.

Since nuclear energy scenarios for the medium- and long-term future differ considerably between countries and in many countries are uncertain, the analyses were performed for steady-state conditions, i.e. for the operation of a reactor park at a constant power level over many reactor generations. However, this approach is quite adequate for discussing the principal differences between the strategies. Some general remarks concerning the start-up and shut-down phases of nuclear energy scenarios are made in Section 2.8.

Figure 2.4. **Overview of principal fuel cycle schemes**

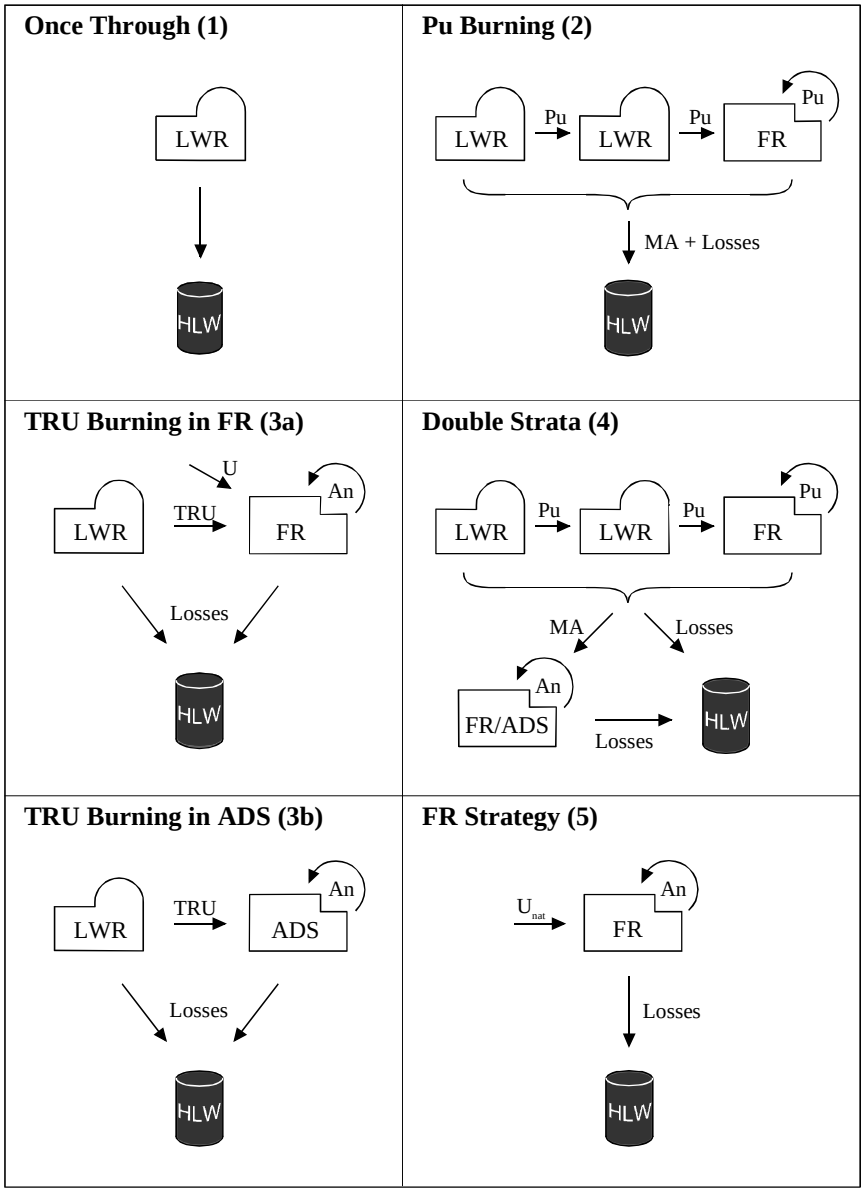
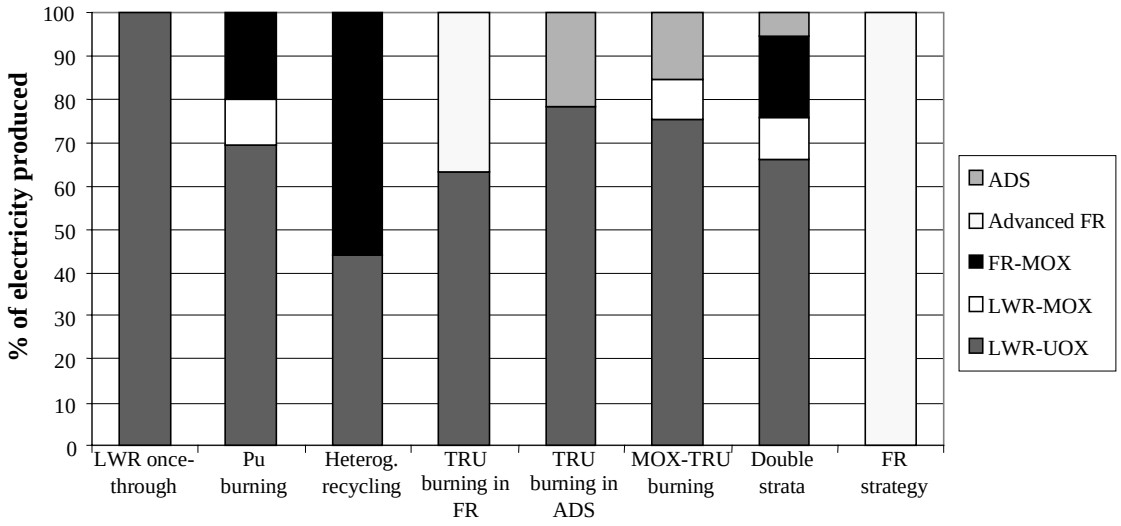


Table 2.3. **Reactor and fuel cycle characteristics**

Schemes	Reactor/ADS	Fuel	Av. Burn-up[1] (GWd/tHM)	Storage/ Cooling[2] (y)	Reprocessing method[3]
1 to 4	LWR[4]	UOX	50	2/4	wet
2, 4	LWR[4]	MOX	50	2/7	wet
2, 4	Pu burner (FR)[5]	MOX	185	2/7	wet
3a	TRU burner (FR)[6]	Ac-Zr	140 (139)	1/2	dry
3b	TRU burner (ADS)[7]	Ac-Zr	140 (250)	1/2	dry
4	MA burner (ADS)[8]	AcN-ZrN	140 (149)	1/2	dry
5	Fast reactor	Ac-Zr	140 (127)	1/2	dry

1. The burn-up for the reactors with dry reprocessing is a reference burn-up for the radiotoxicity reduction comparison. The reactor core analysis is performed for the project-specific burn-up given in brackets.
2. Fuel storage time after fabrication / fuel cooling time before reprocessing.
3. A nuclide-independent recovery yield of 99.9% is assumed.
4. PWR (French N4 reactor).
5. CAPRA design with high-burn-up core.
6. ALMR burner core with a conversion ratio of 0.5.
7. ATW, lead-bismuth cooled core as proposed by ANL.
8. Design according to [20] except for core radius (92 cm instead of 120 cm).

Figure 2.5. **Electricity contributions of reactor components for different schemes**



2.7.2 Resource efficiency and environmental friendliness

As mentioned in Chapter 1, resource efficiency and environmental friendliness are principal axes along the path towards a more sustainable nuclear energy system, together with cost effectiveness and proliferation resistance. The resource efficiency is usually measured in terms of the natural uranium requirement of a fuel cycle strategy; it is currently not a limiting factor, since uranium resources are still plentiful. Important parameters along the environmental friendliness axis are the actinide waste production with emphasis on the transuranic elements, the waste radiotoxicity, the repository requirements and, last but not least, the production of depleted and irradiated uranium.

Figures 2.6 and 2.7 compare the performance of the fuel cycle schemes, relative to the once-through fuel cycle, with regard to natural uranium requirement, TRU and heavy metal losses to repository, and actinide waste radiotoxicity. Important observations are:

- All strategies including LWRs in the reactor park require similar uranium resources. Compared with the once-through strategy, the reductions in the natural uranium requirement are in the range 20 to 37%. Regarding this parameter, only the pure fast reactor strategy achieves a break-through (180-fold reduction when the fuel cycle is fully closed).
- All transmutation strategies with closed fuel cycles have similar TRU-, HM- and radiotoxicity-reduction potentials and these are comparable with those of the pure fast reactor strategy. Under the assumptions made in the analysis (e.g. recovery of 99.9% of all actinides), the mass reduction factors exceed 170 for the transuranics and 1 100 for the heavy metal, and the goal of a hundred-fold radiotoxicity reduction is comfortably met. In particular, this means that the FR and the ADS have similar reduction potentials with respect to these parameters.
- Multiple recycling of plutonium without minor actinide transmutation is useful for the management of plutonium. It effectively reduces the heavy metal losses to the repository and reduces the natural uranium requirement by 30%. However, plutonium burning alone cannot qualify as a transmutation strategy because it reduces the radiotoxicity of the HLW by only a factor of about five.

With regard to the repository requirements for vitrified HLW, it should be noted that the total mass and the initial heat production of the vitrified waste are dominated by the fission products and, hence, mainly depend on the total thermal energy produced.[29] Moreover, due to the non-linear relationship between the actinide content of the waste matrix and the repository release rates, actinide mass reductions achieved by transmutation do not necessarily translate into proportional actinide risk reductions.

To illustrate the non-linear behaviour of the actinide releases, maximum repository near-field release rates for the plutonium burning and the double strata schemes, i.e. the schemes with HLW vitrification, were compared with those of a conventional (LWR) fuel cycle strategy with uranium and plutonium recovery and HLW vitrification (see Chapter 3, Section 3.2.5). It was found that, relative to the latter strategy, plutonium burning generally increases the maximum actinide release rates from the repository near-field, and the additional minor actinide burning reduces the maximum near-field release rates for the more important, but not for all potentially troublesome nuclides. For example, the release of ^{237}Np from the waste matrix is nearly constant because it is solubility limited, whereas the release of the daughter nuclide ^{229}Th, which is about four times more radiotoxic, reduces nearly proportionally when the ^{237}Np content of the glass is reduced.

In the conventional LWR fuel cycle, the radiological hazard of the depleted uranium arising from the enrichment process and the recovered irradiated uranium (irradiated enriched uranium from LWR-UOX fuel and irradiated depleted uranium from LWR-MOX fuel) is only of secondary concern. However, in P&T scenarios with fully closed fuel cycles, the long-term radiotoxicity of this residual uranium becomes comparable with the HLW radiotoxicity.

29. A typical canister with HLW from the reprocessing of LWR-UOX fuel contains 320 kg of glass, 48 kg of fission products, 3.5 kg of actinides and 4.5 kg of activation products [22].

Figure 2.6. **Resource efficiency and HLW production relative to open fuel cycle**

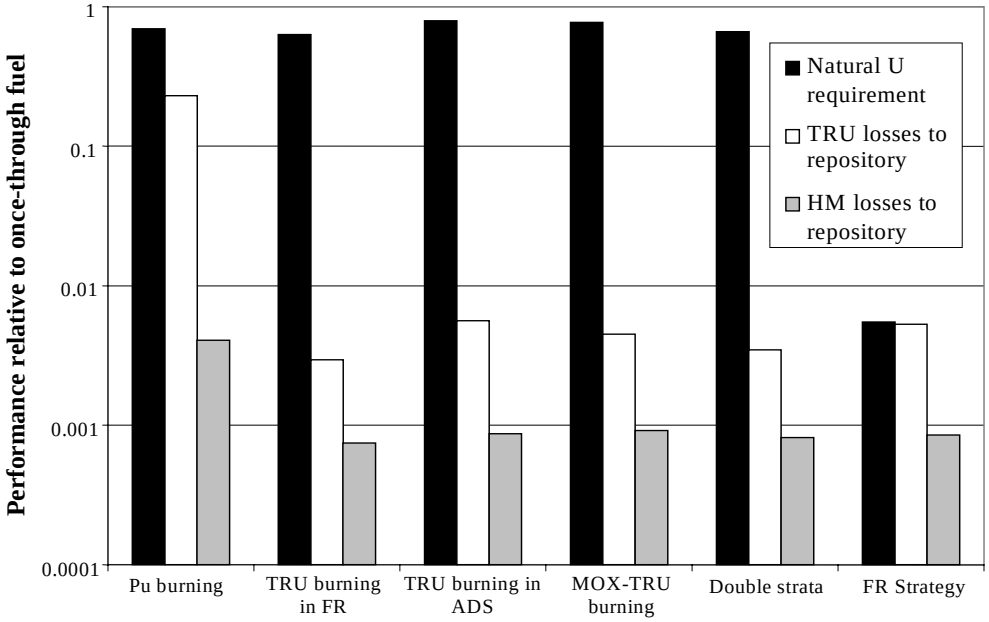


Figure 2.7. **Actinide waste radiotoxicity reduction relative to open fuel cycle**

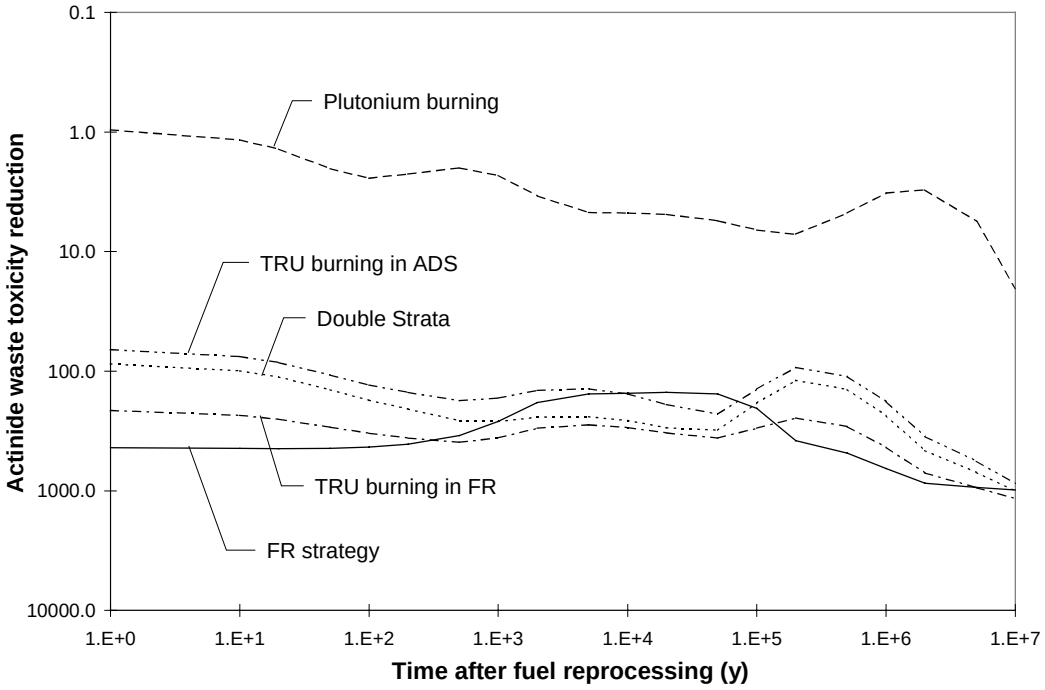
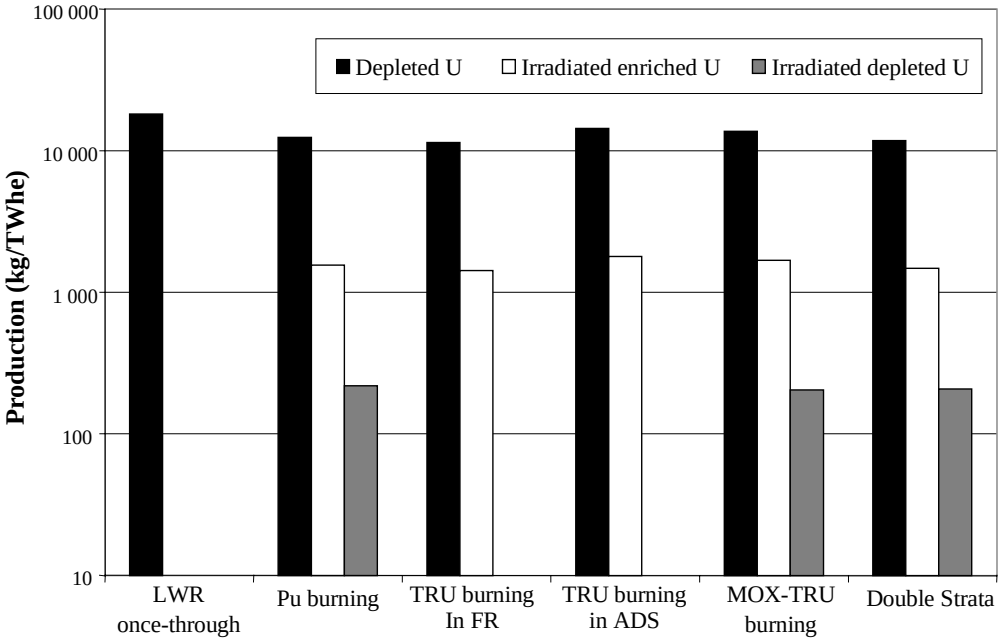


Figure 2.8 shows that all strategies including LWRs in the reactor park produce similar quantities of depleted and irradiated uranium. In the most favourable case (TRU burning in FR), the depleted uranium production relative to the once-through reference case reduces only by 28%. Compared with

the plutonium burning and the other transmutation strategies, the pure TRU burning strategies have the advantage of avoiding the production of additional irradiated depleted uranium streams.[30] The fast reactor strategy is unique in that it does not produce any residual uranium at all.

If the residual uranium is not considered as a resource for future fast reactors, its long-term radiological impact in a P&T scenario must also be assessed. In the nuclear waste discussion, not much attention has yet been given to this issue, since the management and future use of the residual uranium is a political issue.

Figure 2.8. **Residual uranium production**



Notes:
1. In the LWR once-through strategy, the irradiated uranium goes to the repository with the spent fuel.
2. The fast reactor strategy is not included in the figure; it produces no residual uranium.

2.7.3 Consequences for the fuel cycle

Whereas all transmutation strategies with fully closed fuel cycles perform similarly with respect to the resource efficiency and environmental friendliness parameters, different requirements and consequences arise for the establishment of the fuel cycle. In this context, important parameters are the TRU inventory of the fuel cycle and the decay heat and neutron source strength of the fuel.

The analyses show that, among the different transmutation strategies, TRU burning in ADS is associated with the lowest TRU inventory and hence the lowest α activity inventory. This means that this strategy can respond flexibly to unexpected changes in the nuclear energy scenario and has safety advantages.

Table 2.4 shows that the recycling of plutonium and minor actinides in equilibrium scenarios implies the handling of fuels with very high decay heat and neutron source strength levels which are beyond the capability of currently operating fuel cycle facilities. Experience with pilot plants in the

30. Irradiated depleted uranium is more radioactive than depleted uranium and unattractive for re-utilisation.

UK and France shows that the PUREX-type aqueous reprocessing ("wet" reprocessing) can be considered as valid for MOX fuels with high plutonium content such as the plutonium burner fuel arising in schemes 2 and 4. Aqueous reprocessing of this fuel within short cooling times and with the required high recovery yield of 99.9%, however, will require measures to improve the plutonium dissolution yield and modifications of the PUREX flowsheet.

Table 2.4. **Decay heat and neutron source strength**

Reactor/ADS	Fuel at fabrication time		Fuel after cooling time[1]	
	Decay heat (W/kgHM)	Neutron source (10^6 n/s-kgHM)	Decay heat (W/kgHM)	Neutron source (10^6 n/s·kgHM)
LWR-UOX	$1.1 \cdot 10^{-5}$	$2.1 \cdot 10^{-5}$	3.48	0.97
LWR-MOX	1.94	0.10	6.31	10.9
Pu burner (FR)	9.64	0.66	21.8	39.3
TRU burner (FR)	33.8	92.1	46.0	86.1
TRU burner (ADS)	168	670	193	649
MA burner (ADS)	489	1 992	455	1 812
Fast reactor	5.79	9.76	18.6	9.76

1. Fuel burn-up and cooling time see Table 2.3.

On the other hand, the decay heat of the ADS fuels arising in schemes 3b and 4 is well beyond the limit for which the radiation stability of the organic extractant in the aqueous process can be guaranteed. For these and all other systems with fully closed fuel cycles, the less developed pyrochemical reprocessing ("dry" reprocessing) is the appropriate reprocessing method because it circumvents unnecessary separation processes (only fission products are extracted) and can handle highly active product streams without major radiation degradation.

Due to the strong source of spontaneous neutrons, the fuel fabrication will have to adapt its handling technology to reduce the radiation doses to the workers in the plant and during the transport of the fuel assemblies. This also speaks for the pyrochemical reprocessing method which is applicable in small facilities in the immediate vicinity of the reactors, whereas the aqueous process favours large facilities which operate on continental or even world scale, requiring the shipment of fuel over long distances.

2.8 Transient phases in nuclear energy scenarios

2.8.1 Time constants in transient scenarios

Nuclear energy scenarios can be divided into a start-up, an equilibrium, and a shut-down phase. The discussion of the fuel cycle schemes in the preceding section was restricted to the equilibrium phase with the tacit assumption that the latter lasts much longer than the other phases.

In this context, an important aspect of nuclear fission energy are the long time constants for the penetration and phase-out of new reactor and fuel cycle technology. The long time constants reflect fundamental physical limitations in the production and destruction of in-pile and out-of-pile fuel inventories and are of the order of 50 years. This implies that transmutation technology can fulfil its promises only, if it is introduced with the intention to utilise it for at least a century. In view of the required expensive R&D, including the construction and operation of demonstration facilities, economic reasons call for a similar time horizon. More generally speaking: the long time constants inherent in advanced fuel cycle strategies and the necessity of avoiding an interruption of the chosen

strategy require a continuous political, economical and strategic support for the nuclear option to be assured over a period of *at least hundred years*.

It is likely that transmutation technology will penetrate the market only, if the nuclear contribution to the word energy demand has to be stepped up considerably for ecologic reasons. Under this assumption, the start-up phase will also be a growth phase. As was shown by many studies in the past, fast breeder reactors with an appropriately designed core can reach a doubling time in the order of 50 years which allows to support an annual growth rate of 1.4%. If necessary, an ADS could reach a *shorter doubling time* than a normal fast breeder reactor due to the lower fissile inventory and the richness of the spallation process in neutrons. Moreover, the ADS has the unique potential to start a nuclear energy system without an initial inventory of fissile material, i.e. it could be used to launch a new nuclear era at a time when all ^{235}U or plutonium stocks are exhausted. Such applications, however, are not in the scope of the present study.

2.8.2 Role of ADS in the shut-down phase

For a nuclear energy scenario with a finite time horizon, the full benefit from transmutation can be realised only if, in the shut-down phase, the TRU inventory of the system is burnt and not put to waste. Core analyses confirm that sub-critical as well as critical fast-spectrum burners can operate in a multi-recycling mode without any fuel top-up. The inventory burning process will, of course, stop when the remaining fuel mass becomes smaller than a single reactor inventory.

The most rapid nuclear phase-out is achieved, if all LWRs (or other TRU supplier reactors) are taken out of service at the same instant. In this case, the total heavy metal inventory of the burner system decreases exponentially with a half-life of

$$T_{1/2}^{Inv} = \ln 2 \; S/(\gamma \; r \; f),$$

where the parameters S, r and f are the power-specific heavy metal inventory of the burner, the in-pile time fraction of the fuel and the load factor of the burner, respectively. The constant γ, the mass-energy conversion factor, has a value of $\simeq$1.0 kg/GWdth.[31]

Table 2.5 gives a numerical example for each of the four TRU burner options referred to in Table 2.2. It is evident that, in the shut-down phase of a nuclear energy scenario, a thermal neutron spectrum has a theoretical advantage. In more moderate and probably more realistic shut-down scenarios, however, the supplier reactors will remain in service until they have reached their useful life-time; the shut-down phase will consequently be longer and not depend so strongly on the burner type.

2.9 Fission product transmutation

As mentioned before, the primary concern of geologic repositories are possible releases of the relatively mobile fission products. Since the fission product yields are not very sensitive to the fuel composition and the neutron spectrum of the reactor,[32] the fission product risk depends primarily on the number of fissions, i.e. the energy, produced in the fuel. This means that the fission product risk cannot be much influenced by the actinide transmutation strategy and can only be mitigated by separating troublesome fission products from the waste.

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31. In the derivation of this equation it was assumed that the specific burner inventory, S, remains constant during the shut-down phase. Realistic phase-out simulations show that this is a questionable assumption.
32. The neutron spectrum of the reactor can, however, influence the in-situ transmutation of a fission product.

Table 2.5. **TRU burner characteristics in nuclear phase-out scenario**

TRU burner option	HM inventory (kg/MWth)	In-pile time fraction	Load factor	$T_{1/2}^{Inv}$ (y)
Fast, critical[1]	11.14	0.62	0.85	40.1
Fast, ADS[1]	3.48	0.50	0.80	16.5
Thermal, critical[2]	2.2[4]	0.63	1.0[5]	6.6
Thermal, ADS[3]	1.34	0.5	1.0[5]	5.1

1. Same as TRU burners in Table 2.3.
2. AMSTER molten-salt TRU incinerator with uranium top-up [19,23].
3. ATW molten-salt incinerator pure TRU fuel [15,16].
4. TRU inventory only.
5. On-line reprocessing of the fuel.

The neutron capture process is currently the only promising nuclear reaction for transmuting fission products. Other processes which have been proposed in the past rely on technologies which are still at a very early stage of development (e.g. fusion neutron sources) and generally suffer from a poor energy balance. The capture process consumes neutrons, but fast reactors could deliver enough excess neutrons to allow the potentially troublesome long-lived fission products to be completely transmuted to shorter-lived or stable species.[33]

The transmutation of a fission product makes sense only if the reaction rate (microscopic cross-section times neutron flux) is higher than the natural decay rate of the nuclide. With the practically achievable neutron fluxes, this condition cannot be met for the most abundant fission products ^{137}Cs and ^{90}Sr with half-lives of only about 30 years, i.e. these fission products are “non-transmutable”. However, since their radioactive life is limited to less than 300 years, they can be safely enclosed using engineered barriers only. In many cases, the necessity of an isotopic separation and difficulties in the target preparation present other important obstacles for the fission product transmutation.

Long-lived fission products which dominate the long-term risk of HLW repositories are, in order of decreasing half-live, ^{129}I, ^{135}Cs, ^{99}Tc, ^{126}Sn and ^{79}Se. Activation products such as ^{14}C and ^{36}Cl can also contribute to the dose. The relative radiological importance of these nuclides varies depending on the repository concept and the type of host rock (see Section 2.2, Figure 2.2). From the characteristics in Table 2.6, it follows that, in practice, only ^{129}I and ^{99}Tc can be transmuted and the radiological impact of the other long-lived fission products can be reduced only by special conditioning and confinement. More detailed information on the transmutability of long-lived fission products is given in Chapter 8, Section 8.2.

33. Table 2.1 shows that an ADS does not necessarily deliver more excess neutrons than a normal fast reactor. The reason is that the particular ADS core has been optimised for actinide burning. It follows that a burner core cannot be optimised simultaneously for best actinide and best fission product transmutation.

Table 2.6. **Transmutability of long-lived fission products**

Fission product	$T_{1/2}^{Decay}$ **(y)**	$T_{1/2}^{Trans}$ **(y)**[1]	**Isotopic separation**	**Transmutable (yes/no)**
^{129}I	$1.6 \cdot 10^{7}$	51	no	yes[2]
^{135}Cs	$2.3 \cdot 10^{6}$	170	yes	no
^{99}Tc	$2.1 \cdot 10^{5}$	51	no	yes
^{126}Sn	$1.0 \cdot 10^{5}$	$4.4 \cdot 10^{3}$	yes	no
^{79}Se[3]	$6.5 \cdot 10^{4}$	$2.2 \cdot 10^{3}$	yes	no

1. Thermal flux: 10^{14} n/cm^2·s.
2. R&D necessary to improve the iodine separation yield and the stability of the target material.
3. Half-lives for ^{79}Se around $6.5 \cdot 10^{4}$ years have been used widely in waste inventory and repository performance assessments. Recent nuclear data studies, however, indicate a much longer half-live for this nuclide (see http://nucleardata.nuclear.lu.se/nucleardata/).

Finally, it should be noted that the transmutable fission products represent only a small fraction of all fission products and that the vitrified waste mass, which is primarily determined by the fission products, therefore, *cannot be much reduced* by P&T operations. A combined transmutation-conditioning strategy for the long-lived fission products, however, would allow easing the licensing requirements for vitrified waste repositories because the geosphere barrier would no longer have an important safety function.

3. COMPARATIVE ANALYSIS

3.1 Principal fuel cycle schemes

3.1.1 Basis for the selection

A consistent comparison of transmutation systems implies an analysis of the systems using a single nuclear data library, a single reactor code system, and consistent input data for reactor and fuel cycle parameters. It is clear that such an exercise cannot comprehend all possible systems and hence involves a selection of representative systems or "schemes". The selection was guided by the rationale for introducing P&T in future advanced fuel cycles with emphasis on basically different approaches. The applied criteria and assumptions can be summarised as follows:

- The comparative analysis is restricted to actinide transmutation systems based on *critical and accelerator-driven fast reactors*. Most of the currently proposed transmutation systems belong to this category. Actinide burners with a thermal neutron spectrum are described in Chapter 9.
- A particular goal of the study is to compare *evolutionary and innovative approaches* to the transmutation of actinides, characterised by the separate handling or the co-processing of plutonium and minor actinides, respectively.
- Since, in the medium-term, resource efficiency will probably not become an issue of primary importance, LWRs can be expected to remain important components of all advanced nuclear energy systems. The burner reactors of the fuel cycle schemes are therefore optimised for a high burning efficiency so that they can support a *large fraction of LWRs*.
- Assuming radiotoxicity reduction to be the primary goal, only TRU and minor actinide burners with *fully closed fuel cycles* are considered although such systems cannot be expected to be operational on industrial scales before several decades. (Systems with partially closed fuel cycles cannot achieve such high radiotoxicity reductions as systems with fully closed fuel cycles).
- Since the start-up and the shut-down phase in transient nuclear energy scenarios depend on many boundary conditions which are subject to large uncertainties, the analyses of the schemes are restricted to steady-state conditions, meaning that the results apply only if the reactor park is operated for a very long time (transients aspects are briefly discussed in Chapter 2, Section 2.8.1).

Six principal fuel cycle schemes are found to be sufficient to illustrate and quantify the essential features of FR- and ADS-based systems for the burning of plutonium and the transmutation of minor actinides. These include an LWR-once through scheme as a reference case, a plutonium burning scheme, three transmutation schemes in which critical and sub-critical fast reactors perform specific functions, and a pure fast reactor scheme which represents the long-term goal of the nuclear development. Other transmutation approaches as e.g. the heterogeneous recycling of americium and

curium are considered as variants of the six principal schemes (cf. Section 3.3). For convenience, the six principal fuel cycle schemes will be referred to as follows:

1) *LWR once-through*

   A strategy based on modern PWRs with direct disposal of the spent fuel. This is the reference case for the comparisons.

2) *Plutonium burning*

   A representative plutonium burning strategy based on LWRs and fast reactors. An LWR-MOX stage is incorporated in the scheme, because MOX recycling in LWRs is already a standard practice in Europe. MOX-fuelled fast reactors with a low conversion ratio are used to maximise the LWR-to-FR ratio (an accelerator-driven plutonium burner is not considered, since plutonium burning is not in the focus of the present study).

3a) *TRU burning in FR*

   A "two component" transmutation system based on normal LWRs and ALMR-type critical fast reactors. The top-up fuel of the latter contains fertile uranium such that the passive safety regime and applicability of fuels irradiation database remain valid. This precludes reducing the breeding ratio below about 0.5 (the LWR-to-FR ratio is a function of the FR breeding ratio).

3b) *TRU burning in ADS*

   Same strategy as scheme 3a with the difference that the fast reactor is replaced by an ATW-type sub-critical fast system. The capability of the latter to burn pure transuranics improves the LWR-to-FR ratio.

4) *Double strata*

   A second, so-called P&T stratum with accelerator-driven, dedicated minor actinide burners is added to the plutonium burning scheme. A particular advantage of this "three component" transmutation system is that the investment in innovative reactor and reprocessing technology is minimised.

5) *Fast reactor strategy*

   A fast reactor strategy featuring a fully closed fuel cycle and natural uranium top-up. This strategy represents the long-term goal for the nuclear development, since it fulfils the resource-efficiency and environmental friendliness requirements simultaneously.

3.1.2 Reactor and fuel cycle characteristics

The fuel cycles of the principal schemes are sketched in Figures 3.1 to 3.6. The figures contain information on the net electric power installed in the reactor components (all reactors are assumed to be electricity producers), the fuel cooling and storage time before reprocessing and after fabrication, and the reprocessing method ("wet" or "dry"). Additional information on the fuels (composition, burn-up, fuel management, etc.) is given in Table 3.1. It should be noted that the actinide mass flows in the fuel cycles determine the support ratios, and that the values for the latter refer to net electric power delivered to the grid, i.e. account for the electricity recycled in the accelerator of an ADS. The fuel cooling and storage times - the fabrication is assumed to take place immediately after

the reprocessing – determine the out-of-pile time of the fuel and hence influence the total actinide inventory of the fuel cycle.

The fuel cycle parameters, especially the fuel burn-up and the reprocessing losses, strongly influence the results of the comparative analysis. The adopted values represent a consensus of experts from the major OECD countries with reprocessing experience. The value of 0.1% for the reprocessing losses is an extrapolation from the current technology to a technology which can be expected to work at a time when transmutation systems could be introduced on a larger scale. The extrapolation is based on expected and partly at laboratory scale proven advances in the wet and dry reprocessing technology. The assumptions are comparable to assumptions which have been made in other national and international transmutation studies.

The schemes involve the following reactor components:

- *LWR-UOX*

  Fuel as discharged from a French N4 pressurised water reactor. The UOX fuel is irradiated to a burn-up of 50 GWd/tHM.

- *LWR-MOX*

  N4 reactor with a 100% MOX core. The initial plutonium vector is that of the discharged UOX fuel, and the MOX fuel is irradiated to a burn-up of 50 GWd/tHM.

- *Plutonium burner*

  EFR-type reactor with MOX fuel. Low conversion ratio, high burn-up CAPRA core [24]. The plutonium is recycled indefinitely, and the plutonium-to-uranium ratio of the top-up fuel is adjusted to obtain $k_{eff} = 1$ at EOEC assuming a six-batch core. The plutonium feed is the plutonium separated from the LWR discharged MOX fuel. The maximum plutonium enrichment of the fuel for the equilibrium core is 44% at beginning-of-life.

- *Critical TRU burner*

  ALMR-type actinide burner with a conversion ratio of 0.5. The reactor model is the same as that used in the 600 MW(e), metal-fuelled, multiple recycle burner core benchmark exercise of the NEANSC Working Party on Plutonium Recycling [25]. The TRU-to-uranium ratio of the top-up fuel is adjusted to obtain $k_{eff} = 1$ at EOEC assuming a five-batch core. The TRU feed consists of the TRU in the LWR discharged UOX fuel, the U feed is depleted uranium. The TRU content of the fuel for the equilibrium core is 33% at beginning-of-life.

- *Accelerator-driven TRU burner*

  The reactor model is based on the ANL design of a lead-bismuth cooled sub-critical TRU burner [26]. The actinide-to-zirconium ratio of the metal fuel is adjusted to obtain a k_{eff} of about 0.97 at BOEC assuming a six-batch core. The top-up fuel consists of undiluted TRUs as discharged from the LWRs. Since the fuel is uranium-free, the reactor has a low heavy metal inventory.

- *Accelerator-driven MA burner*

  Modular concept which is compatible with the dimensions the ALMR Reference Model A design [27] and, except for a smaller core radius, resembles the lead-bismuth cooled system proposed by JAERI [20]. The reactor model is the same as that used in the NEANSC comparison calculations for an accelerator-driven minor actinide burner [28]. A pure minor

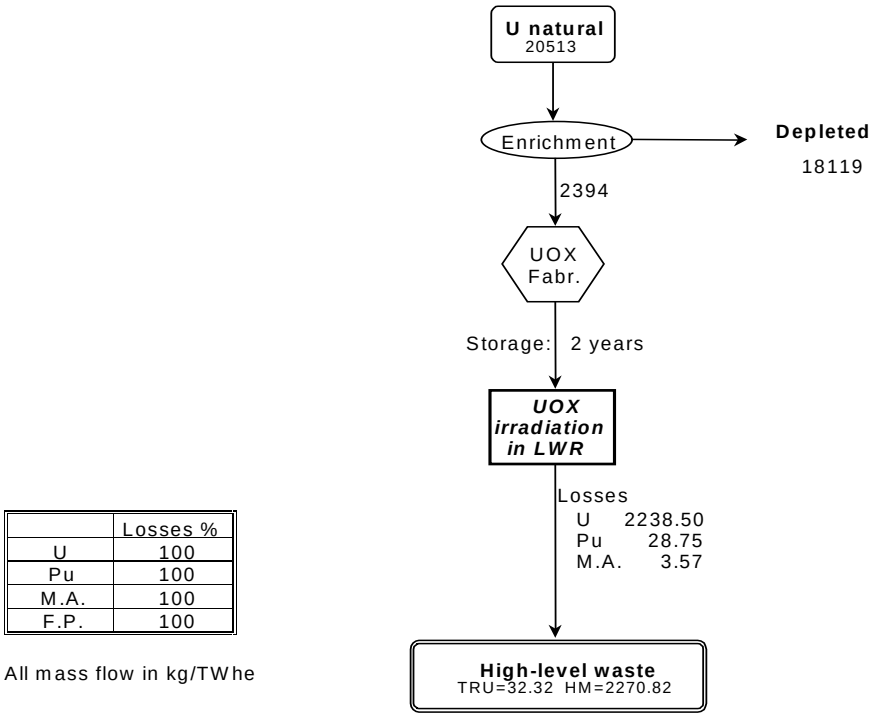
actinide top-up as produced by the first stratum of the plutonium burning scheme (scheme 2) is assumed. Following the Japanese preference, the fuel consists of actinide mononitrides. The actinide-to-zirconium ratio is set to give a k_{eff} of about 0.95.

- *Fast reactor*

  ALMR-type fast reactor. The reactor model is the same as that for the critical TRU burner with the difference that the lower steel reflector and the outermost fuel element ring are replaced by uranium blankets. The fuel and the blankets are reprocessed together, the new blankets are fabricated from reprocessed uranium, and the new fuel is fabricated from reprocessed TRU, reprocessed uranium and a natural uranium top-up. The actinide-to-zirconium ratio is adjusted to give a conversion ratio of 1.0 and a k_{eff} of 1.0 at EOEC assuming a five-batch core.

It should be noted that the TRU burners, the MA burner and the fast reactor have fully closed fuel cycles, i.e. all actinides are recycled until the fuel composition reaches an equilibrium.

Figure 3.1. **Once-through strategy (scheme 1)**



	Losses %
U	100
Pu	100
M.A.	100
F.P.	100

All mass flow in kg/TWhe

Figure 3.2. **Plutonium burning strategy (scheme 2)**

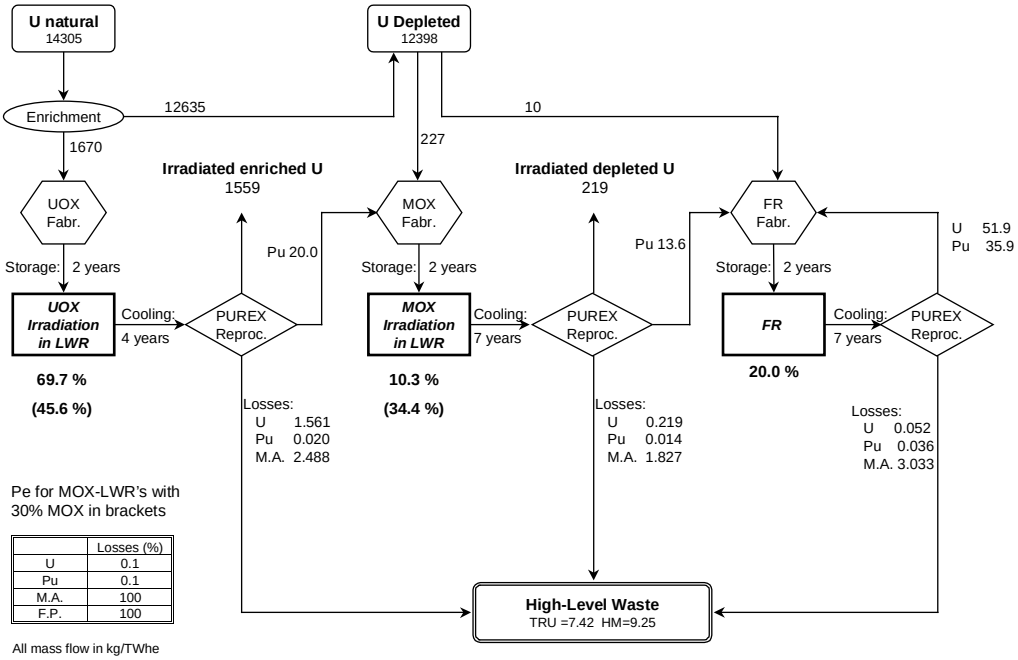


Figure 3.3. **Double strata strategy (scheme 4)**

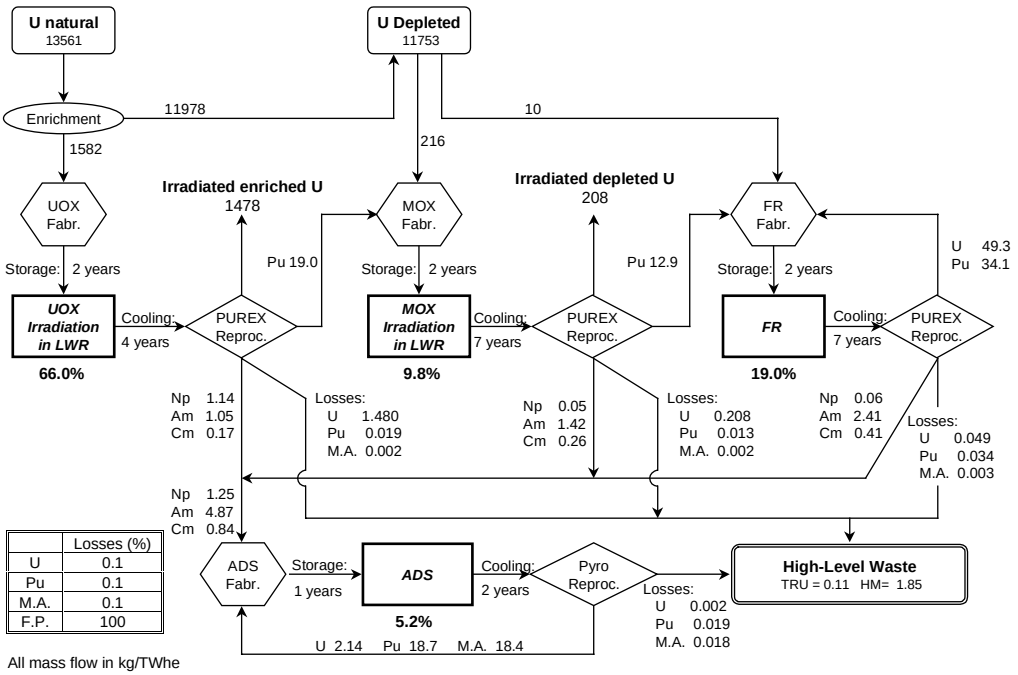


Figure 3.4. **TRU burning in fast reactor (scheme 3a)**

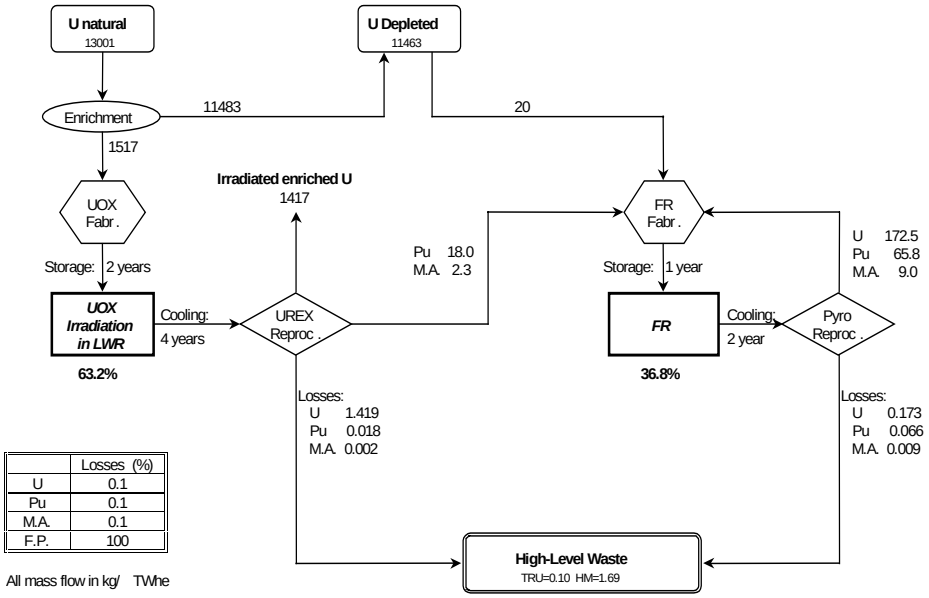


Figure 3.5. **TRU burning in ADS (scheme 3b)**

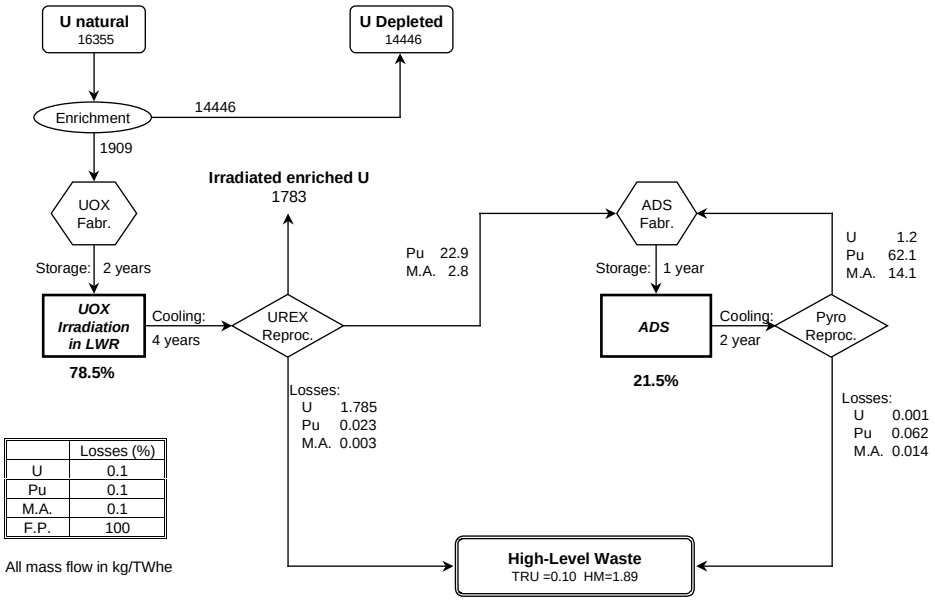


Figure 3.6. **Fast reactor strategy (scheme 5)**

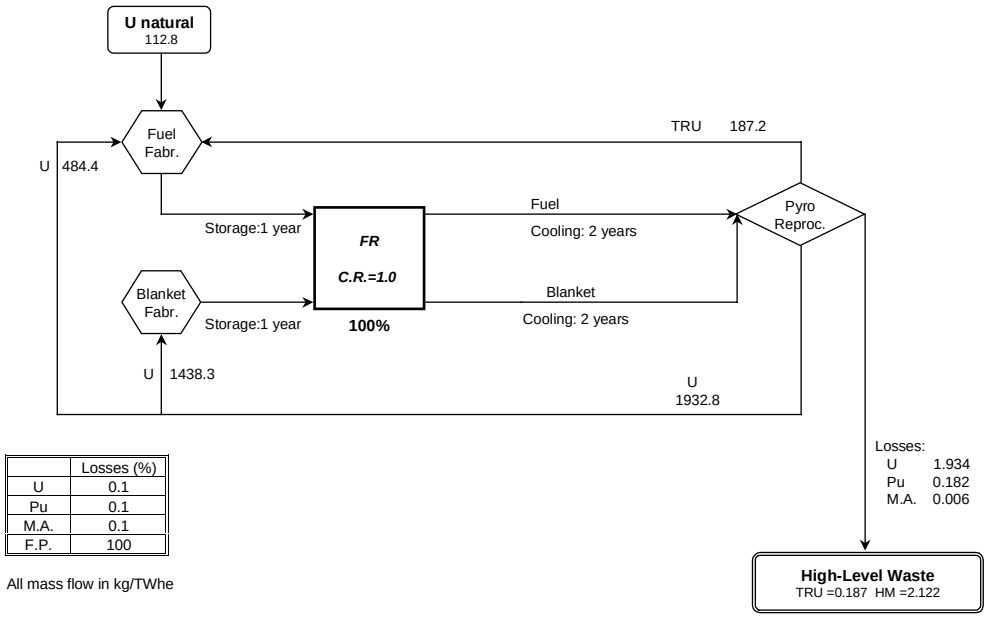


Table 3.1. **Design parameters of fast-spectrum systems**

Design parameter	Pu burner (FR)	TRU burner (FR)	TRU burner (ADS[1])	MA burner (ADS[1])	Fast reactor
Thermal power (MW)	3 600	1 575	840	377	1 575
Net electric power (MW)	1 450	600	275	119	600
Load factor (%)	85	85	80	85	85
Coolant	Na	Na	Pb-Bi	Pb-Bi	Na
Fuel	MOX	^{78}Ac-^{22}Zr	^{25}Ac-^{75}Zr	^{29}AcN-^{71}ZrN	^{79}Ac-^{21}Zr
HM Inventory, BOL (kg)	25’690	17’551	2 923	3 145	17’732[3]
Fuel burn-up (GWd/tHM)	185	139	250	149	127[3]
In-pile time (d)	1 553	1 825	1 093	1 460	1 825
No. of batches	6	5	6	1	5
TRU in top-up fuel (%)[2]	57.6	50.0	100	100	0

1. For the accelerator and the target the following assumptions are made: electrical efficiency = 45%, proton energy = 1 GeV, neutrons per proton = 42.
2. Content of top-up fuel in transuranic actinides.
3. Core without blankets.

3.2 Comparative assessment

3.2.1 Calculation methods

This section overviews the calculation methods used in the analyses of the principal fuel cycle schemes. More detailed information regarding methods and results can be found in [29].

The analyses involved the following well-validated computer code systems:

- *Light water reactors*

  The analyses were performed by CEA Cadarache using the deterministic thermal reactor analysis code system Apollo1-Cesar [30].

- *Fast-spectrum systems*

  The analyses were performed at PSI Villigen using version 1.2 of the European fast reactor analysis code system ERANOS [31] in combination with the reference ERALIB I libraries, based on JEF 2.2 basic nuclear data. The code system had previously been tested for actinide burner applications in the framework of the NEANSC comparison calculations described in [28].

- *Spallation neutron source*

  The PSI version of the high-energy Monte Carlo code HETC was used. HETC simulates the interactions in the target above a cut-off energy of 20 MeV and edits a spatially dependent external neutron source for use with multi-group transport calculations below the cut-off energy. A proton energy of 1 GeV was assumed.

The fast-spectrum systems were modeled in R-Z geometry and analysed using S_4 neutron transport theory with 33 broad energy groups below 20 MeV. The broad group cross-sections were obtained from fundamental-mode cell calculations with 1968 fine energy groups, taking the cell heterogeneities into account. For the sub-critical systems, in-homogeneous reactor calculations with the HETC source were performed and from these the level of the external source for the given thermal power of the systems was evaluated. The equilibrium fuel compositions were calculated using detailed chains including 29 actinides up to ^{248}Cm and 80 explicit fission products.

The evaluation of the equilibrium fuel composition required repeated burn-up calculations with subtraction of the reprocessing losses and addition of the top-up fuel after each iteration. The shut-down times for fuel reloading between reactor cycles and the fuel storage and cooling times were correctly simulated throughout this iterative procedure, and the procedure was repeated until equilibrium was reached, i.e. the differences between two successive fuel compositions were smaller than 0.05% for all relevant nuclides.

For determining the end-of-life fuel composition, it was sufficient to carry out the equilibrium calculation for a single batch core. The neutron multiplication factors for the multi-batch cores were calculated for average fuel compositions which were evaluated as follows: the composition of the fuel at BOEC was obtained by averaging the equilibrium fuel composition at appropriate burn-ups, and this fuel composition was then irradiated during one reactor cycle to obtain the average fuel composition at EOEC.

In an ideal critical reactor, the neutron multiplication factor reaches 1.0 at EOEC. In the core simulations, this was achieved by adjusting the uranium-to-TRU ratio of the top-up fuel. For the sub-critical systems of schemes 3b and 4, this is not possible because the top-up fuel consists of pure TRU. In these cases, the k_{eff} was adjusted by slightly modifying the actinide-to-zirconium ratio of the fuel. This allowed to conserve the geometry of the benchmark models.

The long-term evolutions of the waste activity were calculated with the ORIHET 3 code, an adaptation of the code ORIGEN which uses a decay data library based on NUBASE data. The activities were converted to ingestion doses using the dose conversion factors from [32] (see also

Annex D). The near-field release rates for the vitrified waste arising in schemes 2 and 4 and from an LWR once-through vitrification strategy were calculated by Nagra, Wettingen, using an improved version of the near-field analysis code STRENG.

The decay heat and the neutron source strength of the fuel was calculated with the MECCYCO code [33] which uses nuclear data from JEF. As the 80 explicit fission products used in the establishment of the equilibrium for the actinides were found to be insufficient for accurate decay heat predictions, an additional cycle of the fuel was simulated with 160 explicit fission products. An improvement had also to be applied to the neutron source strength calculations: To account for neutron source contributions from spontaneous fissions in ^{250}Cf and ^{252}Cf, which are significant for the multi-recycled TRU and minor actinide burner fuels, neutron source strength calculations using a special chain library which contains berkelium and californium isotopes had to be carried out.

3.2.2 Equilibrium core characteristics

The principal neutronic parameters of the equilibrium cores, including the k_{eff} at the end of the equilibrium cycle (EOEC), the burn-up reactivity drop during the cycle, the external neutron source strength, the median energy of the core-averaged neutron spectrum, and important safety parameters, are compiled in Table 3.2. Figure 3.7 shows the evolution of the k_{eff} during the equilibrium cycle.

Table 3.2. **Equilibrium core characteristics**

Core parameter	Pu burner (FR)	TRU burner (FR)	TRU burner (ADS)	MA burner (ADS)	Fast reactor
k_{eff}, EOEC	1.005	1.005	0.920	0.932	0.999
Δk_{eff}, BOEC-EOEC	0.045	0.044	0.051	0.019	0.041
Ext. neutron source (n/s)[1]	–	–	$6.36 \cdot 10^{18}$	$2.53 \cdot 10^{18}$	–
Neutron source importance[2]	–	–	0.959	0.843	–
Void effect (pcm)[3], core	2 245	1 775	-3 302	2 861	1 397
Core and upper reflector	1 904	-1 758	-4 933	2 263	-2 025
Fuel Doppler effect[4]	1 908	555	308	75	806
β_{eff} at BOEC (pcm)	318	323	260	161	346
Median energy (keV)[5]	132	242	189	173	228

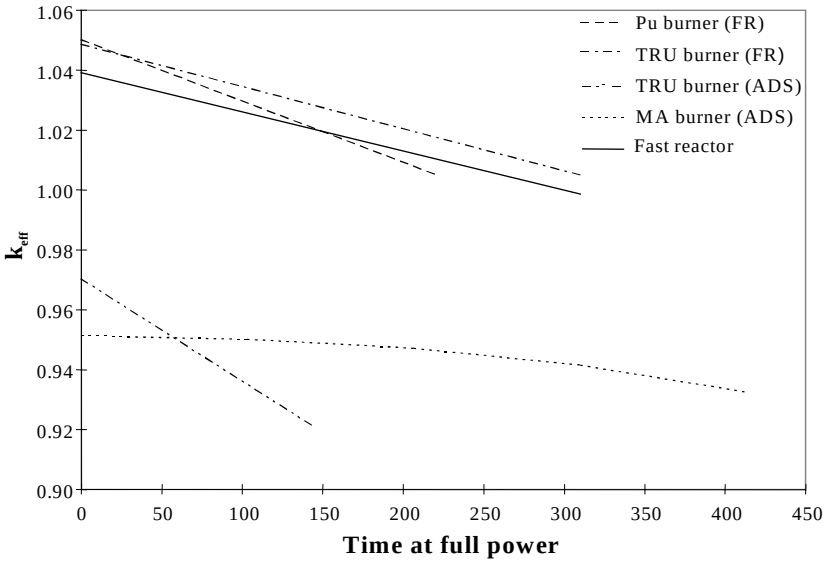
1. No. of source neutrons per second below 20 MeV to maintain design power at EOEC.
2. Neutron source importance at EOEC evaluated as $(1/k_{eff}-1)/(1/k_{source}-1)$.
3. Coolant void reactivity effect at BOEC calculated as $(k_{eff}(\text{voided}) - k_{eff}(\text{reference}))/k_{eff}(\text{reference})$.
4. Fuel Doppler effect at BOEC calculated as 10^6 $(k_{eff}(T_{ref}) - k_{eff}(T_{ref}+600K))/(k_{eff}(T_{ref})\ k_{eff}(T_{ref}+600K))$.
5. Median energy of the core-averaged neutron spectrum at BOEC.

The burn-up reactivity drop depends on the reactor type, the fuel burn-up and the core reloading fraction at the end of a reactor cycle. For the critical cores, the latter was optimised to obtain similar reactivity drops and hence excess reactivities at the beginning of the cycle (BOEC). Due to the steeper reactivity gradient, the plutonium burner core has, therefore, a shorter cycle length than the other critical cores. Assuming the plant availability not to be much influenced by the somewhat different cycle lengths, a constant load factor of 85% was used in the analyses of all critical cores.

It is interesting to compare the different neutronic behaviour of the two ADS cores. Whereas the conversion of fertile into fissile actinide species stabilises the k_{eff} of the minor actinide burner core to the extent that it can operate easily with a single batch core, the fertile-free, sub-critical TRU burner core exhibits the steepest reactivity gradient and hence the shortest cycle length of all cores. In the

analyses, the unusually short cycle length of the sub-critical TRU burner was accounted for by a 5% reduction in the load factor; the limited availability of the accelerator, however, suggested not to credit the load factor of the minor actinide burner for the long cycle length of the single batch core.

Figure 3.7. **Evolution of effective multiplication factor during equilibrium cycle**
(time unit: 3 days for MA burner, 1 day for other systems)



The external neutron source strength at EOEC is important because it determines the accelerator design power. The energy which is recycled to the accelerator increases during the burn-up; its average value amounts to 11.6% for the TRU and 12.6% for the minor actinide burner. It is interesting to note that, due to the different burn-up reactivity behaviour of the two sub-critical cores, the minor actinide burner features a higher recycled energy fraction, but a lower accelerator design power (-8% for the same net electrical output, evaluated from the external sources in Table 3.2).

A comparison of the median energies does not confirm the relevance of the often-cited spectrum hardening effect due to the external spallation neutrons. For example, among the metal-fuelled systems, the ADS has the softest neutron spectrum. This, at first, unexpected result is mainly a consequence of the requirement to reduce the fuel density in transmuters with a diluent such as zirconium (see fuel compositions in Table 3.1). It is clear that, although a very hard neutron spectrum cannot be considered a salient feature of a transmuter, the overall neutron balance condition discussed in Chapter 2, Section 2.4.1, which favours the use of fast neutrons especially for the transmutation of minor actinides, must nevertheless be respected.

As to the safety parameters of the cores, the analyses confirm the well-known fact that they deteriorate with increasing minor actinide contents of the fuel. Consequently, the minor actinide burner core features the least favourable coolant void and fuel Doppler reactivity effects, and the effective fraction of delayed neutrons, β_{eff}, for this core is about halved compared with that of a normal fast reactor. The low β_{eff} value would make the control of a critical minor actinide burner core very delicate and suggests the operation of such a core at substantial sub-criticality (the k_{eff} was adjusted to achieve at any time a sub-criticality level of about 5%). In the case of the accelerator-driven TRU burner, which also features a lead-bismuth cooled core, the safety parameters are much more benign, meaning that the incentive to operate the core in a sub-critical state is reduced.

3.2.3 Actinide waste production

A detailed balance of the actinide waste production, including a breakdown by reactor components, is given in Table 3.3. Important features of this balance are illustrated in Figures 3.8 and 3.9.

Since the actinide waste production depends on the fuel burn-up (see Chapter 2, Section 2.3.2) and the burn-up capability of different types of MA- and TRU-dominated fuels has not yet been verified experimentally and is therefore uncertain, the actinide waste production of the fast-spectrum systems using metal and nitride fuels was scaled so that it corresponds to a fixed reference burn-up of 140 GWd/tHM (using the notation of Section 2.3.2, the scaling factor is $\delta(B_{ref})/\delta(B_{nom})$, where B_{ref} and B_{nom} are the reference burn-up and the nominal burn-up in Table 3.1, respectively). This means that the observed trends are characteristic for the scheme and not for the burn-up of the metal and nitride fuels. It is clear that, for any fuel type, an increase in the burn-up results in a reduction of the fuel losses. For each scheme, the actinide waste production is normalised such that it corresponds to 1 TWhe of energy produced by the reactor park.

It can be seen that all recycling strategies reduce the plutonium waste mass at least by a factor of 150 compared with the once-through case. The plutonium-burning scheme is more effective than any of the transmutation schemes in eliminating the plutonium, but has the disadvantage of producing 3 to 4 times more americium and curium than the once-through scheme (about half of the americium and curium is generated by the fast plutonium burners). Its tendency to convert plutonium into minor actinides limits the TRU waste mass reduction potential to the modest factor of 4.4.

The importance of a fully closed fuel cycle is highlighted by the fact that the transmutation schemes and the fast reactor scheme achieve TRU waste mass reduction factors of at least 175 relative to the once-through case. Among the innovative transmutation schemes, scheme 3a and scheme 3b, the former appears to have a small advantage due to the higher uranium and hence lower TRU fraction in the fuel (the compensation of the difference in the TRU waste production between the two schemes would require an increase in the average burn-up of the ADS fuel from 140 to 270 GWd/tHM). The double strata scheme performs nearly as well as scheme 3a.

The fast reactor scheme is a special case because it represents a breeding strategy and the generation of plutonium is part of this strategy. It is interesting to note that, nevertheless, it achieves similar TRU and heavy metal waste mass reduction factors as scheme 3b; for the elimination of the minor actinides, it is more effective than any other scheme. The unique features of the fast reactor scheme appear, again, in the isotopic composition presented in Figure 3.9. Due to the breeding of new plutonium in the blankets, the plutonium vector is strongly dominated by ^{239}Pu. The absence of thermal-spectrum systems in the reactor park and the small abundance of the higher plutonium isotopes in the fuel explain the low minor actinide content of the waste.

It can be noted that all recycling strategies reduce the heavy metal waste mass by a factor of 1 100 or more compared with the once-through reference case. The variation in this factor of only 15% is mainly due to the different fractions of fast-spectrum systems in the reactor park: since the fuel in the fast-spectrum systems reaches about three times the burn-up of that in the thermal-spectrum systems, a high fraction of fast-spectrum systems is associated with a lower actinide waste production.

Depleted and reprocessed uranium, which is produced by all LWR-based schemes, does not appear in Table 3.3 because it is considered as a resource. The non-negligible long-term radiological impact of uranium is discussed in Section 3.4.4.

Table 3.3. **Actinide waste production for principal fuel cycle schemes (g/TWhe)**

	O-T	**Pu burning**			**TRU burning in FR**			**TRU burning in ADS**			**Double strata**				**FR strategy**		
	LWR	LWR	FR	Total	LWR	FR	Total	LWR	ADS	Total	LWR	FR	ADS	Total	Fuel	Blanket	Total
^{234}U	11.19	0.06	0.24	0.30	0.01	0.62	0.63	0.01	1.65	1.66	0.06	0.22	1.57	1.85	0.23	0.46	0.70
^{235}U	18356	13.07	0.07	13.15	11.63	0.18	11.81	14.63	0.39	15.03	12.39	0.07	0.37	12.84	0.48	0.68	1.16
^{236}U	13207	9.28	0.14	9.42	8.37	0.24	8.61	10.53	0.51	11.04	8.80	0.14	0.35	9.28	0.49	1.22	1.71
^{238}U	2207E3	1757.9	51.5	1809.4	1398.7	171.5	1570.2	1759.5	0.0	1759.5	1666.5	48.8	0.0	1715.3	493.6	1264.2	1757.9
^{238}Pu	1006	1.39	0.88	2.27	0.64	3.55	4.19	0.80	10.66	11.46	1.32	0.83	6.84	8.99	2.08	0.02	2.10
^{239}Pu	14915	15.57	10.03	25.60	9.45	25.49	34.94	11.89	22.95	34.84	14.76	9.51	1.56	25.83	77.88	34.20	112.09
^{240}Pu	6847	8.91	15.75	24.66	4.34	24.86	29.20	5.46	57.25	62.71	8.44	14.94	7.83	31.21	42.49	1.13	43.62
^{241}Pu	3711	4.39	2.22	6.61	2.35	3.31	5.66	2.96	9.98	12.94	4.16	2.11	0.89	7.15	4.25	0.02	4.27
^{242}Pu	2274	3.43	7.06	10.49	1.44	8.56	10.00	1.81	26.27	28.09	3.25	6.70	2.95	12.90	3.52	0.00	3.52
^{237}Np	1724	1257.2	65.6	1322.9	1.09	1.61	2.70	1.37	3.64	5.01	1.19	0.06	2.57	3.83	1.02	0.19	1.21
^{241}Am	909.8	1626.5	1394.6	3021.1	0.58	2.55	3.13	0.73	6.59	7.32	1.54	1.32	4.84	7.71	2.29	0.00	2.29
^{242m}Am	0.0	6.7	32.4	39.0	0.00	0.18	0.18	0.00	0.49	0.49	0.01	0.03	0.45	0.49	0.14	0.00	0.14
^{243}Am	670.3	964.7	1112.1	2076.9	0.42	2.22	2.65	0.53	7.51	8.04	0.91	1.05	4.47	6.44	0.70	0.00	0.70
^{242}Cm	0.0	0.02	0.09	0.11	0.00	0.01	0.01	0.00	0.03	0.03	0.00	0.00	0.01	0.01	0.01	0.00	0.01
^{243}Cm	0.0	2.60	6.01	8.61	0.00	0.02	0.02	0.00	0.08	0.08	0.00	0.01	0.05	0.06	0.01	0.00	0.01
^{244}Cm	239.4	396.54	373.95	770.49	0.15	1.86	2.01	0.19	7.93	8.12	0.38	0.35	5.39	6.12	0.51	0.00	0.51
^{245}Cm	23.9	56.77	45.67	102.45	0.02	0.35	0.36	0.02	1.54	1.56	0.05	0.04	1.12	1.22	0.09	0.00	0.09
U	2238E3	1780.3	52.0	1832.3	1418.7	172.5	1591.2	1784.7	2.6	1787.3	1687.7	49.3	2.3	1739.3	494.8	1266.6	1761.4
Pu	28753	33.68	35.95	69.63	18.22	65.78	84.00	22.92	127.12	150.04	31.93	34.08	20.08	86.08	130.23	35.37	165.60
Np	1723.8	1257.2	65.6	1322.9	1.09	1.61	2.70	1.37	3.64	5.01	1.19	0.06	2.57	3.83	1.02	0.19	1.21
Am	1580.1	2597.9	2539.1	5137.0	1.00	4.95	5.95	1.26	14.59	15.85	2.46	2.41	9.77	14.64	3.13	0.00	3.14
Cm	263.4	455.93	425.72	881.65	0.17	2.24	2.41	0.21	9.57	9.78	0.43	0.40	6.57	7.41	0.62	0.00	0.62
TRU	32320	4344.7	3066.4	7411.1	20.48	74.57	95.06	25.77	154.92	180.69	36.02	36.95	38.99	111.96	135.00	35.57	170.57
HM	2271E3	6125.0	3118.4	9243.4	1439.2	247.1	1686.3	1810.5	157.5	1967.9	1723.7	86.2	41.30	1851.2	629.8	1302.2	1932.0

Figure 3.8. **TRU waste production per heavy element**

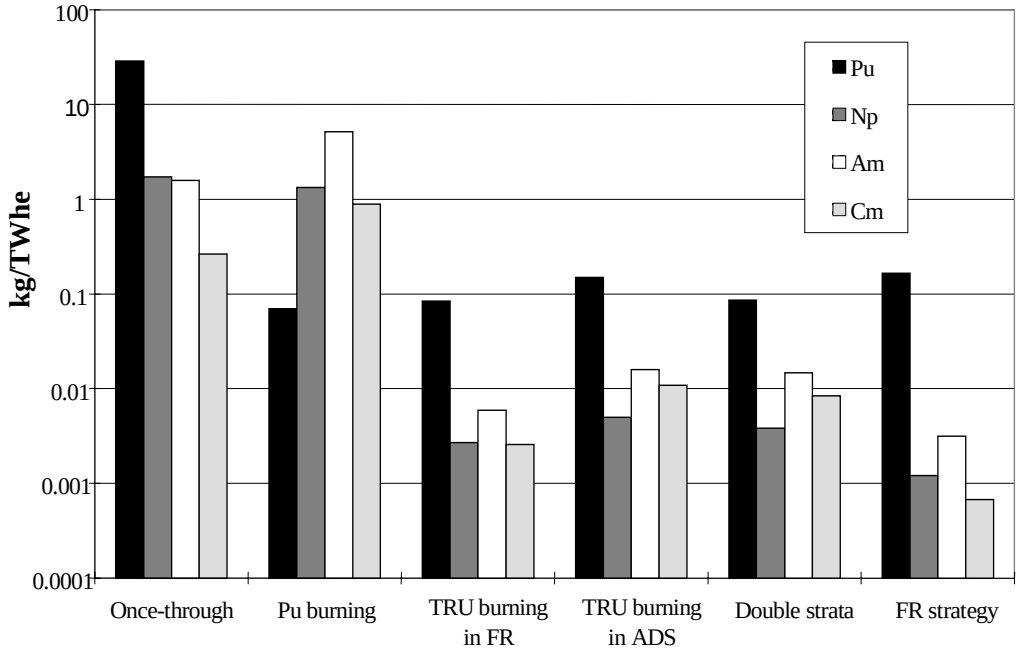
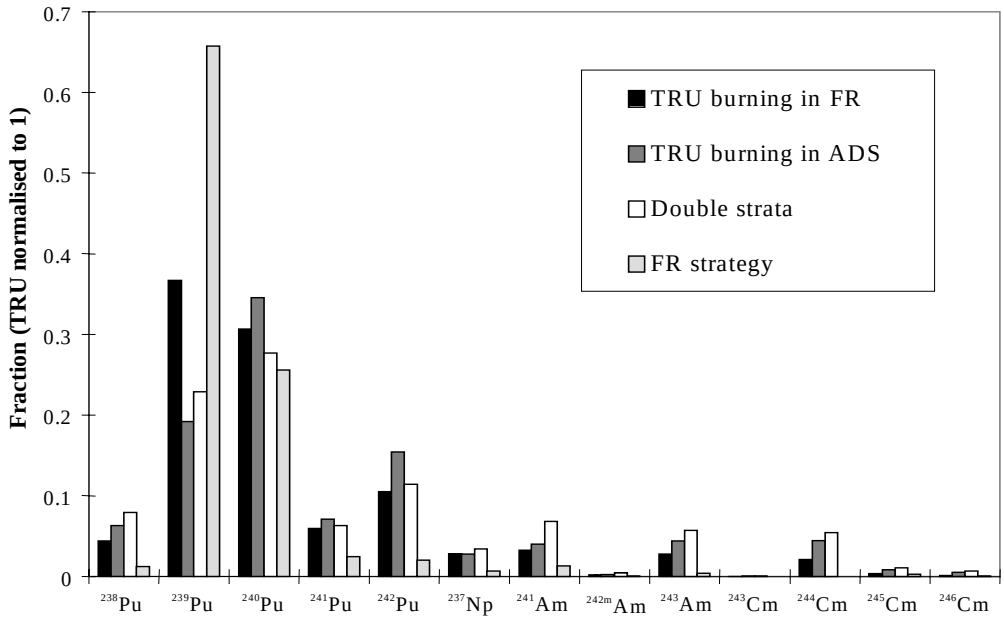


Figure 3.9. **Isotopic composition of TRU waste**



3.2.4 Radiotoxicity reduction

The actinide waste radiotoxicity is shown in Figure 3.10. It can be seen that, in the important time frame from 10^3 to 10^5 years, all transmutation schemes comfortably meet the goal of a hundred-fold radiotoxicity reduction relative to the once-through fuel cycle. On the other hand, plutonium recycling

alone reduces the radiotoxicity in this time frame only by a factor of about five. This confirms that the plutonium burning strategy cannot qualify as a transmutation strategy.

In the time frame from 10^3 to 10^5 years, schemes 3a and 4 (TRU burning in FR and double strata strategy) achieve the highest radiotoxicity reduction, and beyond 10^5 years the fast reactor strategy achieves the highest radiotoxicity reduction. For all transmutation schemes, however, the differences and fluctuations are relatively small.

To understand the strategy-dependent trends in the time evolution of the radiotoxicity, a decomposition into nuclide contributions is necessary. Figure 3.11 shows that the dominant radiotoxicity contributors are ^{238}Pu up to a few hundred years, ^{240}Pu and ^{239}Pu in the range 10^3 to 10^5 years, and ^{226}Ra (including daughter products) in the range 10^5 to 10^6 years. The latter is predominantly produced by the decay of ^{238}Pu. As this applies to all transmutation schemes, one can expect the trends to be correlated with the ^{238}Pu-to-^{239}Pu ratio of the fuel. An inspection of the plutonium isotopic composition of the fuels (see Figure 3.9) confirms the existence of such a correlation.

In accordance with the assumptions in Section 3.1.2, the afore-mentioned results apply to fuel losses of 0.1% for all actinides and reprocessing methods. For a single-stratum strategy, the influence of the fuel losses on the transmutation performance can be assessed using the expressions in Chapter 2, Sections 2.3 and 2.5. Figure 3.12 shows parametric results for the radiotoxicity reduction in the case of the double strata strategy. It can be seen that the goal of a hundred-fold radiotoxicity reduction allows the minor actinide losses in the plutonium-burning stratum to be increased by a factor of ten, but that fuel losses of only 0.1% in the minor actinide burning stratum are essential.

Figure 3.10. **Evolution of the actinide waste radiotoxicity**
(Average burn-up of metal and nitride fuel: 140 GWd/tHM)

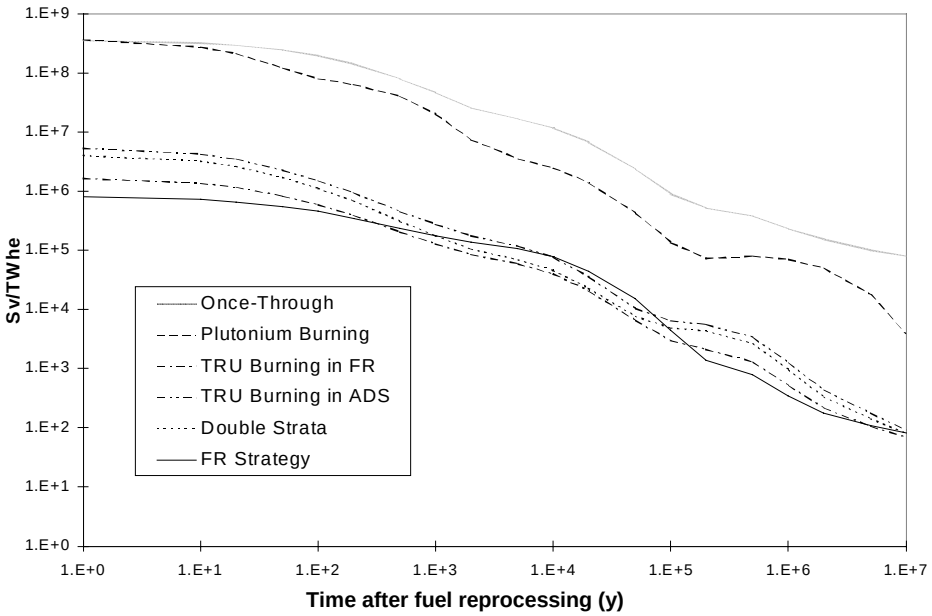


Figure 3.11. **Nuclide contributions to the actinide waste radiotoxicity for scheme 3a**

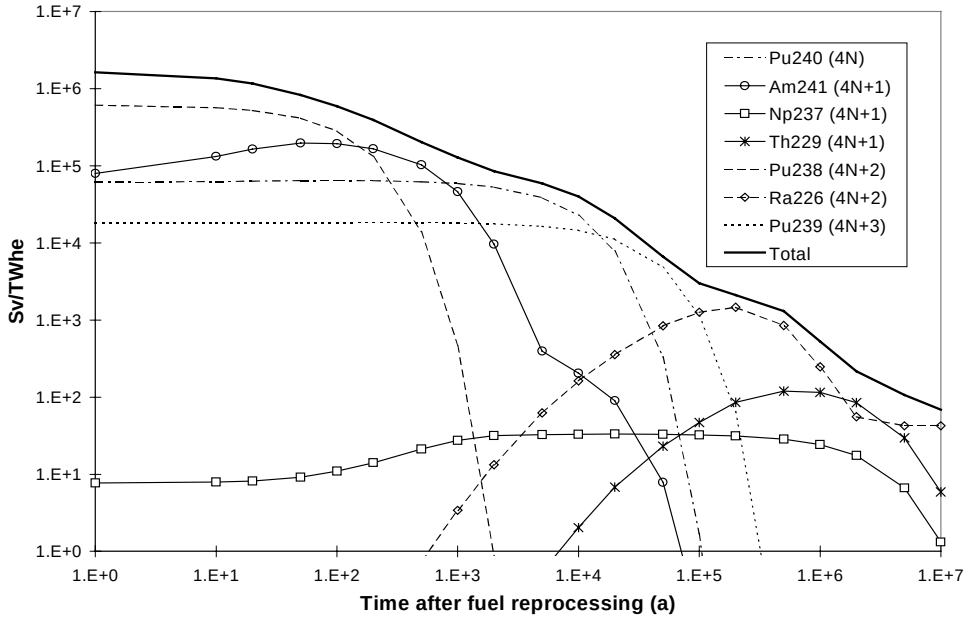
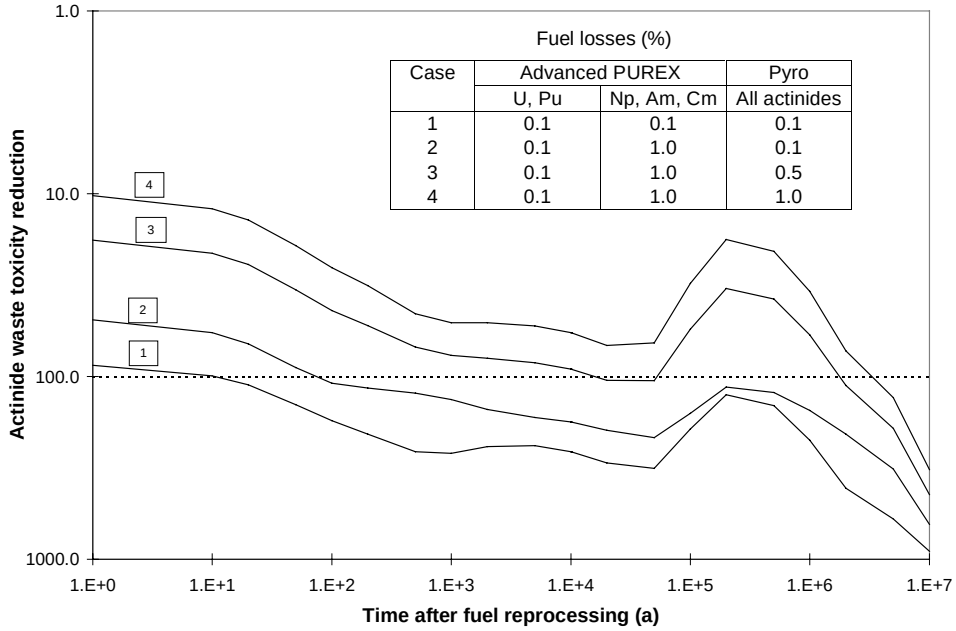


Figure 3.12. **Influence of fuel losses on the actinide waste radiotoxicity for scheme 4**



3.2.5 Long-term risk

Since the principal fuel cycle schemes involve different waste forms and repository concepts, a comparison of the associated long-term risks is not an easy task.

In principle, three cases can be distinguished:

- For the once-through fuel cycle, the direct storage of the spent fuel elements in geologic repositories is the appropriate disposal concept. Risk analyses for this concept show that the long-lived fission product ^{129}I usually dominates the annual individual dose for the population and that the risk from the actinides is generally smaller than that from the fission products. (In the US study for a repository in tuff, the dominating nuclides up to about 65 000 years are ^{99}Tc and ^{129}I, with ^{237}Np becoming predominant thereafter).
- In connection with wet reprocessing, the vitrification, i.e. the immobilisation, of the fission products and unrecovered actinides in a glass matrix, is the favoured concept. The glass is enclosed in steel canisters and the space between the canisters and the near-field/host rock interface is filled with bentonite. Whereas the nuclide transport properties of the near-field are well-defined, those of the host rock can vary considerably depending on the material (salt, clay or granite) and structure of the host rock. Different studies have identified ^{135}Cs, ^{79}Se, ^{99}Tc and ^{126}Sn as dominant dose contributors. ^{129}I is not contained in vitrified waste because it escapes during the reprocessing. However, the current sea disposal of iodine may no longer be practicable in advanced nuclear fuel cycles, and a special treatment of this nuclide will therefore also be an element of future waste management schemes.
- In the case of dry reprocessing, the unrecovered materials are processed into different waste forms which depend on the particular flow sheet. For the principal fuel cycle schemes with dry reprocessing, respective information will be given in Section 3.5. On the whole, these wastes are very stable; the associated long-term risk, however, is difficult to assess because detailed concepts for the ultimate storage of the wastes have not yet been developed.

Compared with the direct storage of spent fuel elements, the reprocessing of spent fuel has the advantages of reducing the actinide mass and the plutonium content of the HLW. In combination with vitrification, it minimises the risk of a clandestine recovery of fissile material; due to the low mobility of plutonium, it does, however, not significantly reduce the actinide part of the long-term risk of a repository. Drawbacks of the reprocessing are the extra investment in the reprocessing plant and the potential proliferation risk associated with the handling of the separated pure plutonium.

Judging the advantages of the reprocessing to be important and the recovered plutonium to be an asset rather than a risk, several countries have already embarked on a fuel cycle which involves the wet reprocessing of the spent fuel from LWR-UOX reactors, followed by the vitrification and final geologic disposal of the remaining fission products and minor actinides. In this conventional fuel cycle with vitrification (CFC-vitrification), the fate of the recovered plutonium is open; the plutonium will probably be recycled once and the spent MOX will be treated as HLW.

According to [22], a respective canister with vitrified waste contains 320 kg of glass, 48 kg of fission products, 3.5 kg of actinides and 4.5 kg of activation products. This means that the CFC-vitrification strategy reduces the actinide content of the HLW already to a level where the fission products are dominating the waste mass. Since the fission product inventories depend primarily on the number of fissions, i.e. the energy, released in the fuel, it is obvious that the HLW mass cannot be further reduced by moving from a CFC-vitrification to a transmutation strategy. Such a move, however, affects the actinide releases from the waste repository.

Since the repository behaviour for the CFC-vitrification strategy has been assessed in the framework of different national waste management projects, it is interesting to make a comparison with the plutonium burning and the double strata strategies which also involve vitrification. In the latter case, a direct comparison is possible, if the HLW from the dry reprocessing, which contributes

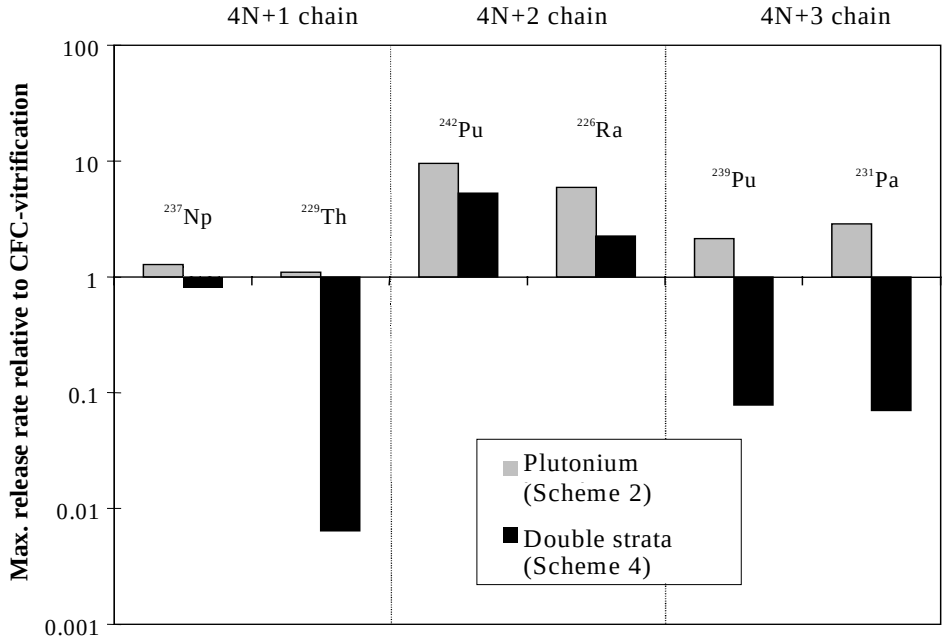
35% to the TRU going to waste (see Table 3.3), can be neglected. This is a reasonable approximation because the waste form bearing actinides from dry reprocessing is at least as stable as vitrified waste.

Taking into consideration that the geosphere and biosphere responses are site-specific and vary considerably between projects, it is advantageous to perform the comparison on the basis of the maximum release rates of potentially troublesome actinides from the repository near-field. Potentially troublesome actinides which can contribute to the annual individual dose beyond about one million years are ^{231}Pa, ^{237}Np and their respective daughter products ^{227}Ac and ^{229}Th, ^{226}Ra, a decay product of ^{238}Pu and ^{234}U, and the long-lived plutonium isotopes 239 and 242. Respective release rates were calculated for the plutonium burning, the double strata and a consistent CFC-vitrification strategy using the same near-field characteristics and nuclide properties as in the Kristallin-I safety assessment study [12].

The relative maximum release rates in Figure 3.13 confirm the expected strongly non-linear relationship between release rates and actinide concentrations in the glass. In particular, it can be seen that:

- Compared with the CFC-vitrification strategy, the plutonium burning strategy generally increases the maximum release rates (this is not surprising, since, in the reference case, most of the plutonium is recovered before vitrification).
- The addition of the P&T cycle results in a reduction of the maximum release rates for the more important, but not for all potentially troublesome actinides.
- Nuclides from the same decay chain can behave very differently (the maximum release rate for ^{237}Np, which has a very low solubility in the glass, is practically unchanged, whereas there is a large benefit for its radiologically more hazardous daughter product ^{229}Th).

Figure 3.13. **Maximum near-field release rates for fuel cycles with HLW vitrification**
(Near-field assumptions as in the Kristallin-I safety assessment study [12])



With regard to the long-term risk arising from the fission products, it is important to note that the fission product inventory depends primarily on the number of fissions, i.e. the energy, produced in the fuel, and that the fission product risk can, therefore, not be much influenced by the actinide transmutation strategy.

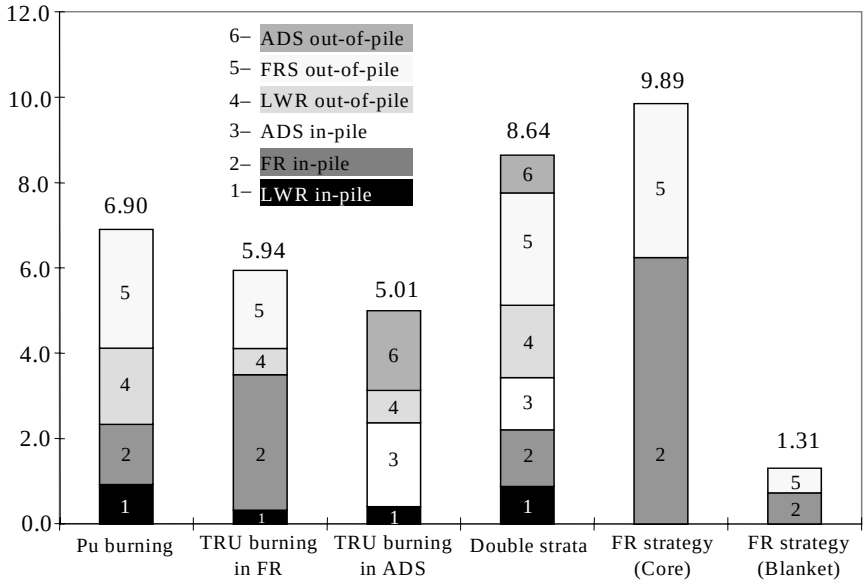
As already noted, the energy released in the fuel determines the fission product inventory and, in a reprocessing strategy, the mass of the high-level waste. Since the dependence of the cumulative fission product yields on the reactor- and fuel-type is small, the fission product release rates of a vitrified waste repository are practically proportional to the released fission energy. Neutron-spectrum dependent variations in the fission product composition and hence release rates can, however, arise from the in-situ transmutation of fission products. For example, capture of thermal neutrons in the precursor ^{135}Xe reduces the ^{135}Cs production in an LWR by as much as 70% [34].

3.2.6 Consequences for the fuel cycle

Whereas the principal transmutation schemes perform similarly with respect to the important goal of the TRU waste mass and radiotoxicity reduction, different requirements and consequences arise for the establishment of the fuel cycles. In this context, important parameters are the in-pile and out-of-pile TRU inventories, the throughput requirements for the fuel cycle facilities, and the decay heat and neutron source strength of the fuels. The TRU inventory has safety implications and plays a role in the shut-down phase of a nuclear energy strategy (see Chapter 2, Section 2.8), the fuel throughput determines the capacity of the fuel cycle facilities, the decay heat affects and limits the applicability of different reprocessing methods, and the neutron source strength has consequences for the shielding and the remote handling of materials.

Figure 3.14 shows that, among the transmutation schemes, the TRU burning in ADS scheme and the double strata scheme feature the lowest and highest total TRU inventories, respectively. This implies that the TRU burning in ADS strategy can respond more flexibly to unexpected changes in nuclear energy scenarios than other transmutation strategies.

Figure 3.14. **In-pile and out-of-pile TRU inventories**



The reprocessing and fuel fabrication requirements in terms of heavy metal throughput are compiled in Table 3.4. Regarding the reprocessing, the ADS-based transmutation schemes (TRU burning in ADS and double strata strategy) have the advantage of a modest demand for innovative (pyro-) reprocessing. Except for the FR strategy which is fully pyro-based, the schemes require a comparable wet reprocessing capacity, either PUREX or UREX. A similar situation exists on the side of the fuel fabrication, where the ADS-based transmutation schemes have the lowest demand for advanced fuel fabrication (metal and nitride fuel). This applies particularly to the double strata strategy which is, however, penalised by an increased MOX fuel demand for the plutonium burner.

Table 3.4. **Reprocessing and fuel fabrication requirements**

	Pu burning	**TRU burning in FR**	**TRU burning in ADS**	**Double strata strategy**	**FR strategy**
Reprocessing requirements (kgHM/TWhe)					
PUREX for LWR-UOX	1 583	–	–	1 501	–
PUREX for LWR-MOX	234.7	–	–	222.9	–
Adv. PUREX for FR-MOX	90.9	–	–	86.4	–
UREX	–	1 439	1 810	–	–
Pyro	–	247.3	77.5	39.3	2 122
Fuel fabrication requirements (kgHM/TWhe)					
LWR-UOX	1 669	1 517	1 909	1 583	–
LWR-MOX	247.4	–	–	2 34.5	–
FR-MOX	111.5	–	–	105.7	–
Ac-Zr	–	288.0	103.2	–	2 233[1]
AcN-ZrN	–	–	–	46.2	–

1. Including 1 438 kg/TWhe of uranium for the blankets.

Tables 3.5 and 3.6 provide information on the activity, decay heat and neutron source strength of the fuels after cooling (cooling times as indicated in Figures 3.1 to 3.6) and after fuel fabrication, which is assumed to take place immediately after the reprocessing. In general, the decay heat is dominated by the actinides and lies beyond the operating limits of existing fuel cycle facilities.

Experience with pilot plants in the UK and France shows that the PUREX-type aqueous reprocessing can be considered as valid for the FR-MOX fuel of schemes 2 and 4. Reprocessing of this fuel within short cooling times and with the required high recovery yield of 99.9%, however, will require measures to improve the plutonium dissolution yield and modifications of the PUREX flowsheet. An interim solution for schemes 2 and 4 could be the blending of the irradiated LWR-MOX and FR-MOX fuels before reprocessing. Such blending would reduce the decay heat of the FR-MOX fuel by a factor of about two.

The decay heat of the ADS fuels arising in schemes 3b and 4 lies well beyond the limit for which the radiation stability of the organic extractant in the aqueous process can be guaranteed. For these and all other systems with fully closed fuel cycles, the pyrochemical reprocessing is the appropriate reprocessing method because it circumvents unnecessary separation processes (only fission products are extracted) and can handle highly active product streams without major radiation degradation. Figure 3.15 shows that the decay heat decreases only slowly with increasing cooling time and that very long cooling times – resulting in very large out-of-pile fuel inventories – would be required to mitigate the problem and facilitate the use of aqueous reprocessing methods.

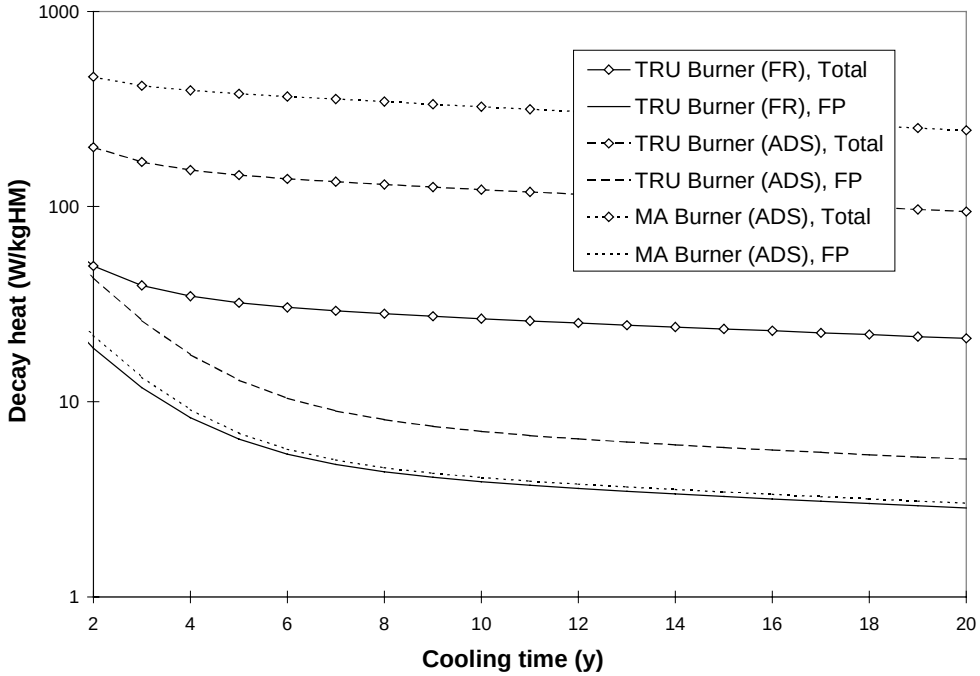
Table 3.5. **Activity, decay heat and neutron source strength of fuel after cooling**

	LWR-UOX	**LWR-MOX**	**Pu burner (FR)**	**TRU burner (FR)**	**TRU burner (ADS)**	**MA burner (ADS)**	**Fast reactor**
Fuel cooling time (a)	4	7	7	2	2	2	2
Activity (10^{12} Bq/kgHM)							
Actinides and FPs	36.24	50.30	157.0	232.4	691.7	725.4	160.4
FPs	30.15	17.26	62.38	155.3	342.0	183.7	131.7
Decay heat (W/kgHM)							
Actinides and FPs	3.48	6.31	21.77	46.00	192.6	455.1	18.56
α	0.52	4.86	16.76	30.51	156.4	435.4	5.63
β	1.63	0.66	2.32	11.38	27.12	14.83	9.34
γ	1.34	0.79	2.67	4.09	8.96	4.42	3.58
FPs	2.96	1.41	4.86	15.34	35.50	18.02	12.88
β	1.62	0.63	2.21	11.28	26.64	13.83	9.31
γ	1.34	0.79	2.65	4.07	8.88	4.19	3.58
Neutron source strength (10^6 n/s kgHM)							
Total	0.97	10.93	39.28	86.08	649.1	1 812	9.76
Spontaneous fission	0.95	10.77	38.72	85.02	643.6	1 797	9.57
(α,n)	0.02	0.16	0.56	1.06	5.46	15.22	0.19

Table 3.6. **Activity, decay heat and neutron source strength of fuel at fabrication**

	LWR-UOX	**LWR-MOX**	**Pu burner (FR)**	**TRU burner (FR)**	**TRU burner (ADS)**	**MA burner (ADS)**	**Fast reactor**
Activity (10^{12} Bq/kgHM)							
Actinides	$1.53 \cdot 10^{-5}$	38.28	148.1	111.5	470.6	598.2	28.89
Decay heat (W/kgHM)							
Actinides	$1.1 \cdot 10^{-5}$	1.94	9.64	33.79	168.1	489.3	5.79
α	$1.1 \cdot 10^{-5}$	1.91	9.49	33.60	167.2	487.4	5.74
β	$4.4 \cdot 10^{-8}$	0.03	0.13	0.14	0.59	1.14	0.03
γ	$9.0 \cdot 10^{-8}$	$1.0 \cdot 10^{-3}$	$2.6 \cdot 10^{-3}$	0.02	0.09	0.30	$4.3 \cdot 10^{-3}$
Neutron source strength (10^6 n/s kgHM)							
Total	$2.07 \cdot 10^{-5}$	0.10	0.66	92.05	669.9	1992	9.76
Spontaneous fission	$2.06 \cdot 10^{-5}$	0.04	0.37	90.89	664.0	1975	9.57
(α,n)	$1.32 \cdot 10^{-7}$	0.06	0.29	1.16	5.82	17.04	0.19

Figure 3.15. **Evolution of the decay heat of the fuel**



Due to the strong source of spontaneous neutrons, the fuel fabrication will have to adapt its handling technology to reduce the radiation doses to the workers in the plant and during the transport of the fuel assemblies. The increased requirements for shielding during transport also speak for the pyrochemical reprocessing method which is applicable in small facilities in the immediate vicinity of the reactors, whereas the aqueous process favours large centralised facilities which imply shipment of fuel over long distances.

In summary, it appears that all transmutation strategies with fully closed fuel cycles will have to rely on pyrochemical reprocessing. The ADS-based schemes have the advantage of modest throughput requirements, but rely on the development of the technology for fuels with very high decay heats.

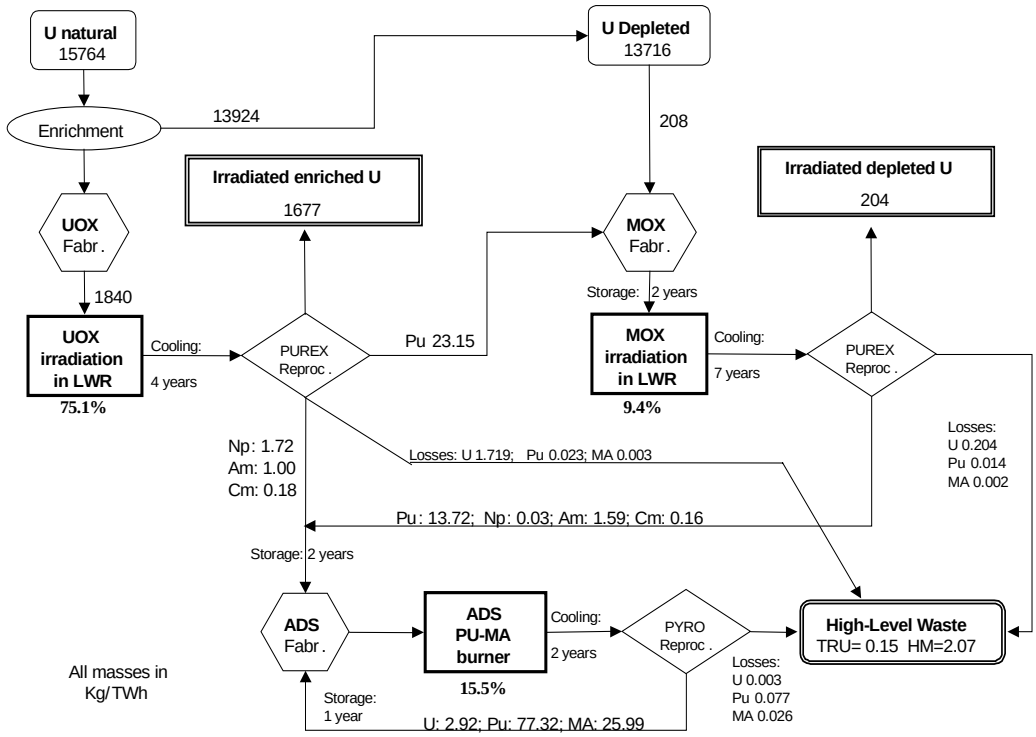
3.3 Other fuel cycle schemes

3.3.1 TRU burning with preceding MOX recycling

A country which has already committed itself to the conventional reprocessing of LWR spent fuel with recovery of the plutonium and vitrification of the HLW, or plans to introduce plutonium recycling as an intermediate step before actinide burners become commercially available, could be interested in a TRU burning strategy with an LWR-MOX stage between the normal LWR and the TRU burner. Figure 3.16 shows the ADS variant of this strategy which, in the following, will be referred to as scheme 3c. The strategy represents a mix between the evolutionary and the innovative approach to transmutation and has the advantage that it does not depend on the installation of MOX-fuelled fast plutonium burners and requires a smaller number of fast-spectrum systems than any of the afore-mentioned transmutation schemes. However, due to the presence of a plutonium separation stage, the strategy is not a pure co-processing strategy as in the other TRU burning cases.

A study of scheme 3c [35] was carried out in parallel with that of the principal fuel cycle schemes using consistent assumptions for the reactor and the fuel cycle parameters (fuel burn-up, reprocessing losses, fuel cooling and storage time). In particular, the TRU burner in Figure 3.16 has the same core characteristics (power, coolant- and fuel-type) as that in scheme 3b. The additional LWR-MOX reactors, however, imply a change in the actinide composition of the ADS fuel.

Figure 3.16. **TRU burning with preceding MOX recycling (scheme 3c)**



More specifically, the effect of the plutonium burning in the LWR-MOX reactors is to reduce the fissile and correspondingly enhance the fertile MA component of the ADS fuel. This results in a reduced reactivity drop of the core and hence mitigates the inherently large reactivity drop of TRU burner cores. A reduced burn-up reactivity drop means either a reduced number of fuel batches or a smaller accelerator current at the end of the reactor cycle and hence translates immediately into an economic advantage. The reduced requirements for reactivity compensation represent also a safety advantage.

The actinide waste production for scheme 3c is given in Table 3.7. The values in this table can be compared directly with the actinide waste production for the principal fuel cycle schemes in Table 3.3. It can be seen that the TRU mass in the waste is dominated by the ADS as in the other TRU burning cases; the TRU waste mass is slightly reduced compared with scheme 3b, but is still larger than that of scheme 3a. As expected, the transuranics in the waste have a somewhat lower plutonium content, but this does not change the radiotoxicity of the waste significantly. The general conclusion that all transmutation strategies with closed fuel cycles can achieve a similar radiotoxicity reduction is herewith confirmed.

Table 3.7. **Actinide waste production for scheme 3c (g/TWhe)**

	LWR-UOX	LWR-MOX	ADS	Total
^{238}Pu	0.75	0.70	9.33	10.78
^{239}Pu	12.71	4.28	8.81	25.79
^{240}Pu	5.97	5.15	35.53	46.65
^{241}Pu	2.42	1.95	6.70	11.06
^{242}Pu	1.32	1.66	17.03	20.01
^{237}Np	1.73	0.03	2.98	4.74
^{241}Am	0.61	1.09	5.99	7.70
^{242m}Am	0.00	0.01	0.48	0.49
^{243}Am	0.38	0.49	6.58	7.46
^{242}Cm	0.00	0.00	0.04	0.04
^{243}Cm	0.00	0.00	0.08	0.08
^{244}Cm	0.16	0.14	6.12	6.42
^{245}Cm	0.01	0.01	2.05	2.08
Pu	23.16	13.74	77.40	114.29
Np	1.73	0.03	2.98	4.74
Am	1.00	1.59	13.05	15.65
Cm	0.18	0.16	9.98	10.31
TRU	26.06	15.52	103.41	144.99

3.3.2 Heterogeneous recycling of americium and curium

As discussed in Section 3.2.5, the homogeneous recycling of minor actinides in the fuel implies very hot fuels which are also strong neutron emitters and consequently necessitates considerable investments in improved and new reprocessing technologies as well as remote handling and shielding of materials. To avoid the contamination of the fuel cycle with minor actinides, it has been proposed to separate americium and curium from the spent fuel, fabricate them into separate targets with a neutron-inert support material like $MgAl_3O_4$ (spinel), "incinerate" the targets, and dispose of the spent targets without further reprocessing. The targets could be placed in special fast reactor subassemblies containing a neutron moderator such as $^{11}B_4C$, ZrH_2 or CaH_2 which enhances fission [36].

For neptunium, the homogeneous recycling is the generally preferred method, although a large fraction of the neptunium in the fuel is converted to ^{238}Pu, an alpha emitter with a half-life of only 88 years. The resulting heat load and neutron emission from (α,n) reactions could still have economic consequences for the fabrication, transportation and storage of the fuel.

The heterogeneous recycling concept for americium and curium could be implemented either in a pure fast reactor scheme (scheme H1), or a two-component scheme which consists of a mix of UOX-fuelled LWRs and fast reactors (scheme H2). Actinide mass balances for both schemes have been calculated by CEA under the following assumptions [37]:

- The fast reactor is the 1 450 MWe European Fast Reactor (EFR) with a MOX core which is optimised for an average fuel burn-up of 140 MWd/tHM [38]. In scheme H1, the reactor is self-sustaining in plutonium thanks to thin uranium blankets; in scheme H2 the reactor operates as a plutonium burner without blankets. The average plutonium content of the fuel is

about 20% in the former and about 23% in the latter case, meaning that it is close to that of a normal MOX-fuelled fast reactor.

- The LWR in scheme H2 is the 1 450 MWe European Pressurised Reactor (EPR) with an average burn-up of 60 GWd/tHM [39]. The plutonium and minor actinide mass balance for an optimised EFR core with target subassemblies restricts the fraction of LWRs in the two-component scheme to about 44%, meaning that this scheme features the least favourable support ratio of all investigated schemes.

In the case of scheme H1, it was found that an EFR core which consists of 42 target and 346 fissile sub-assemblies allows to incinerate 90% of the minor actinides in the targets. The necessary residence time of the targets is about 10 years to be compared with a 6-year residence time for the fissile subassemblies. In the case of scheme H2, it was found necessary to complement the 42 target subassemblies in the core with a complete ring of 78 target subassemblies at the core periphery, i.e. in the place of the radial blanket region of the self-sustaining core configuration.

It is worth noticing that the moderator in the target subassemblies has a beneficial effect on the fuel Doppler and the coolant void reactivity effect (the former increases and the latter is reduced). Moreover, it has been ascertained that, in spite of the long residence time of the targets, the limit of 200 dpa for the radiation damage to the structural material is not exceeded.

The two-component scheme is illustrated in Figure 3.17. It can be seen that the neptunium from the spent fuels is fabricated into new $(U,Pu,Np)O_2$ fuel for the EFR, and the americium and curium from the spent fuels is separated and fabricated into targets which are disposed of after irradiation. The assumptions for the recovery yield (99.9% for Pu, Np, Am and Cm) and the fuel cooling and storage time before reprocessing and after fabrication are consistent with the respective assumptions for the principal fuel cycle schemes.

Table 3.8 gives the resulting actinide waste production under equilibrium conditions in units which allow a direct comparison with the results in Tables 3.3 and 3.7. In particular, it can be seen that scheme H2 allows to reduce the TRU mass in the waste by a *factor of about 60* compared with the LWR once-through reference case. The analyses performed by CEA show that the TRU mass reduction factor is nearly the same for scheme H1, and drops to about 40, if americium only is separated and incinerated in the targets while curium is directly rejected to waste.

A TRU waste mass reduction factor of 60 may already be attractive, but is modest compared with a factor of at least 175 achieved by the principal fuel cycle schemes (see Section 3.2.3). However, as pointed out in [40], this limitation of the heterogeneous recycling concept should be balanced against the advantages that:

- The fabrication, irradiation and final disposal of the highly active americium and curium targets is disconnected from the uranium-plutonium fuel cycle which is that of a normal fast reactor.
- The advanced PUREX process with americium and curium separation is valid for the reprocessing of the fuel and the development of the pyrochemical reprocessing method and ADS technology is not required.
- By abstaining from the reprocessing of the targets, the minor actinide mass flows can be reduced significantly (40% for Am, factor 3 for Cm) with a corresponding cost benefit compared with the homogeneous recycling concepts based on fully closed fuel cycles.
- At the initial stage of development, americium only could be loaded into the targets.

- The remaining technological uncertainties, which relate mainly to the behaviour of the targets, are small and alternatives are available (for example, the coated particle concept [41] can be considered as an alternative to the inert matrices).

In summary, it appears that heterogeneous transmutation systems as described in this section are less performant than systems with fully closed fuel cycles. On the other hand, the heterogeneous systems have the advantage that they could be deployed earlier than other systems because they are mostly based on existing technology.

Figure 3.17. **Heterogeneous recycling of americium and curium (scheme H2)**

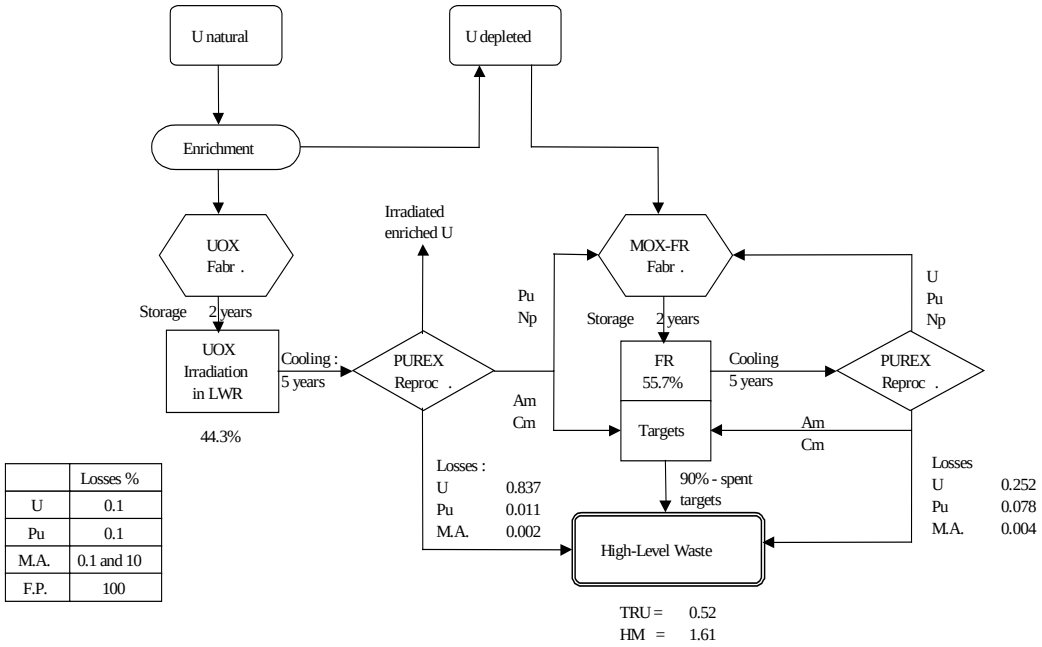


Table 3.8. **Actinide waste production for heterogeneous scheme H2 (g/TWhe)**

	LWR-UOX	FR	Targets	Total
U	837.5	252.5	2.5	1 092.5
Pu	11.4	78.0	150.0	239.4
Np	0.8	1.2	–	2.0
Am	0.7	2.8	37.5	40.0
Cm	0.1	0.3	235.0	235.4
TRU	13.0	82.3	422.5	516.8
HM	850.5	334.8	425.0	1 609.3

3.4 Fuel cycle issues and challenges

The previous Sections 3.2 and 3.3 highlighted the fuel cycle schemes under consideration and indicated in Section 3.2.6 the consequences for the fuel cycle. The following sections will address these consequences in some more detail and will highlight the technological challenges faced by the

fuel cycle operations in order to achieve the performance as supposed in the previous fuel cycle schemes.

Four topics will be highlighted in that respect:

- Fuel and target fabrication.
- Reprocessing.
- Secondary waste arising.
- The management of depleted and reprocessed irradiated uranium.

3.4.1 Fuel and target fabrication and behaviour

Several studies have been undertaken in the recent past addressing the different aspects of fuel and target fabrication and behaviour. The reports by JAERI, the European Commission as well as the Roadmap-reports on ATW and by the European Technical Working Group on ADS include comprehensive reviews of fuel/target technologies for application in transmutation systems, and the interested reader is referred to those documents for details [42,8,9]. In the latter report, the focus is limited to non-fertile (i.e. not uranium-bearing) fuels and to transmutation targets containing ^{99}Tc and ^{129}I. Here, the consideration is somewhat broader, with the various fuel cycle schemes including fertile (schemes 2, 3a, 5) and non-fertile (schemes 3b and 4) fuels, plutonium recycle only (scheme 2), and recycling with co-processing (schemes 3a, 3b, 5) or separation (scheme 4) of plutonium and minor actinides.

3.4.1.1 Oxide fuels and targets.

The schemes under consideration use uranium-bearing oxide fuels and $(Am+Cm)O_2$ inert matrix targets. Considerable experience exists with the performance of mixed-oxide (MOX) fuels to high burn-up levels in both thermal and fast reactor applications, with plutonium fractions ranging from 4 to as high as 30 weight percent. The design of MOX fuels for any of the pertinent fuel cycle schemes of this report is therefore a well-founded technology. On the other hand, fabrication of MOX fuels containing recycled plutonium (i.e. with large percentages of ^{240}Pu and higher Pu isotopes) requires a high degree of automation and remote operation of fuel fabrication processes that is not currently in common practice. Innovations in MOX fuel fabrication technology will be necessary to ensure worker protection and the economic viability of the fuel cycle. This may require a departure from the standard powder synthesis, cold pressing and sintering process.

Very little experience exists for fertile oxide fuels containing representative quantities of the minor actinides neptunium, americium and curium. The French SUPERFACT experiment comprised eight fuel pins with $(U,Pu)O_2$-NpO_2/AmO_2 and UO_2-NpO_2/AmO_2 fuel pellets, irradiated to modest burn-up (4.5-6.5 atom percent) in the Phénix reactor. Post-irradiation examination showed nominal performance of the fuel pins, with evidence of the onset of fuel-cladding mechanical interaction due to fuel swelling in those pins containing higher minor actinide fractions. Greater helium production, well in excess of fission gas generation, was observed in the americium-containing fuel and attributed to the high specific alpha activity of the americium daughters ^{242}Cm and ^{238}Pu. At the burn-up levels thought to be needed for efficient performance of a minor actinide transmutation system, it will be necessary to provide accommodation for internal pressurisation and fuel swelling by reducing fuel smear density and increasing plenum volume. A trade-off study between desired core performance and fuel burn-up is clearly needed. Also of concern is the fabrication of minor actinide-bearing oxide fuels.

As curium isotopes build up with repeated recycling, the limitations on fuel handling and facility operation will be challenged. Alpha decay and neutron emission from curium isotopes may impose a need for chemical separation of curium from the other transuranics and storage of the curium until some of its isotopes decay to lower transuranics. This issue requires further study, as the storage of curium poses criticality and heat removal problems.

Non-fertile TRU oxide fuels (targets) use an inert matrix material as a diluent to maintain specific heat generation levels within the proper range. Desired properties of the matrix material are: (1) chemical compatibility with the reactor coolant, (2) chemical and physical compatibility with the fuel, (3) resistance to radiation damage, and (4) a low neutron absorption cross-section. Both metal and ceramic matrix materials have been considered for this application. Ceramic materials include zirconia (ZrO_2, stabilised in the fluorite cubic structure by additions of CaO, Y_2O_3 or MgO), magnesia, and spinels such as $MgAl_2O_4$. A dispersion of TRU oxides in zirconium or in a refractory metal such as molybdenum offers the advantage of a low fuel operating temperature, but provisions must be made to prevent a chemical interaction between the fuel and matrix (e.g. $PuO_2 + Zr \rightarrow ZrO_2 + Pu$) that could lead to undesirable redistribution of the fissile constituents. The ceramic matrix must be stable under irradiation and capable of accommodating the very large quantities of helium produced in minor actinide-bearing fuels.

Fabrication processes for inert matrix oxide fuels include dry powder operations and wet processes such as co-precipitation or sol-gel reactions. There has been some experience with infiltration of a nitrate solution of transuranics into inert ceramic particles, followed by calcinations/denitration and sintering. In all cases, particular care must be taken to avoid the loss of volatile TRU oxides.

3.4.1.2 Metal fuels

Considerable experience with uranium alloy metal fuel has been gained in the course of operations with DFR (U-Mo) and EBR-II (U-Fs[34], U-Zr) over a number of years. A limited number of U-Pu-Zr fuel elements were irradiated in EBR-II to reasonable high peak burn-up levels, over 20 atom percent. The 33-cm long EBR-II fuel slugs were fabricated by injection casting the molten fuel alloy into quartz tubes. After solidification, the quartz molds were broken and separated from the fuel. Because the fuel rods were designed for about 75% smear density, with liquid sodium serving as a thermal bond between the fuel and cladding, the fuel slugs could be used in the as-cast condition with little concern for minute dimensional variations. In only one experiment was there made an attempt to incorporate minor actinides in the injection casting process, with the result that most of the americium was lost by vaporisation. It may be possible to suppress such losses by over-pressurisation of the casting system, but this remains a major limitation to the use of minor actinide-bearing metal alloy fuel. Powder metallurgy methods may provide the means for avoiding americium losses. The performance of metallic fuel is well-established; it offers many safety advantages provided that steady-state fuel temperatures are kept below the temperature range for the formation of a low-melting eutectic composition with iron that is present in the cladding alloy.

3.4.1.3 Nitride fuels

It is generally accepted that the use of nitride fuels in accordance with contemporary environmental standards would require the use of nitrogen highly enriched in the isotope ^{15}N, to avoid the production of ^{14}C from (n,p) reactions with the more common ^{14}N isotope. Nitride fuels offer a number of advantages: (1) high thermal conductivity, (2) extensive mutual solubilities of the actinide

34. Fs: “fissium”, the designation given to a collection of transition metal fission products remaining in the uranium recovered in the original EBR-II melt-refining process.

nitrides, (3) a single common valence state of the actinide nitrides, and (4) very low (and sometimes negative) values for void worth. No nitride fuel performance results are available at high burn-up levels, with past experience limited to burn-ups in the range of 6.5-8 atom%. Nitride fuels can be operated at low temperatures, and this gives some reason for optimism in their ability to reach the high burn-ups required for efficient transmutation systems. There is a concern that, under accident conditions in which the nitrogen over-pressure in a fuel pin is lost, the nitride can decompose rapidly, leading to extensive core damage. This concern should be ameliorated in an inert-matrix fuel in which the matrix is ZrN or Zr (this applies, e.g. to the fuel of the minor actinide burner in scheme 4).

Fabrication processes for nitride fuels commonly involve powder pressing and sintering, with the powders produced either by direct nitridation of metals or by carbothermic reduction of oxide powders followed by nitriding. Losses of americium by volatilisation of the nitride are a problem during high-temperature sintering of minor actinide-bearing fuels.

3.4.1.4 Other fuel types

Carbide fuels and coated-particle fuels are also candidates for application in transmutation systems. Carbide fuels may be of less interest because they do not offer substantial advantages over the other fuel types while presenting some specific complications such as the formation of complex phases and the potential for significant chemical and mechanical interactions with the fuel cladding. Coated-particle fuels, on the other hand, may be favoured in a nuclear system in which a key component is a high-temperature gas-cooled converter reactor, by providing a commonality of fuel cycle processes. These fuels are known to be capable of very high burn-ups and, by virtue of high mechanical integrity coatings, are able to retain the large amounts of helium and fission products generated.

3.4.1.5 Long-lived fission product targets

Important studies of the transmutation of ^{99}Tc and ^{129}I have been carried out within the framework of the EFTTRA program [43]. The general consensus arising from this and other studies is that ^{99}Tc is best transmuted to stable ^{100}Ru in the metal form, while the preferred form for ^{129}I is as a metal iodide. Sodium iodide may become the target of choice due to the nature of the process whereby it is recovered from the dissolver solution during aqueous reprocessing. Irradiation experiments have not been carried to high fractional transmutation levels, but results indicated that the metallic technetium targets are well behaved. The technetium experiments also have shown the importance of target geometry, for exploitation of neutron absorption at resonant energy levels. The behaviour of sodium iodide targets (or silver iodide or lead iodide) may not be so benign, because ^{129}I is transmuted to stable ^{130}Xe, with attendant build-up of gas pressure internal to the target capsule. Meanwhile, as ^{129}I is transmuted, the remaining cations form free metal which can interact with the capsule material, potentially leading to capsule failure through corrosion or formation of low-melting compounds. Capsule material composition is a factor that is often overlooked in transmutation studies. Because the objective of transmutation is a substantial reduction in the radiotoxicity of nuclear wastes to be disposed, it is important that the process not generate more radiotoxic waste than is destroyed. The use of common stainless steel capsules, for example, can lead to the generation of activation products that are more radiotoxic than the technetium or iodine originally present if irradiation is carried out in a thermal flux.

Clearly, long-term irradiation tests and materials compatibility studies are needed for successful implementation of a regime for the transmutation of long-lived fission products.

3.4.2 Reprocessing techniques

3.4.2.1 Aqueous reprocessing

Introduction to aqueous reprocessing

Aqueous reprocessing is based on the use of the PUREX process, which has been since the mid-1950s the industrial technique to separate uranium and plutonium from spent fuel. PUREX is a solvent extraction process that takes advantage of the multi-valent nature of the actinides. When present in nitric acid solution, uranium and plutonium exist in the U(VI) and Pu(IV) states, which are readily extracted into an organic molecule such as tributylphosphate (TBP, typically mixed with a kerosene or dodecane diluent) that is brought into contact with the acid solution. Americium, neptunium and curium are not extracted, and remain in the aqueous phase. Plutonium can then be reduced to the Pu(III) state and stripped out of the organic phase, accomplishing the desired separate recovery of plutonium and uranium. The minor transuranic elements in spent fuel (Np,Am,Cm) typically have long half-lives and/or high toxicity, and are commonly mixed with the fission products and treated as waste in a vitrification process following PUREX reprocessing. The discharge burn-up of LWR fuel has been increased over the years from 33 to 50 GWd/t, with a resulting increase of the decay heat in spent fuel assemblies and the occurrence of higher concentrations of trans-plutonium elements.

Fast-reactor based plutonium-burning technologies, such as that entailed in the French CAPRA program, require an increase in the plutonium content of fuels to as much as 50 weight percent and an increase in goal burn-ups to levels as high as 210 GWd/tHM or more. This makes aqueous reprocessing more difficult, because of the low solubility of plutonium and the radiation damage to the organic extractant (tributyl phosphate). Industrial “pilot” scale work (at research sites in Dounreay, Scotland and Marcoule, France) has shown that with the introduction of advanced technologies (such as pin-choppers and centrifugal contactors), aqueous reprocessing can be considered as valid for fast reactor and future ADS fuel if the decay heat can be mitigated by longer cooling periods or by dilution with LWR fuel.

Special chopping or shearing systems have been developed for fast reactor fuel (e.g. for the reprocessing of FFTF fuel, although the process was never used) in order to replace the single or multiple pin shearing process with a more economical complete bundle shearing approach. Laser cutting of PFR ducts was employed at the Dounreay reprocessing facility.

Many proposals for P&T technology rely on the aqueous reprocessing of spent fuel as a preliminary step preceding minor actinide partitioning. In the case of pyrometallurgical processing of TRU’s (the US ATW Roadmap project), a mechanical head-end and an aqueous processing step (called UREX) for the prior removal of uranium, as the main fertile element, precedes the sequence of pyrometallurgical separation steps. Spent fuel arising from a composite reactor park (70% LWR-UOX, 10% LWR-MOX, 20% FR-MOX) must be reprocessed in order to facilitate plutonium recycle and the stabilisation of the transuranic inventory; aqueous reprocessing methods are favoured in such scenarios. According to the Pu burning scheme (scheme 2, see Figure 3.2), the spent fuel generation rate from a 100 GWe composite reactor system leads to the outputs shown in Table 3.9.

Table 3.9. **Spent fuel discharge rates for components of scheme 2**

Fuel type	Burn-up GWd/tHM	Electrical output GWe	Spent fuel discharge tHM/y
LWR-UOX	50	69.7	1 387
LWR-MOX	50	10.3	206
FR-MOX	185	20.0	80

The reprocessing operations supporting such a system are presumed to be carried out 4 to 7 years after discharge of spent fuel from the reactors. The residual decay heats for these types of fuel are summarised in Table 3.10 (See also Tables 3.5 and 3.6).

Table 3.10. **Residual decay heats for scheme 2 fuel types**

Fuel type (cooling time, y)	**Burn-up, GWd/tHM**	**Total decay heat, kW/tHM**	**Fission products, kW/tHM**	**Actinides, kW/tHM**
LWR-UOX (4)	50	3.48	2.96	0.52
LWR-MOX (7)	50	6.31	1.41	4.90
FR-MOX (7)	185	21.77	4.86	16.91

In conventional aqueous reprocessing of LWR-UOX fuel, the fuel elements are chopped into small pieces and transferred to the dissolver. Large LWR fuel elements contain between 450 and 500 kg of UO_2 or MOX. For simplicity, we will consider two fuel elements per tHM, which are dissolved in 5 m^3 HNO_3. Conceptually we will consider the ratio 1:1.8 between the aqueous HNO_3 feed solution and the organic (30% TBP-diluent) extractant phase. The throughput of a typical plant is about 200kg HM chopped spent fuel per hour or 4.8 tHM/day (800-960 tHM/year) depending on the plant load factor. In this case, we assume the contact time between the aqueous and organic phase to be 1 hour. The radiation damage in the extractant is proportional to the contact time of the highly radioactive aqueous feed solution and the TBP-solvent mixture.

Radiation damage to solvent

The radioactive decay energy dissipated by the feed solution into the extraction mixture as a radiation dose D is calculated as follows:

$$D = 0.35 \text{ kW}/4.8 \text{ m}^3 \cdot \text{hour} = 0.073 \text{Wh/l}$$

Using this expression and the data given in the previous tables, the following doses are calculated (Table 3.11):

Table 3.11. **Radiation damage to solvent in different fuel cycle schemes**

Fuel type	**Dose emitted by aqueous phase (Wh/l)**	**Dose received by solvent (Wh/l)**
LWR-UOX	0.57	0.20
LWR-MOX	1.36	0.48
FR-MOX, 185 GWd/t	6.72	2.40

The main contribution to the radiation dose comes from the alpha decay of the actinides, which is very damaging to organic molecules. For LWR-MOX the alpha radiation represents 60% of the total radioactivity and for the FR-MOX fuel it amounts to 80%. In order to keep the radiation damage to the solvent to the safe limit of 0.1 Wh/l, the residence time for FR-MOX has to be reduced from the conceptual residence time of one hour to 2 or 3 minutes, which cannot be achieved with pulse columns. The use of centrifugal contactors in the first extraction cycle must be investigated as to its feasibility in current reprocessing facilities.

Several other possibilities are open to cope with very high burn-up fuel: (1) dilution of FR-MOX with LWR-UOX and LWR-MOX; (2) special head-end for FR-MOX; (3) installation of fast contactors throughout the reprocessing facility; and (4) very long term cooling.

- *Dilution of FR-MOX with LWR-MOX and LWR-UOX*. In a complex reactor park with 70% LWR-UOX, 10% LWR-MOX and 20% FR-MOX, the bulk mass of discharged spent fuel is given in Table 3.9. An equilibrium flowsheet of an advanced reprocessing plant could be established on the base of a homogeneous throughput of all the fuel types according to their discharge fractions. This option is in any case possible for the mixture of 7 tHM LWR-UOX and 1 tHM LWR-MOX. The total Pu inventory (without burn-up credit) would increase, but the radiation dose to the TBP would only increase marginally. In the case of a joint treatment of all fuel, including FR-MOX, the Pu inventory would drastically increase (by a factor 4.8) and the overall radiation exposure of the solvent would double, with peaks of more than a factor of 10 when FR-MOX is extracted.

- Some 100 tHM have been processed in reprocessing pilot plants after various cooling times and a representative campaign of FR-MOX reprocessing has taken place in the COGEMA UP2-400 plant at La Hague [44]. However, the technology to handle FR fuel in the head-end was different from that of LWR fuel assemblies, due to design differences and to higher fissile content in the irradiated FR fuel. The reprocessing of industrial quantities in a conventional plant will have an impact on the design philosophy of the PUREX extraction (pulse) columns, the slab-tanks and the criticality control devices. These issues are common for conventional PUREX reprocessing and for the newly proposed UREX process (should it be applied to fuel other than LWR spent fuel).

- *Special head-end for FR-MOX*. In order to improve the Pu dissolution yield and to avoid solvent radiation peaks during the extraction, a separate dissolver dedicated to FR-MOX treatment could be installed and connected to the main dissolver by a metering system. By connecting the dedicated FR-fuel dissolver to the main LWR dissolver a constant radiation level can be kept throughout the process campaign. The second dissolver could also serve as a "residue dissolver" by making use of highly oxidizing compounds (e.g. electrochemically-generated Ag(II)) to dissolve the insoluble fraction of the initial Pu inventory (2). Under increased radiolysis of TBP the fission product decontamination factors will decrease, the plutonium losses will increase, and the production of secondary waste (dibutyl phosphate, Na_2CO_3, …) will also increase.

- *Installation of fast contactors*. By replacing the pulse columns with fast centrifugal contactors, a gain of a factor three or more, depending on the number of stages and the scheduled contact time per stage, can be expected on the radiolysis of TBP and consequently on the feasibility of using aqueous reprocessing for high-burn-up FR-MOX fuel. Implementation of such a technology requires the complete refurbishment of an existing reprocessing plant. The importance of fast contactors in the first extraction stage warrants additional full-scale hot tests in order to establish whether the design of these components is sufficiently robust that they can be serviced and maintained in a very hostile radioactive environment.

- *Very long cooling times*. The radioactivity concentration in spent nuclear fuel decreases with the mean half-life of the fission products and with the half-life of the "short lived" actinides. The fission product activity decreases with a factor of 2 when delaying the reprocessing from 3 to 7 years. However, in the case of the actinide contribution to radioactivity, which is essentially determined by ^{244}Cm and ^{238}Pu (with half lives of 18 and 87 years, respectively),

the delays have to be much longer to be effective (see Figure 3.15). A long-term storage (~100 years) of separated ^{244}Cm with the formation of ^{240}Pu, has been considered as a strategy for Cm management. The ^{238}Pu contamination of separated plutonium from LWR-MOX and FR-MOX is an issue that must be examined in the framework of Pu purification and fuel fabrication operations.

Recovery of minor actinides

Recovery of minor actinides: neptunium

Although the conventional PUREX solvent extraction process is considered to be a method for extraction of uranium and plutonium from spent fuel, with the remaining actinides and fission products being sent to the waste stream, it is possible to recover neptunium from the dissolver solution by minor modifications to the PUREX flowsheet. Neptunium in the Np(IV) or Np(VI) valence states is reasonably extractable with TBP, while in the Np(V) state it is essentially inextractable. Under process conditions in which plutonium is placed in its extractable Pu(IV) state, neptunium tends to reside in the inextractable Np(V) state. Reagents such as tetravalent uranium, ferrous sulfamate or hydroxylamine can be used to reduce neptunium to the Np(IV) state; nitrite ion or pentavalent vanadium can be used to oxidise Np to the Np(VI) state. Neptunium then is co-extracted with either uranium or plutonium and must be subsequently separated by re-oxidizing it to Np(V) and acid stripping.

Recovery of minor actinides: americium and curium

In the PUREX nitric acid dissolver solution, americium and curium will reside in the +3 valence state, as will the lanthanide elements. The lanthanide fission products comprise about 30% of the total fission product mass in ten-year cooled spent LWR fuel, and their mass is more than ten times that of americium and curium combined. Neither Am(III) nor Cm(III) are extractable with TBP in the PUREX process, and in normal commercial practice there is no need for their recovery. But in a transmutation system that is dedicated to the near-total elimination of the highly radiotoxic transuranic elements, it is necessary to extract these elements. Modification of the valence states of Am and Cm in the mainstream PUREX process through redox reactions adds process complications and could affect other extractions, so the current thinking is to recover these constituents from the PUREX high-active raffinate before that stream is sent to waste processing. The lanthanide fission products complicate the recovery of americium and curium, and a number of processes have been developed for this very complex problem. Because the acidity of the raffinate is rather high, ≥2M in HNO_3, and the radiation level is significant, it has proven challenging to extract americium and curium separately in the presence of a large mass of +3 lanthanides [Ln(III)]. Therefore, initial efforts were directed toward co-extraction of Am and Cm and their subsequent separation. The compromise in such systems is the attendant substantial increase in high-level liquid waste generation.

Co-extraction of americium and curium

Several processes have been developed for the co-extraction of americium and curium from a raffinate solution containing lanthanides and other fission products. The first was the TRUEX process [45], which employs as extractant CMPO [n-octyl(phenyl)-N,N-diisobutyl carbomoylmethylphosphine oxide] in TBP. CMPO is a powerful extractant with high affinity for +3 actinides [An(III)] at high acidities, but it does not discriminate between An(III) and Ln(III). Other processes that feature co-extraction of actinides and lanthanides are TALSPEAK, DIDPA, DIAMEX and TRPO. The TALSPEAK and DIDPA [46] processes utilise an acidic organophosphorous extractant, di-2-ethylhexylphosphoric acid (HDEHP) or diisodecylphosphoric acid (DIDPA), for extraction of An(III) and Ln(III). This extraction is followed by stripping of the An(III) constituents from the solvent with a

combination of a carboxylic acid and diethlyenetriaminopentaacetic acid (DTPA). Both the TALSPEAK and DIDPA processes require reduction in the acidity of the aqueous feed solution, either by dilution or by denitration.

The DIAMEX process [47] is based on the use of malonamide extractants such as DMDBTDMA (dimethyldibutyltetradecylmalonamide) in a diluent such as kerosene. Because the DIAMEX extractant contains no metal ions, following the principle of incorporation only of constituents easily converted to innocuous volatile compounds (the so-called "CHON" principle), the waste arisings from the DIAMEX process are minimised relative to the previously described processes. The SANEX process can be coupled with DIAMEX for An(III)/Ln(III) separation. The SANEX process selectively extracts the +3 minor actinides from the +3 lanthanides with a BTP extractant, bis-1,2,4-triazinylpyridine, from relatively concentrated aqueous solutions. Recently, extractant mixtures of bis-chlorophenyldithiophosphinic acid and tri-n-octylphosphine oxide (TOPO) have been successfully tested.

The TRPO (trialkylphosphine oxide) process [48] capitalises on the high affinity of trivalent actinides and lanthanides for TRPO in moderately low acid concentrations. The extraction requires additional operations for separation of An(III) and Ln(III). The need for reduction in acidity leads to additional waste generation.

Direct separation of An(III) and Ln(III) has become possible with development of the CYANEX 301 process [49], using bis-2,4,4-trimethylpentyldithiophosphinic acid. Very high An(III) decontamination factors can be obtained with purified CYANEX-301 extractant. A complication of this process is the necessity to dilute the aqueous feed solution to quite low acidity.

Separation of americium from curium

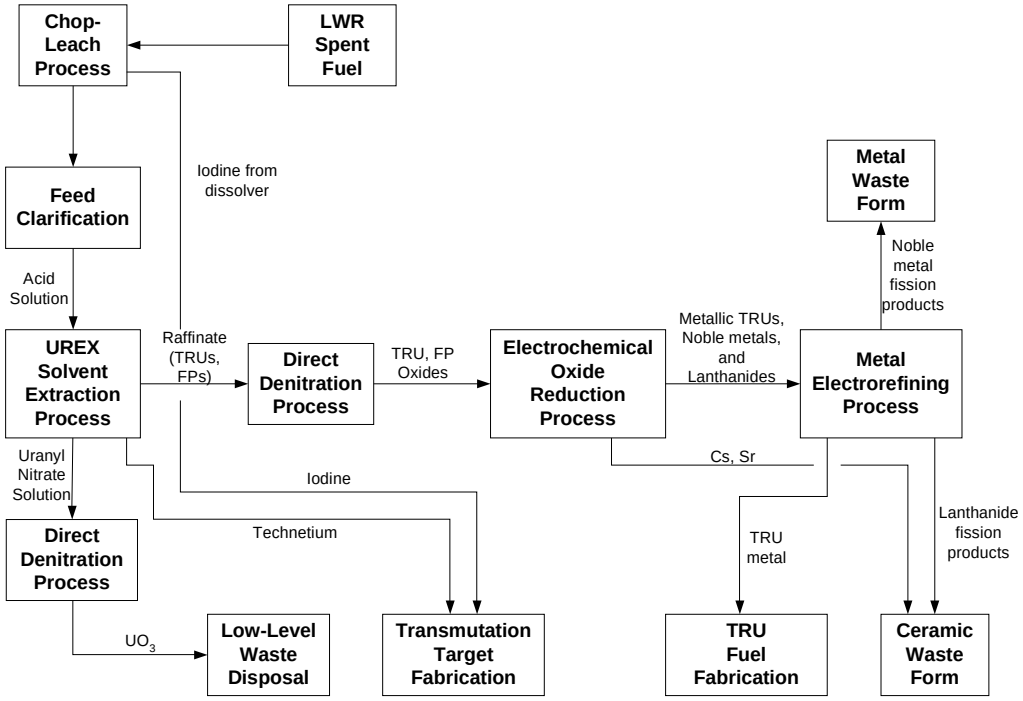
This separation can be accomplished by changing the oxidation state of americium to the extractable (IV) or (VI) states. Curium (III) is very difficult to oxidise to higher states in aqueous solution, making the separation possible. The SESAME process has been developed for this separation. It involves the electrochemical oxidation of americium in conjunction with a complexant to stabilise americium in the higher oxidation state. Potassium phosphotungstate, for example, is a good complexant for tetravalent americium.

A combination of process such as DIAMEX-SANEX-SESAME thus can serve to provide an acceptably efficient separation of americium and curium in pure form, well decontaminated of the lanthanide fission products. Such complete separations can be accomplished, as has been demonstrated, but at a cost of added expense and added high-level liquid waste volume. The question then arises as to the need for separate extraction of all of the actinides present in spent nuclear fuel. The transuranic elements are readily fissioned in the fast neutron spectrum provided by either a fast reactor or an accelerator-driven system. It is possible that certain benefits may accrue in fabrication of the fuel feed to a transmutation system if problems associated with the presence of curium can be avoided. Fabrication of transmuter fuel without the need to account for the neutron emission from curium could provide certain benefits, but at the cost of the need to store curium until its decay is complete (requiring nearly 200 years for ^{244}Cm). It may also prove advantageous to route certain transuranic elements to different transmutation systems optimised for specific elements. It is too early to make a judgement on this question, but the technology for performing any required separations will be available; it is only a matter of cost.

Alternative approaches

An example of a different approach is the hybrid processing system envisioned for the US Accelerator Transmutation of Waste (ATW) program [50], as illustrated in Figure 3.18. In this system, an aqueous solvent extraction process (UREX) is used to extract uranium from spent LWR fuel, using a complexant/reductant such as acetohydroxamic acid (AHA) to prevent the extraction of plutonium and neptunium. In this way, the transuranics are left in the first-stage raffinate solution along with all of the non-volatile fission products. The raffinate is then denitrated and the resulting oxide solids are pyrochemically processed to separate the transuranics from the fission products. The pyrochemical processes proposed will not separate the transuranics one from another, and decontamination of the lanthanide fission products will be low, on the order of a decontamination factor of 20 for individual lanthanides. These are inherent features of the processes selected, and serve the purpose of increasing the proliferation resistance of the overall system.

Figure 3.18. **Hybrid processing system utilising aqueous and dry processes for separation of transuranics and long-lived fission products for subsequent transmutation**



3.4.2.2 Pyrochemical (dry) processing

Introduction to pyrochemical processing

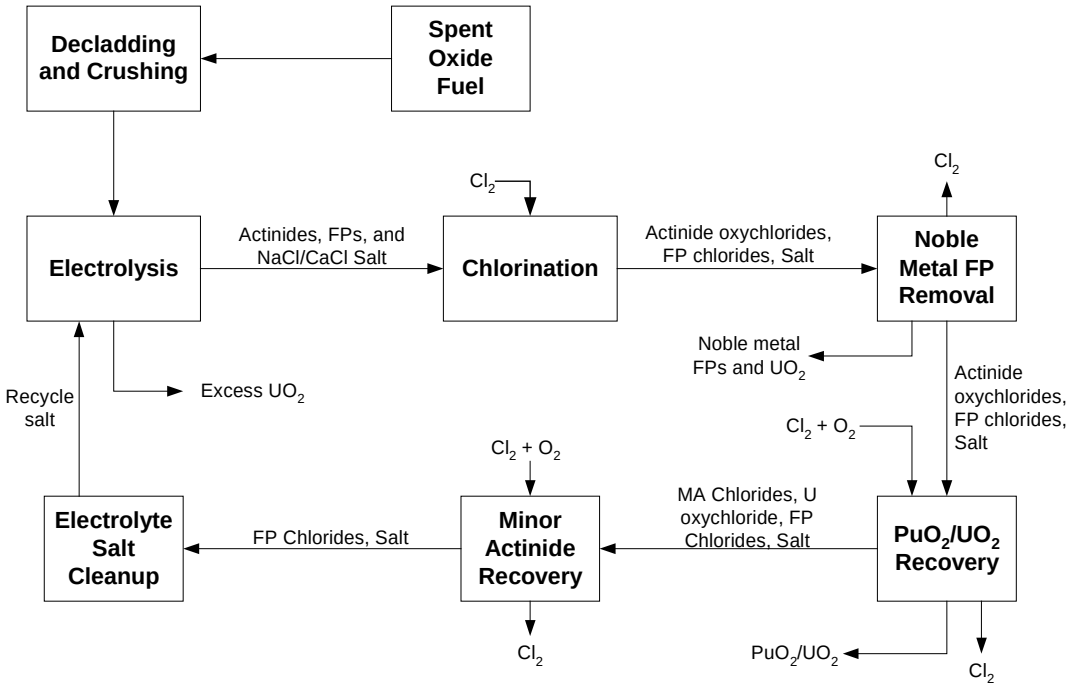
The quest for an alternative to aqueous reprocessing has been underway since the 1960s, when a rudimentary pyrochemical process was implemented for the purpose of processing spent fuel discharged from the U.S. EBR-II reactor. This process, known as “melt-refining,” simply involved the removal of volatile fission products by melting of the fuel material. Plutonium was lost to the melting crucible and later recovered, and the noble metal fission products were collectively recycled with the newly-constituted fuel as a constituent referred to as “fissium”. More recently, an advanced pyrochemical process has been applied to the processing of EBR-II fuel and blanket assemblies. This process, referred to as “pyroprocessing”, involves the electrorefining of the uranium present in the

fuel. Provided that the U:TRU ratio in the electrolyte salt is kept sufficiently high (i.e. >0.5 or so), pure metallic uranium can be electrodeposited at a solid cathode. Recovery of transuranic elements requires the use of a liquid metal cathode such as Cd, which facilitates the deposition of TRU elements by reducing their chemical activity. However, because EBR-II fuel treatment is intended only as a waste management scheme, the uranium is consolidated for storage and the transuranics and fission products present in the fuel/blanket material are left in the electrolyte salt and are subsequently incorporated in a durable high-level waste form. The metallic uranium deposit is freed of adhering electrolyte salt by vacuum melting of the deposit, which results in preferential vaporisation of the salt. This consolidation is normally done at temperatures in the 1 400°C range; at such temperatures the extremely reactive combination of uranium and salt leads to rapid attack of crucible materials.

A related pyrochemical process has been used in Russia for the processing of fast reactor spent oxide fuel. This process, also under development for a number of years [51], is illustrated schematically in Figure 3.19 and is essentially an electrowinning process in which oxide fuel is chlorinated in a chloride salt bath (e.g. NaCl-CsCl) to form oxychlorides (such as PuO_2Cl_2) of the actinide elements. The electrolysis process, typified by the reaction $PuO_2^{2+} + 2e^- = PuO_2$, results in the co-deposition of uranium, neptunium and plutonium oxides at the cell cathode and liberation of chlorine at the anode. Some contamination of the deposit with americium and curium occurs. The balance of the americium and curium remain in the salt bath. The cathode deposit is separated from adhering salt by washing with water. The recovered actinide oxides are incorporated into fresh fuel rods by vibratory compaction. Recycle of some oxide fuel elements in the BN-600 reactor has been accomplished.

Figure 3.19. **Oxide electrowinning process for treatment of oxide spent fuel and production of MOX fuel**

Process as developed by RIAR, Dimitrovgrad, Russia. Note that all steps following decladding and crushing are conducted in the same pyrolytic carbon vessel.



Pyrochemical separations processes that would find application in partitioning and transmutation systems must meet demanding requirements on actinide recovery and, in some cases, on long-lived

fission product recovery from spent fuel. The nature of the pyrochemical process to be used in particular applications depends strongly on the fuel composition, both chemical and isotopic. It also depends on economic factors related to the fuel composition and the required plant throughput rate.

Pyrochemical operations, at the present stage of technology development, are batch processes. The cost of such operations is approximately linearly dependent upon the number of batch operations to be run simultaneously. Some economies of scale can be expected from large-volume equipment fabrication, commonality of repair parts, and standardisation of procedures, but the effects are not at all comparable to the considerable economies of scale available with continuous aqueous processing. Furthermore, many pyrochemical processes require the recovery of materials from each batch operation and transport and loading of the materials to the next batch operation; this must generally be done by electromechanical means, as opposed to the simple liquid pumping operations in aqueous processing. All of these handling and transfer operations add complexity to the process, and duplication of complex equipment to provide for multi-batch operations means an increase in the probability for failure somewhere in the system. Intuitively, such considerations must impose a limit on the economic throughput capacity of a pyrochemical separations process. The limited industrial experience with such processes at present precludes a quantitative evaluation of the practical limits to throughput capacity. Nor is it possible to estimate with certainty the capital and operating costs of these processes.

Nevertheless, there are certain applications in which the use of a pyrochemical process is clearly indicated. These processes, typically operated at elevated temperatures, are ideally suited to the treatment of spent fuel that has been cooled for a short time and is generating considerable decay heat. Passively-cooled electrorefining or pyrochemical processing vessels of modest size containing, for example, about 1 m^3 of molten salt can accommodate decay heat levels of a few tens of kilowatts. As seen in Table 3.5, decay heat levels encountered with 2-year-cooled fuel in fuel cycle schemes 3b and 4 can be substantial (about 200 and 450 watts per kg heavy metal, respectively). Batch sizes, however, are limited by the amount of TRU product that can be concentrated at any given step in the process, due to criticality concerns. Therefore, even though 200 kg TRU may be critically-safe when distributed in the molten salt or molten metal in the process vessel, it is not possible to amass such a quantity in the TRU product that must eventually be consolidated by melting or other means. Batch sizes with such material must therefore be limited to around 5 kg TRU, corresponding to a batch heat load of 1-2 kilowatts or less. Process vessels must be provided with adequate heating system capacity to maintain the process medium at a constant operating temperature even after extraction of the TRU content of the batch. In addition, redundant heater capacity must be available in the event of heater element failure, and the equipment must be provided with means for expedited heater replacement so that inadvertent freezing of the molten salt can be precluded.

Another factor that can be more limiting in determining the acceptable cooling time before initiation of processing is the need to prevent fuel failure before it is sent to the fuel chopping step. Movement of fuel rods prior to that step generally assumes that the fuel is intact, so the temperature of the fuel cladding must be kept within safety limits to avoid exceeding failure strain levels. This is particularly important when handling complete fuel assemblies where the total amount of TRU elements can be 35-50 kg, sometimes necessitating active fuel assembly cooling.

So, in general, a fuel cooling time of two years is adequate for pyrochemical processing, provided that the fuel can be handled without exceeding temperature limits for the fuel cladding or the handling equipment. High radiation levels (particularly the high levels of α-radiation from the transuranic elements) that would result in serious reagent deterioration in aqueous systems are tolerable in molten salt systems because the reagents are stable. In addition, because of the absence of a moderating agent

(i.e. water), the processes can safely handle large masses of fissile isotopes, thus reducing the size and floor area required for process equipment.

For the ADS-related fuel cycle schemes that produce irradiated fuel with high decay heat levels, the total amount of fuel that must be processed annually, given the design parameters of Table 3.1, is quite small, on the order of one tonne per year for one reactor. Conservatively assuming 100 days productive operation per year, the processing rate in this case need be only 10 kg per day, or two 5-kg batches. This can be easily accommodated in pyrochemical process equipment of very modest size. Following the same rationale, pyrochemical processing operations supporting the three fast reactor based fuel cycle schemes would be presented with much less challenging heat load problems. The comparison, paralleling that from earlier in this chapter, is best seen in Table 3.12, where the calculations are based on the same parameters presented in Table 3.1.

Table 3.12. **Process decay heat loads for pyrochemical processing in the various fuel cycle schemes**

	Pu burner (FR)	**TRU burner (FR)**	**TRU burner (ADS)**	**MA burner (ADS)**	**FR (core)**
Fuel discharge, tHM/y	4.9	3.0	0.7	0.7	3.1
Decay heat, W/kgHM	21.8	46.0	192.6	455.1	18.6
Process decay heat load, kW/day	1.3	1.6	3.3	4.1	0.7

Here, it has been assumed that the processing plant operates for 100 days per year. With proper batching and equipment/facility design, these fuels can be processed without difficulty.

A variety of pyrochemical processing methods are available today, the most widely used being (1) molten salt electrochemistry (electrorefining, electrowinning); (2) volatility processes, including vacuum distillation; and (3) molten salt/molten metal reductive extraction processes. More insight in the current technological maturity and R&D needs to develop these processes to an industrial scale will be given in Chapter 7.

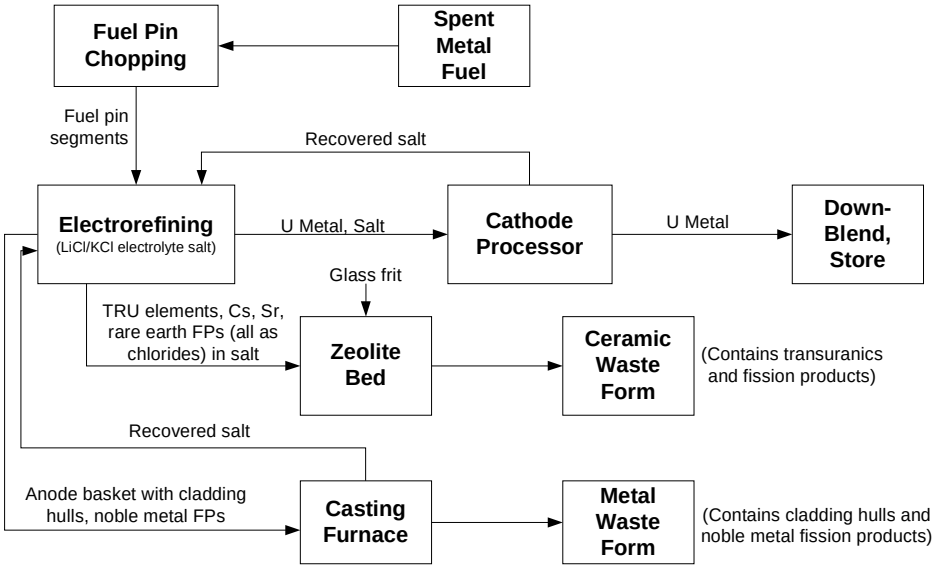
Electrorefining

The electrorefining process can be characterised simply as a process in which an impure material is made as the anode in an electrochemical cell and, with the passage of current in the cell, pure material is deposited at the cathode. In spent fuel treatment applications, the anode contains the spent fuel, generally after chopping to expose the fuel material to the electrolyte salt. The fuel can be either metal, oxide or nitride. It is not necessary to remove the fuel cladding or any other materials internal to the fuel rod. In the case of metal fuel electrorefining in which the fuel material is an alloy of uranium, transuranics and zirconium, the electrolyte salt of choice is a eutectic mixture of LiCl and KCl, operated at a temperature of 500°C. A concentration of about 2 mol% of actinide chlorides is maintained in the electrolyte salt to support electrotransport of the actinides to the cathode. With this concentration of actinide ions in the salt, electrotransport can be sustained at a high rate when a potential of 0.5-1.0 volt is imposed between the solid cathode and the anode. Reduction of the actinide chlorides occurs at the cathode and deposition of an actinide atom is accompanied by dissolution of a corresponding atom from the spent fuel. As anodic dissolution of the chopped fuel proceeds, fission products dissolve by reducing some of the actinide chlorides. This necessitates the occasional addition of a compensating amount of oxidant, usually $CdCl_2$ or UCl_3. If uranium is present in the spent fuel, and if the ratio of the concentration of UCl_3 to transuranic chlorides in the salt is greater than about

0.1, then only uranium will deposit at the solid cathode. The alkali metal and alkaline earth fission products (e.g. Cs and Sr) will not deposit because their chlorides are much more stable than the actinide chlorides. Similarly, the chlorides of the transuranic elements and the rare earth fission products are more stable than uranium chloride. So, these elements will not deposit at the solid cathode as long as the uranium concentration in the electrolyte salt is sufficient. The transition metal fission products will not be oxidised and will remain in the anode basket as metallic elements. This process is presently in use for the treatment of spent sodium-bonded driver fuel and blankets from the EBR-II reactor and is illustrated schematically in Figure 3.20.

Figure 3.20. **Schematic of pyrochemical process used for treatment of EBR-II spent driver fuel and blankets**

Because this is a waste management system, no attempt is made to recover transuranics for recycle. Recovered uranium is downblended to less than 20% enrichment in ^{235}U before storing.



The transuranic elements can be recovered in the case of the treatment of uranium-bearing fuels by the use of a different cathode, a liquid metal cathode. A common liquid metal cathode consists of a small amount of liquid cadmium contained in a ceramic crucible suspended in the electrolyte salt, with electrical contact made to the cadmium. The transuranic elements will electrodeposit in the liquid cadmium because their chemical activities are reduced by formation of intermetallic compounds such as $PuCd_6$. Uranium will also deposit in the cadmium cathode as an intermetallic compund. Even though the rare earth fission products will also form intermetallic compounds with cadmium, their chlorides are more stable than those of the actinides; thus, only small amounts of rare earths are deposited in the cadmium cathode. But it is important to recognise that in this system: (1) the uranium deposited at the solid cathode will be pure; (2) the transuranics deposited in the liquid cathode will be contaminated with 10-30 wt.% uranium and a few percent of the lanthanide elements; and (3) the properties of the transuranic elements are sufficiently similar that they cannot be separated in this process. The cladding materials of the spent fuel will not be dissolved, provided that the cell voltage is maintained below the decomposition potential of the constituents of the cladding.

In the case of non-fertile fuels (no uranium content), the transuranic elements can be electrodeposited directly on a solid cathode. Minor contamination of the transuranics with lanthanide

fission products can be expected, and there will be no separation of the transuranic elements from one another.

If the fuel to be electrorefined is not metallic, one has the option of reducing the fuel to the metallic state, as in the case of oxide fuel using the Direct Oxide Reduction process with calcium as the reductant. Alternatively, it is possible to use the Dimitrovgrad approach and capitalise on the increased electrical conductivity of oxides at elevated temperatures, directly electrodepositing the actinide oxides. Although this process has proven successful with MOX fuel, it requires further development or the use of additional process steps for complete recovery of all of the actinide oxides.

In the case of nitride fuel, actinides can be recovered much the same as with metallic fuel. Actinide elements can be electrotransported to a cathode, with nitrogen gas being liberated at the anode. An off-gas recovery system would be necessary in the case of nitride fuels highly enriched in the ^{15}N isotope to prevent build-up of ^{14}C. Noble metal fission products would remain in the anode basket, and other fission products would form stable chlorides and reside in the electrolyte salt, which could be a LiCl-KCl eutectic mixture. In the processing of non-fertile fuel, transuranic elements could be collected on a solid cathode; with nitride fuels containing uranium, the collection of pure uranium at the solid cathode would not be possible, and the use of a liquid cathode for collection of transuranics may not be necessary. Experimentation is necessary to resolve this question.

Electrowinning

Molten salt electrowinning is simply an electrolysis process in which the material to be recovered is present as a halide compound in an electrolyte salt. For example, a metal or nitride fuel could be chlorinated in a chloride carrier salt to produce chlorides of the actinide elements and most of the non-gaseous fission products (depending on the chlorinating agent). Most fuel types could be fluorinated to produce fluorides in a fluoride carrier salt. The molten salt containing the dissolved spent fuel constituents could then be placed in an electrolytic cell, with a potential applied between the anode and cathode. At the appropriate voltage level (the decomposition potential), which depends on the species to be electrolysed and the cell temperature, actinides can be deposited at the cathode and the halogen gas will be liberated at the anode. Electrowinning can also be important in certain applications of volatility processes.

Volatility processes

The most common of the volatility processes applied in spent fuel processing are chloride volatility and fluoride volatility. Such processes can be extremely useful in the processing of complex fuel types, including inert-matrix fuels. A small-scale fluoride volatility processing system was operated jointly by NRI (Czechoslovakia, now Czech Republic) and RIAR (Russia) in the 1980s. This process was used for treatment of oxide fuel from the BOR-60 reactor. Powdered fuel was fluorinated, converting uranium to UF_6 and most of the plutonium to PuF_6, both very volatile products. Re-fluorination was necessary to convert the remaining plutonium to the PuF_6 species. Most of the fission products formed non-volatile fluorides, while the noble metal fission products formed semi-volatile fluorides that tended to accompany the uranium and plutonium. After extraction of niobium and ruthenium fluorides, uranium and plutonium fluorides were then condensed at -60 and -80°C. After collection in the condensers, the uranium and plutonium fluorides were reheated, decomposing the PuF_6 to solid PuF_4, thereby allowing the volatile UF_6 to be separated from the plutonium. Column distillation was then used to purify the UF_6 by separating molybdenum, iodine and technetium fluorides.

Volatility processes have been proposed for application in the various separation schemes envisioned for the US Accelerator Transmutation of Waste (ATW) programme. A chloride volatility process could form the head-end of the treatment process for a fuel consisting of a metallic dispersion of TRU-Zr alloy in a zirconium matrix. Chlorination of the fuel in a LiCl carrier salt is followed by volatilisation of the zirconium as $ZrCl_4$. Digestion of the matrix material then provides physical access to the transuranics and fission products, which are subsequently separated by a reductive extraction process. A direct electrorefining process is currently favoured over the chloride volatility process due to the problem of contamination of the $ZrCl_4$ deposit. Fluoride volatility processing has been proposed for ATW processing of TRISO (pyrolytic carbon coated) fuel [52]. Without question, fluoride volatility processing is a powerful technology; on the other hand, it is a technological challenge to control the process so as to achieve very high efficiency, avoid ancillary recovery steps, and comply with environmental and safety regulations. Much more development is needed to realise the full benefits of this family of process methods.

Reductive extraction processes

Reductive extraction processes exploit certain well-behaved replacement reactions to separate certain fission products and actinides. Molten metal/molten salt systems are particularly useful for application of reductive extraction. An illustrative example is the treatment of chlorinated metallic fuel to separate the actinides from active metal fission products. The fuel can be chlorinated with $CdCl_2$ in a LiCl carrier salt, resulting in the formation of chlorides of the actinides, the Group I/II fission products, and the lanthanide fission products. The noble metal fission products will not form chlorides and will either sink to the bottom of the chlorination vessel or remain in the basket originally containing the fuel. The salt can then be contacted with a molten metal consisting of a dilute solution of lithium in cadmium. Lithium will reduce the actinide chlorides and, to some extent, the lanthanide chlorides. If the process is operated in counter-current mode in a train of high-temperature centrifugal contactors, about eight contactor stages can effect extraction of over 99.99% of the actinide elements into the cadmium phase, with a lanthanide contamination of this product of less than 10% of the lanthanide elements present; i.e. excellent recovery of actinides contaminated with about 10 weight percent lanthanides. Greater purification requires an increased number of contactor stages. The process has been demonstrated with four contactor stages and provided excellent separation [53]. A similar metal/salt reaction process has been reported for removal of noble metal fission products from oxide fuel that has been reduced with hydrogen and then fluorinated, with the noble metal fission product elements being digested in liquid antimony, tin, or zinc.

As described earlier in this chapter, several of the possible fuel cycle schemes can make use of pyrochemical processing methods. Because these methods tend to be non-selective with respect to the transuranic elements, they are applicable in a broad variety of systems. Process selection must be made with full awareness of the potential for contamination of the separated TRU product with traces of lanthanide fission products. If the performance of the FR or ADS fuel is sensitive to the presence of lanthanides, then certain steps, such as the use of several reductive extraction stages, must be called upon to reduce the lanthanide contamination.

Wastes from pyrochemical processing

Pyrochemical processing operations tend to produce little secondary waste because there is little or no reagent degradation. If the processes are properly designed and operated, high-level waste volumes can be minimised by recovery and recycle of salt and metal reagents. There is little published information available on wastes from pyrochemical process operations due to the relative technological immaturity and lack of industrial-scale experience. Therefore, the technology can be assessed only on the basis of extrapolations from laboratory-scale studies, and most of this experience

has been with the electrorefining process used for separations of actinides and fission products in the metallic state.

In the case of the electrorefining process developed at the Argonne National Laboratory in the US, when applied in fuel cycle schemes 3a, 3b, 4 and 5, the high-level waste forms are identical. This is the case regardless of whether the fuel type being treated is metal, oxide or nitride, metmet, cermet, or cercer. Different waste forms would probably arise in the case of coated-particle graphite reactor fuels or in the case where a fluoride salt mixture is utilised in the process, but the discussion will be limited here to the chloride-based processes for the sake of brevity.

In the reference process, then, the active metal fission products (Cs, Sr, etc.) will reside in the electrolyte salt together with trace amount of actinide elements. The transition metal fission products will remain in the anodic dissolution baskets together with the cladding hulls. So, there are two waste streams to deal with, one salt and one metal. Because a chloride salt is not amenable to vitrification, development of a different waste form was necessary. A natural chloride-bearing mineral, sodalite ($NaAlSiO_4.NaCl$), exists in the geologic structures of the proposed Yucca Mountain repository in the United States. This mineral can be synthesised by mixing the fission product-loaded electrolyte salt with a zeolite (Zeolite A, $Na_{12}Al_{12}Si_{12}O_{48}$) and heating to temperatures near 900°C. The synthesis is catalysed by the presence of borosilicate glass frit, which also serves to encapsulate the sodalite particles, providing an additional barrier to radionuclide release. As is the case with vitrification of high-level waste, the fission product loading of the waste form is constrained by the limiting centerline temperature of the waste form and the total heat generation per unit area acceptable in the high-level waste repository of interim storage site.

The composite glass-ceramic waste form developed by Argonne is limited by the temperature at which transformation of the sodalite to nepheline (a related mineral but one which does not retain fission product cations well) occurs. This restricts centerline temperatures to around 650°C, and because the thermal conductivity of the composite is similar to that of borosilicate glass, the maximum fission product loading is about the same as for glass, around 15 weight percent. The limits on actinide content are higher than for glass (which is limited by the solubility of actinides to around 1 weight percent), but the fission product decay heat limit prevails. Little is known about long-term degradation of the sodalite structure by α-particle damage.

A variety of leach tests conducted with differing leachant chemistries and environmental conditions have shown that the release rate of important radionuclides from the pyrochemical process ceramic waste form is comparable to the releases from the best vitrified waste forms and well over an order of magnitude less than the release from spent fuel under direct disposal conditions. These tests have been carried out in evaluation of the potential behaviour of this waste form in the oxidising environment characteristic of the Yucca Mountain repository. Under reducing conditions, the behaviour is not known.

The metallic waste stream is generated by removing the anode baskets from the system and melting the baskets together with the metallic fission products and the cladding hulls to produce a corrosion-resistant metal alloy. This alloy is dependent in composition on the nature of the cladding material, which dominates the mass of the waste form. A metal waste form based on stainless steel cladding hulls has shown a release rate for technetium that is several orders of magnitude less than than the release of technetium from spent LWR fuel.

The volume of waste emanating from pyrochemical processing operations is somewhat dependent on the nature of the fuel being processed and even more dependent on the complexity of the overall process. Although there is little extended large-scale experience with such operations, it is possible to

estimate the resultant waste volumes on the basis of laboratory-scale testing. Table 3.13 shows estimated waste volumes for the fuel cycle schemes of this report that involve pyrochemical processing (schemes 3a, 3b, 4 and 5). Process wastes, also known as secondary wastes, are difficult to estimate; therefore, a fixed value of 0.2 m^3 per tonne was used in this table.

Table 3.13. **Estimated waste volumes from pyrochemical processing operations in fuel cycle schemes 3a, 3b, 4, and 5**

Fuel cycle scheme	Waste stream and type	Est. volume (m^3/TWhe)
3a	LWR uranium (LLW)	0.20
	LWR ceramic waste form (HLW)	0.27
	LWR metal waste form (HLW)	0.08
	FR ceramic waste form (HLW)	0.20
	FR metal waste form (HLW)	0.01
	Process wastes (LLW)	0.7
3b	LWR uranium (LLW)	0.25
	LWR ceramic waste form (HLW)	0.33
	LWR metal waste form (HLW)	0.10
	ADS ceramic waste form (HLW)	0.07
	ADS metal waste form (HLW)	0.02
	Process wastes (LLW)	0.8
4	LWR-UOX PUREX wastes (HLW)	0.38
	LWR-MOX PUREX wastes (HLW)	0.06
	FR-MOX PUREX wastes (HLW)	0.02
	ADS ceramic waste form (HLW)	0.03
	ADS metal waste form (HLW)	0.002
	Pyrochemical process wastes (LLW)	0.1
	PUREX process wastes (LLW)	2.0
5	FR ceramic waste form (HLW)	0.56
	FR metal waste form (HLW)	0.04
	Process wastes (LLW)	0.2

(HLW: high-level waste; LLW: low-level waste).

3.4.3 Secondary wastes arising in fuel cycle schemes

It is generally acknowledged that in addition to the unwanted fission products, fuel cladding, etc., that would constitute the primary fuel cycle PUREX waste from a P&T scheme, there would be generation of secondary waste. This secondary waste would comprise all insoluble active residues, degraded solvents or salts, ancillary materials, and analytical wastes, etc. that arise during additional fuel cycle operation. It is suggested that the majority of secondary wastes which arise during processing operations would be generated during solvent/salt cleanup and recovery operations [54]. Table 3.14 summarises the potential secondary waste forms that could be generated with the operation of P&T fuel cycles. Note that in this analysis of secondary wastes, construction and decommissioning wastes have not been included. However, it is obvious that the increased number of multistage processes and greater shielding requirements would generate considerably more decommissioning wastes that would include secondary, active, wastes owing to sorption of material onto process surfaces and accumulation of active fines, etc. but these cannot yet be realistically quantified.

3.4.3.1 Front-end secondary wastes

The front-end of fuel cycle schemes 1, 2, 3a, 3b, and 4 have common stages associated with the production of UOX-fuel for irradiation in conventional thermal reactors. These fuel cycles will generate wastes during the mining and milling stages, which are reported to represent the major health and environmental hazards of the nuclear industry [55]. Secondary wastes in the form of uranium residues in the tails from milling operations represent a definite anthropological hazard with respect to the in-growth of radioactive progeny. The generation of fugitive dust, from tailings dams can have considerable impact on the localised populations. Additional secondary wastes include sulphuric acid and ammonia that are used during the leaching and purification of uranium ore concentrate, used filter cloths, contaminated solvent extraction raffinate, dilute hydrofluoric and hydrochloric acids and magnesium or calcium fluoride slag. The use of process and ancillary service materials such as petrol and diesel will also generate additional secondary wastes. However, the stages that follow mining and milling generate considerably less waste and have a lower impact on the environment. Conversion and enrichment processes are essentially clean processes that generate very small quantities of secondary waste mostly in the form of scrap metal and analytical wastes.

The power production, or reactor, stage of the nuclear fuel cycle generates relatively little secondary waste. The secondary waste that does arise will be due mainly to the treatment of coolant, such as corrosion products, spent demineralisation resins, filters, ancillary service material and general maintenance and analytical wastes. However, the stages downstream of thermal reactor operation in each of the fuel cycles differ considerably. Analysis of the back-end of the nuclear fuel cycle will show the increased secondary waste generation of P&T schemes becomes more apparent.

3.4.3.2 Back-end secondary wastes

It is assumed that final disposal stage would entail long-term storage in a deep geological repository. The primary waste associated with this stage would be the waste rock inventory, which itself is radioactive, generated during construction of the repository. The secondary wastes are difficult to identify, however it seems feasible to assume that similar secondary wastes from the related ancillary services to those generated during mining operations would arise. In contrast, the irradiated fuel from the thermal reactor in the P&T schemes would be subjected to processing following an appropriate post-reactor cooling period. The secondary wastes and the associated increase in dose in most P&T scenarios will arise from the separation and treatment of irradiated fuel components, and their subsequent fabrication into new fuel or transmutation target materials.

PUREX is proposed for the processing of thermal UOX fuel, and it is also proposed that advances in PUREX technology would allow the recovery of Np. It is suggested that a multistage processing operation involving the combination of DIAMEX-SANEX-SESAME technologies is implemented to allow the efficient separation of americium and curium from PUREX HAR and to strip the undesirable lanthanide fraction. The primary wastes produced by this multiplex processing system would comprise HLW of lanthanides, fission products and trace quantities of actinides. The secondary wastes are summarised in Table 3.14. The solvent used in DIAMEX (DMDBTDMA in kerosene or dodecane), conforms to the CHON principle, which implies that the spent solvent would be totally incinerable and as such would reduce the total solid secondary waste arising. However, a solvent waste stream containing up to 11% of the ruthenium in the feed to the DIAMEX process would be generated during solvent processing [56]. The ruthenium fraction could be oxidised to volatile RuO_4 prior to DIAMEX separation although this effectively generates a new RuO_4 secondary waste, which would require scrubbing from the solvent. Iron could accumulate in the solvent phase also, potentially resulting in the formation of a problematic third phase, which would require subsequent removal from the solvent during clean-up processing. Additional scrubbing cycles would be required to remove any residual

ruthenium prior to incineration of the spent solvent. Radiolysis of the solvent would lead to the formation of carboxylic acids, amine, amide-acid and other degradation products, which decreases the solvent extracting properties [47].

SANEX would generate secondary waste from off-gas scrubbing and by radiolysis of the organic solvent. The presence of ^{242}Cm and ^{241}Am would increase the α-level activity and promote solvent degradation. The organic solvent degradation products would depend on which soft donor extractant is used, as several have been suggested (See §3.5.1). Similarly, SESAME would generate waste streams contaminated with trace quantities of Am and Cm from product finishing. The waste solvent could be polluted with activated ^{110}Ag and quantities of waste complexing agents, heteropoly acids such as potassium phosphotungstate, would be generated. The advanced separation processes referred to above are currently in the development stage and as such it is not possible to provide an accurate account of the potential secondary wastes associated with their operation. However, it seems feasible to suggest that implementing such complex multistage processes would generate additional secondary wastes, which would further contribute to the operational dose of the fuel cycle and toxicity of the wastes.

The majority of secondary wastes that arise during processing operations would be generated during solvent cleanup and recovery operations [54]. It is feasible to assume that secondary wastes generated during PUREX processing for thermal UOX compared to FR-MOX fuels would be similar, although probably different in magnitude. Solvent radiolysis or hydrolytic decomposition during PUREX operation would generate the major fraction of secondary wastes, including organic degradation products such as dibutylphosphate (DBP), monobutylphosphate, alkanes, nitro-alkanes, carboxylic acids, carbon dioxide, and phosphoric acid. The α-level associated with recycle FR-MOX fuel would be approximately 30 times greater than the equivalent UO_2 fuel of the same irradiation (See Table 3.5). Consequently, the extraction of FR-MOX fuel that has been subjected to high irradiation would result in increased formation of DBP via solvent radiolysis, due to the increased Pu and MA inventories in the spent fuel (average 140 kg DBP/TWhe for fast reactor fuel). Trace quantities of insoluble crud would be found in solvent wash raffinates. For example, the solvent wash raffinate from the processing of thermal oxide fuel would typically contain around 0.8 g/l U, 8 mg/l Pu and 0.3 mg/l Np. The increased degradation of solvent and diluent expected during the processing of FR-MOX fuel might be accompanied by an increased inventory of insoluble actinide residues, the magnitude of which would depend on the complex interactions of many operating variables such as initial fuel composition, irradiation, dissolution conditions, etc. Distillation of the solvent wash raffinate reduces the volume but generates a higher-boiling residue containing traces of uranium, plutonium, and minor actinides. The recycle of Pu in the FR stage of the fuel cycle would generate quantities of secondary waste subject to accumulation with each cycle.

Secondary waste generation during fast reactor operation would be relatively small in comparison to that from recycling operations. Ancillary service wastes would be expected to be similar to those of the thermal reactor stage; however, the secondary wastes associated with coolant could differ and would depend on the coolant media, e.g. sodium, employed in the fast reactor system. A coolant treatment system would be necessary that would operate on a continuous or periodic basis to remove potential corrosion and activation products.

The operation of an ADS would incur the generation of different secondary wastes to those associated with conventional thermal and fast neutron reactor systems and in greater quantities. The formation of activation and spallation products would have serious safety implications and require continuous or regular periodic removal. The use of Pb or Pb-Bi eutectic (LBE) target and coolant media could result in the generation of isotopes such as ^{202}Pb, ^{205}Pb, ^{208}Bi and ^{210}Po and other possible reaction products (mostly lanthanides). Preliminary calculations have indicated that fission products would account for as little as 10% of the activity in the target [57]. In addition, radiation damage in

structural materials such as the beam tube and beam window could occur due to the formation of spallation and activation products such as isotopes of Fe, Cr, and Ni, depending upon the alloy employed [58]. The generation of gaseous secondary wastes would be expected from sources such as target venting, the coolant system, the supply of an inert operating atmosphere e.g. helium, and beam dumps. Solid secondary wastes associated with the ADS operation would be expected to comprise PPE, HEPA filters, batteries, scrap metals and glass, etc. [59].

The secondary wastes associated with the pyroprocessing of the post-ADS target material would include zeolites, used for ion exchange purification of the process salt medium, and waste eutectic salt and inclusions such as $CaCl_2$, FP, $MgCl_2$, LiCl, KCl, Li_2O, etc. It has been estimated that 0.3 m^3 of mineral wastes and 0.05 m^3 of metallic wastes would be generated per tonne of reprocessed material [60]. Zeolite ion exchangers would be used for extracting fission products and rare earths, after which they would be collapsed and mixed with anhydrous zeolite to promote salt uptake into the crystalline matrix prior to conditioning for final disposal (See 3.5.2). It is suggested that such waste forms would typically contain around 4 wt% FP. Recycling of the molten salt eutectic mixes could potentially reduce the generation of secondary wastes during pyroprocessing [61]. In addition there would be generation of process fines that would be subject to sorption onto the internal surfaces of equipment, cells and filters. However, the magnitude of secondary wastes generated during pyroprocessing is hypothesised as being relatively low owing to the reduced or negligible reagent degradation, based on extrapolations of current knowledge (See 3.5.2).

Generally, it is proposed that schemes 1 and 5 would generate the smallest secondary waste inventories owing to their relatively simple designs, i.e. fewer multiplex processes. However, scheme 1 generates the greatest secondary waste associated with raw materials extraction and scheme 5 assumes sole use of fast reactor technology and no LWR fuel cycle operations, which does not account for any phase out of current technology. In addition scheme 5 would incur penalties associated with the recycle of TRU cycle including increased criticality risks during FR-MOX dissolution, which would require the construction of dedicated head-end facilities (See §3.5.1). The dose due to recycling of Pu and MA would result in greater handling risks owing to larger inventories of ^{238}Pu and ^{241}Am, which would generate increased neutron and gamma radiation around the glove boxes in fuel fabrication [62]. In addition, further doses would arise through (α, n) reactions in equipment handling ^{238}Pu. This is exhibited in the greater levels of radiation following cooling, and during fabrication, associated with the TRU and MA burner schemes shown in Tables 3.5 and 3.6. Scheme 4 the double strata strategy (Figure 3.4) would generate the greatest quantity of secondary waste owing to its multiplex nature and multiple recycling (See Section 3.1.2).

Categorisation of the secondary waste associated with fuel cycle operation is difficult owing to the uncertainty inherent in any assessment of potential inventories. However, it is possible to suggest that secondary wastes arising from ancillary service materials and secondary liquid effluent treatment, e.g. secondary filter cartridges from steam generator blowdown systems that are contaminated with small quantities of fission products and activation products would be considered LLW. Sludges and concentrates, contaminated with fission products and activation products with trace quantities of actinides would be categorised as ILW. Similarly, material contaminated with activation products, fission products, actinides and neutron-activated products would also be categorised as ILW. In Table 3.14 categories are assigned to the secondary wastes that would potentially arise during the operation of the five principal fuel cycle schemes selected by the expert group.

3.4.4 Depleted and reprocessed irradiated uranium

The management of depleted uranium (DU) and reprocessed irradiated uranium has received relatively little interest over the past years as their environmental impact is very low and, in today's fuel cycles, is overwhelmed by the potential radiological impact from the intermediate and especially high level waste in the long-term. The P&T-schemes under consideration in this study may however reduce the amount of long-lived high-level waste with a significant factor, i.e. a factor hundred or more. Therefore, the management of this depleted uranium and reprocessed irradiated uranium may become a more apparent issue in the future if such P&T-schemes would be deployed.

Table 3.14a. **Summary of secondary wastes arising from fuel cycle front-end and reactor**

FUEL CYCLE STAGE	CATEGORY[a)]	SECONDARY WASTES
Mining & milling	LLW	Residual uranic wastes contained in drilling mud and displaced rock.
	LLW	Sulphuric acid, ammonia, used filter cloths, activated solvent extraction raffinate, dilute hydrofluoric and hydrochloric acids, and magnesium or calcium fluoride slag.
	LLW	Emissions from ancillary services.
Enrichment & conversion	LLW	Maintenance wastes: small quantities of coolant, inert gases, scrap metal, polythene, paper towels, etc.
	LLW/ILW	Analytical wastes
	ILW	Conversion: trace uranic quantities from scrub liquor and crud.
	LLW	Conversion: wastes from ancillary services such as polythene, paper towels, carbon and cloth filters, etc.
Power production/ transmutation	ILW	Cooling system maintenance – corrosion products, isotopic activation products
	LLW/ILW	Spent demineralisation resins, and ancillary service material e.g. cooling tower blowdown, sewage, used oil, contaminated oil, wastewater treatment sludge, etc.
	ILW	ADS: Spallation and activation products associated with coolant irradiation, e.g. formation of ^{210}Po from neutron capture in ^{209}Bi (in LBE coolant / target material), ^{202}Pb and ^{205}Pb, ^{208}Bi. Activation products such as Mo, Ni, Cr, Fe, etc., from irradiation of structural components such as the beam window and beam tube.
	ILW	Corrosion products from mechanical degradation of structural materials
	LLW	Activated air & offgas emissions from: target venting, coolant system, inert purge system, beam dumps
	ILW	PPE, HEPA filters, batteries, scrap metal & glass, etc.

a) Category = radioactive waste category: LLW: Low-level Waste; ILW: Intermediate Level Waste.

Today, the strategy for the long-term management of depleted uranium is based on the consideration that this depleted uranium is a valuable material, which may have various applications, and is not considered a waste. The use of this depleted uranium in fast reactor systems, as shown in

this study, is one of the applications where the re-enrichment is a second potentially valuable source of ^{235}U for LWRs, whilst the remaining ^{238}U may again be used in future fast reactor systems. In the absence of these, or other, large-scale applications, however, final disposition in some form of "repository" would have to be considered.

Table 3.14b. **Summary of secondary wastes arising from fuel cycle back-end**

FUEL CYCLE STAGE	CATEGORY	SECONDARY WASTES
PUREX	ILW	Degraded solvent and wash raffinate containing organic solvent degradation products such as DBP, MBP, alkanes, nitro-alkanes, carboxylic acids, carbon dioxide, and phosphoric acid.
	ILW/HLW	Trace quantities of insoluble U, Pu, FP, and MA dissolver solids, and soluble traces in solvent wash raffinates, e.g. from processing of thermal oxide fuel, typically around 0.8 $g.l^{-1}$ U, 8 $mg.l^{-1}$ Pu and 0.3 $mg.l^{-1}$ Np.
DIAMEX (fission product and transuranic separation)	ILW/HLW	11% initial Ru in solvent wash raffinate, possibly removed in scrub liquor following oxidation to RuO_4.
	ILW	Possible Fe (corrosion product) accumulation in solvent.
SANEX (Trivalent actinide and lanthanide separation)	ILW	Scrub liquor, from off-gas scrubbing.
	ILW	Organic solvent degradation products – dependent upon extractants, e.g. TPTZ, or *di*-thiophosphoric acids in combination with TBP.
	ILW	Trace quantities of Am, Cm, from process raffinate.
SESAME (Am/Cm separation)	ILW	Contaminated condensate containing Am and Cm from product streams and associated scrub liquor.
	ILW	Waste solvent (solvent cleanup and recovery operations) possibly containing activated ^{110}Ag and waste heteropoly acid.
	LLW	Acidic wastes, phosphoric acid.
Pyroprocessing (Pu/MA recovery from spent ADS fuel)	ILW	Scrub liquor from off-gas treatment.
	ILW	Cladding hulls, noble metals.
	ILW	Waste eutectic salt & trace inclusions (including the more electropositive FPs).
	HLW/ILW	Ion exchange zeolites, waste crucibles, scrap metallic waste.
Storage (intermediate) & Final disposal (repository)	LLW	Ancillary service wastes.

Depleted uranium, initially occurring as UF_6, can be stored safely for many decades in steel containers in the open air in storage yards. However, depleted uranium stored in the UF_6 form may represent a potential chemical hazard if not properly managed. Alternatives for the strategic management of depleted uranium therefore include the deconversion of UF_6 stocks to stable forms

more suitable for long-term management. Due to their high chemical stability and low solubility, uranium oxides in general are the favoured form for this. Generally, though not exclusively, storage as U_3O_8, the most stable oxide, is considered for long-term storage where continued storage as UF_6 is not appropriate. Large-scale storage of compacted U_3O_8 has been undertaken in France since 1984. About 130 000 tU of UF_6 have already been converted into U_3O_8. The powder is held in about 3 m^3 painted mild steel DV 70 type containers stacked three high in "warehouses".

The radiological characteristics of DU are a consequence of the properties of the three uranium isotopes: ^{238}U, ^{235}U and ^{234}U and their daughter products. DU is safe against criticality under all naturally occurring conditions. The initial activity of DU when it is newly produced is very low, around 23 Bq/g and the toxicity is also low, around 0.75 Sv/g. Whatever the ^{235}U assay of uranium, as time passes, decay products will appear meaning that the activity and the radiotoxicity will increase. The activity and radiotoxicity levels for DU will become the same as for uranium ore (with the same original amount of uranium) after a time period of around 1 million years.

Reprocessed irradiated uranium is currently not systematically recycled in UOX- or MOX-fuel form. The reprocessed uranium distinguishes itself from natural uranium by the occurrence of higher amounts of α-emitting isotopes:

- ^{232}U, not present in natural uranium, has a higher specific activity than ^{235}U and some of the daughter-products are α-emitting, i.e. ^{228}Th with a half-life of 1.9 years, as well as emitting hard γ-rays, i.e. ^{208}Tl. The γ-activity attains a secular equilibrium after about 10 years attaining significant higher values than for natural uranium.

- ^{234}U occurs as a natural α-emitter and is accumulated in this irradiated uranium. In addition, Np and Pu are also occurring in reprocessed irradiated uranium and add to the α-activity.

- Trace amounts of ^{106}Ru occur which increase the γ-activity.

The nuclear industry has in place the facilities that are needed to recycle reprocessed uranium (REPU) on a semi-industrial scale. This includes chemical conversion of REPU, enrichment, fuel fabrication and transport as well as reactor irradiation of REPU-based fuel. In Japan, JNC carried out experiments on the REPU conversion technology at Ningyo-Toge conversion facility. The amount of REPU converted to UF_6 for re-enrichment reached 336 tU. Recycling of REPU from LWR fuel is now demonstrated, albeit limited to a fraction of the available material and to a few numbers of reactors.[35] The enrichment of REPU is today based on centrifuge technology and is currently performed in Russia and in the Netherlands. Altogether, the reprocessing plants have delivered more than 12 000 tonnes of REPU where this amount will further grow in the years to come in a pace comparable to the output of a large uranium mine. More recently, an alternative way of REPU reuse has emerged which relies on blending REPU with high-enriched uranium resulting in a reduced ^{236}U-content in the enriched REPU fuel, thus reducing neutron absorption and improving the economy of REPU recycling. If REPU is considered as a by-product from reprocessing, the economics realised in replacing natural uranium by REPU compensate for the extra expenditures related to storage, conversion, enrichment and fabrication depending on the source material. The competitiveness of REPU is strongly related to the cost of reprocessing and of natural uranium, but also to its isotopic composition that varies from a batch to the other. The future of REPU is anticipated to be governed by economic and strategic, e.g. conservation, considerations. Considering the savings which can be made by recycling REPU of relatively good

35. Substantial quantities of REPU from MAGNOX fuel have been re-enriched in the UK to natural level for recycling, while the Doel-1 reactor in Belgium has been operated exclusively with re-enriched REPU for a number of years.

specifications, despite an extremely depressed natural uranium market, it is possible that in the coming years, those utilities having selected reprocessing will also recover uranium through recycling.

In conclusion, as long as LWRs would make up a significant part of the nuclear power plant park in future fuel cycle schemes, an increasing amount of depleted and reprocessed irradiated uranium would emerge. Especially the latter would be of increasing importance, as depleted uranium would be used in fast reactor systems where the recycling of reprocessed irradiated uranium would not compensate for its production. Only the use of this reprocessed irradiated uranium in future all FR-scenarios (scheme 5) may indicate a steady decrease of the build-up inventory of depleted uranium and the reprocessed irradiated uranium. In the very long-term (about 1 million years), disposition of this depleted and reprocessed irradiated uranium results in an activity and radiotoxicity level comparable to natural uranium.

3.5 Conclusions

The results of the comparative analysis of fuel cycle schemes, described in this chapter, can be summarised as follows:

- All transmutation strategies with closed fuel cycles could, in principle, achieve high reductions in the actinide inventory and the long-term radiotoxicity of the waste, and these are comparable with those of a pure fast reactor strategy. With respect to these reductions, the potentials of the FR and the ADS are very similar. The choice of the fuel cycle scheme affects the radiotoxicity reduction factor only within a factor of about two.
- Under the assumptions used in the analysis, these strategies can achieve a more than hundred-fold reduction in the long-term waste radiotoxicity and even higher actinide inventory reduction factors (more than 1 100 for the heavy metal and 175 for the transuranics), compared with the once-through fuel cycle.
- The reduction factors are primarily determined by the fuel burn-up and the reprocessing and fuel fabrication losses. An ambitious goal for the recovery of all actinides (99.9%, as already achieved for uranium and plutonium with aqueous processes) must be set, if the quoted reduction factors are to be realised.
- With regard to actinide waste production and technological aspects, the TRU burning in FR and the double strata strategies are similarly attractive. The former can gradually evolve to a pure fast reactor strategy, but requires a higher initial investment in fast reactor and advanced fuel cycle technology. The latter confines the minor actinides to a small part of the fuel cycle, but calls for particularly innovative technology for this part of the fuel cycle.
- Transmutation systems with partially closed fuel cycles, e.g. heterogeneous transmutation schemes, in which americium and curium are separated from the fuel and recycled in special “target” pins which are disposed of after irradiation, are technologically less demanding than a closed fuel cycle strategy, but are also about a factor of two less effective in reducing the radiotoxicity. They are being explored as a near-term transmutation option which does not depend on an ADS.
- Multiple recycling of plutonium without minor actinide transmutation is useful for the management of plutonium, but cannot qualify as a transmutation strategy because it reduces the long-term radiotoxicity of the high-level waste by only a factor of about five.
- The sub-critical operation of an actinide burner with a fast neutron spectrum offers interesting additional parameters of freedom in the core design. In particular, the possibility of operating

such a burner with a uranium-free (or thorium-free) fuel supply allows the burner effectiveness to be maximised and hence the fraction of specialised transmuters in the reactor park to be minimised.

- A further advantage of the sub-critical operation mode is the tolorance of the system against degradations in the safety characteristics of the core. Both of these advantages are of particular relevance for systems which burn pure minor actinides, i.e. minor actinide burners in a double strata strategy.
- Actinide transmutation implies the handling of unusual fuels with very high decay heats and neutron source strengths. A significant effort is required to investigate the manufacturability, burn-up behaviour and reprocessability of these fuels. This applies particularly to fuels with high minor actinide content, which can probably be reprocessed only with the help of pyrochemical methods. These methods have to be further developed to tolerate from ten to more than twenty times higher decay heat levels than those encountered in the pyrochemical reprocessing of normal fast reactor fuels.
- The introduction of pyrochemical processing techniques at the industrial level will require the development of new process flowsheets and the use of potentially very corrosive reagents at high temperatures. These processes will generate chemical and radiological hazards which will have to be mitigated. A compensating benefit of elevated-temperature operation is the increased reaction rates that prevail. The recovery efficiency of such processes is yet to be proven on an industrial scale.
- The PUREX aqueous reprocessing can be considered as valid for the FR-MOX fuel in the plutonium-burning and double strata schemes. Reprocessing of this fuel within short cooling times and with the required high recovery yields, however, will require the plutonium dissolution yield to be improved and the PUREX flowsheet to be modified.
- Due to the high radioactivity of multiple recycled FR-MOX fuel, its handling will require measures to be taken to reduce the radiation doses in the fabrication plant and during the transportation of the fuel assemblies. The increased requirements for shielding, and preference for short transportation paths, of multiple recycled fuels also favour the pyrochemical reprocessing method.
- All transmutation strategies which include LWRs in the reactor mix produce large streams of depleted and irradiated uranium. If this uranium is not considered as a resource for future fast reactors, its long-term radiological impact has also to be taken into account.

4. ACCELERATOR-DRIVEN SYSTEM (ADS) AND FAST REACTOR (FR) TECHNOLOGIES

4.1 Introduction

The previous chapters have indicated that advanced nuclear fuel cycles incorporating P&T may include fast neutron spectrum reactors, whether of FR- or ADS-type. While both share, in principle, the characteristics of a fast spectrum and consequently share also fuel, material and coolant technology, distinctive technological differences occur between FRs and ADSs. These differences relate to:

- *Fuel*: as ADS would use more fertile-free fuel and would also allow higher concentrations of minor actinides in the fuel, this influences the fabrication and the reprocessing potential of such fuels.
- *Materials*: while the fast neutron spectra are quite comparable in a FR and in the core lattice of an ADS, the harder neutron spectrum in the source region of an ADS as well as the emission of energetic charged particles from the spallation source impose additional constraints on the choice and behaviour of the target materials, the adjacent reactor structures, and especially the beam window. Activation by high-energy particles is also a new issue.
- The *target and sub-critical lattice* in an ADS present new challenges for technology and is one of the major differences between ADS and FR.
- Finally, the need for an *accelerator* to drive the ADS is an additional component which needs further development towards higher performance and reliability.

This chapter will deal with these technological differences and especially the new requirements for ADS development. Chapter 5 will deal with the question of safety of ADSs versus FRs and will highlight additional technological aspects to complement those mentioned in the following chapter. Chapter 3 has already introduced the fuel fabrication and reprocessing issues and we shall therefore focus in this chapter on the reactor technology for FR and ADS.

In the absence of any specifically agreed international design, this chapter will discuss the main technological aspects which are design-independent and will highlight the following three questions:

- To what respect would the required ADS-technology differ from the already developed FR-technology and what are the additional developments needed?
- Can the existing FR technology basis be of specific use for ADS-development and is there scope for synergy between both developments? In other words, what is the extra effort needed?
- What are the main technological bottlenecks in ADS or FR developments for deployment of an industrial-scale waste transmutation system?

After a short history of FR-development and the current status of FR-technology, this chapter will give an overview of some ADS concepts and detail the technological challenges related to the development of ADS in general. A summary of the comparison of these technological differences will conclude this chapter.

4.2 Common grounds of ADS and FR technology

4.2.1 History and current status of existing FR technology

The FR history is as old as that of thermal reactors. For the first 20 years of their existence, these two systems advanced side by side. The first FR was Clementine at Los Alamos (USA) in 1946 with a power of 150 kW. The first nuclear reactor in the world to generate electricity was a FR, the EBR-1 in the United States, in 1951. Around the sixties, as shown in Figure 4.1 and Table 4.1, four experimental fast reactors of about the same power went critical, and the oldest and largest of them, DFR (72 MWth), was successfully operated over 18 years. So also was Rapsodie later on. BOR-60 is still in operation now. After DFR, which used sodium-potassium, sodium was adopted as primary coolant.

The first prototype fast reactor for power generation was the US Enrico Fermi reactor (1964, 66 MWe). After three years of operation, this reactor suffered a fuel melting incident and was finally shut down in 1972. From 1972 to 1974, three prototypes of comparable size were successively brought into operation: BN-350 in the USSR (now in Kazakhstan), Phénix in France and PFR in the UK. The second is still in operation. BN-350 and PFR were finally shut-down in 1999 and 1994, respectively. The cores of these two plants operated satisfactorily, but the plants experienced steam generator problems. The SNR-300 prototype was built in Germany by a German-Belgian-Dutch consortium; plant and fuel were ready in 1985, but owing to a political impasse, the plant was never allowed to start up. The first criticality of the Japanese prototype Monju occurred in April 1994. Monju experienced a sodium leakage in the secondary loop in December 1995.

The stage of the large (pre-industrial) demonstration plants began with the start-up of BN-600 (600 MWe) in the USSR (Russia) in 1980 and Superphénix (1 240 MWe) in France in 1985. These achievements are further discussed below, together with those in Japan and other countries. Due to technical difficulties associated with the use of sodium as a coolant and economic problems in a saturating rather than expanding nuclear energy market, BN-600 and Superphénix remained the only industrial-scale fast reactors, meaning that the experience base for such reactors is much smaller than that for thermal reactors.

The motivation for building fast reactors has progressively changed. At the outset, the main objective for developing the FR was breeding in order to conserve uranium resources. It is easy to see the advantage of such a technology in an era of uranium shortage and price increase, as was forecast in the nineteen-seventies. In reality, however, uranium remained abundant and cheap, mainly because the growth rate of nuclear energy was lower than had been expected. Consequently, the use of FRs in a “burner” mode for managing excess plutonium gained in importance and remains today a particular focus of fast reactor R&D activities. Moreover, the desire to further optimise the back-end of the fuel cycle including the disposal of high-level waste has recently been stimulating an increasing interest in extending the application of the FR from the burning of plutonium to the burning (transmutation) of all transuranic actinides.

Table 4.1a. **Main features of constructed fast reactors**

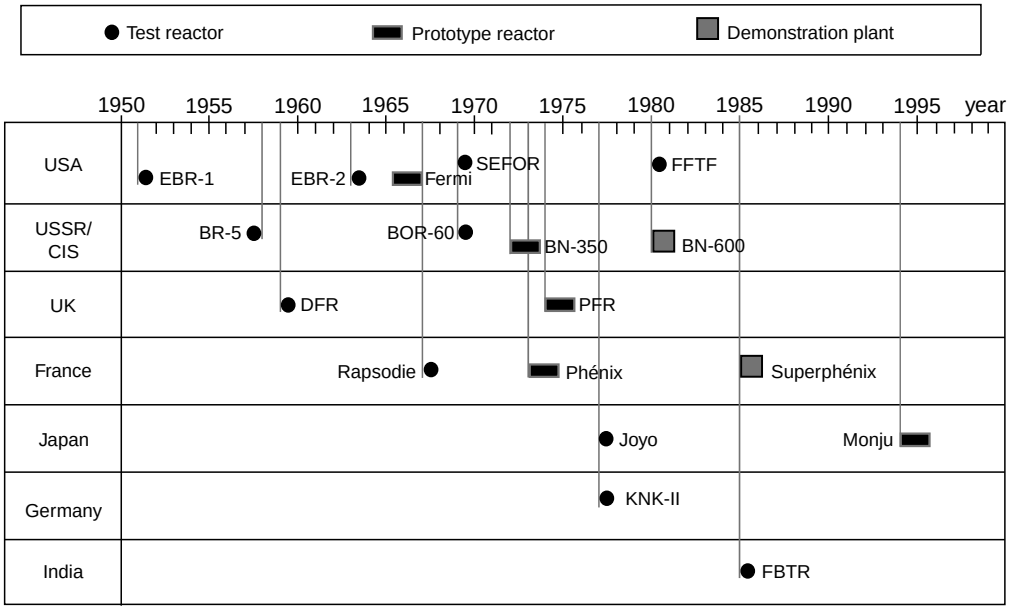
Reactor name	Country	Location	First criticality date	Stutdown date	Thermal capacity (MW)	Electric capacity (MW)	Fuel	Primary circuit configuration	Primary coolant	Primary coolant temperature (°C) In/Out
Clementine	USA	Los Alamos	1946	1953	0.025		Pu metal		Mercury	140/40
BR-2	CIS	Obninsk	1956	1957	0.1		Pu metal		Mercury	70/40
EBR-1	USA	Argo (Idaho)	1951	1963	1.4	0.2	U		Sodium/ potassium	
BR-5	CIS	Obninsk	1959	1971	5		PuO_2, UC		Sodium	450/375
BR-10			1971		10		MOX, UN	Loop		
DFR	UK	Dounray	1959	1977	72	15	U-Mo	Loop	Sodium/ potassium	350/230
EBR-2	USA	Argo (Idaho)	1963	1994	62	20	U-Zr, U-Pu-Zr			482/370
E. Fermi (EFFBR)	USA	Detroit	1963	1972	200	66	U-Mo	Loop	Sodium	427/268
Rapsodie	France	Cadarache	1966	1982	20/40		MOX	Loop	Sodium	510/404
BOR-60	CIS	Dimitrovgrad	1969		60	12	MOX	Loop	Sodium	550/360
Joyo	Japan	Oarai	1977(mark-I)		100 (Mark-II)		MOX	Loop	Sodium	500/370
FBTR	India	Kalpakhan	1985		40		(U,Pu)C	Loop	Sodium	518/400
KNK-11	Germany	Karlsruhe	1977	1991	58	21	MOX/UO_2	Loop	Sodium	
SEFOR	USA	Arkansas	1969	1972	20		MOX	Loop	Sodium	430/370
FFTF	USA	Hanford	1980	1994	400		MOX	Loop	Sodium	590/370
PFC	Italy	Brasimone	Aband.		125		MOX	Loop	Sodium	525/375
BN-350	CIS	Chevenko	1972	1999	1 000	150 and desalinisation	UO_2	Loop	Sodium	500/300
PFR	UK	Dounray	1974	1994	600	270	MOX	Loop	Sodium	560/400
Phénix	France	Marcoule	1973		560	250	MOX	Loop	Sodium	552/385
SNR-300	Germany	Kalkar	Aband. in 1991		770	327	MOX	Loop	Sodium	560/380
BN-600	CIS	Beloyarsk	1980		1 470	600	UO_2	Loop	Sodium	550/550
CRBR	USA	Clinch River	Aband. in 1983		975	380	MOX	Loop	Sodium	
Monju	Japan	Tseruga	1994		714	280	MOX	Loop	Sodium	529/397
Superphénix	France	Creys-Malville	1985	1996	3 000	1 240	1 240	Loop	Sodium	545/395
BN-800	CIS	Beloyarsk	Suspended			800	800		Sodium	550/350

Table 4.1b. **Main features of fast reactors which have been operated**

	Joyo (Mark II) Japan	**Phénix France**	**Monju Japan**	**BN-350 Kazakhstan**	**BN-600 Russia**	**Superphénix France**
CAPACITIES						
Thermal capacity (MW)	100	560	714	1 000*	1 470	3 000
Gross electric capacity (MW)	0	250	280	150	600	1 240
Net electric capacity (MW)	0	233	246	135	560	1 200
CORE						
Active height/active diameter (m)	0.55/0.72	0.85/1.39	0.93/1.8	1.06/1.5	1.02/2.05	1/3.66
Fuel mass (tHM)	0.76	4.3	5.7	1.17 ^{235}U	12.1 (UO_2)	31.5
Number of assemblies	67	103	198	226	370	364
Maximum power (kW/l)	544	646	480	–	705	480
Average power (kW/l)	475	406	275	400	413	280
Expected burn-up (MWd/t)	75 000	100 000	80 000	100 000	100 000	70 000 (first core)
FUEL						
Fissile material	MOX	MOX	MOX	UO_2	UO_2	MOX
Enrichment (%) first core	30 Pu	19.3 Pu	15/20 Pu_f	–	–	15.6 Pu
Mass of plutonium (t) first core						6
Enrichment (%) reloads	30 Pu	27.1 Pu	16/21 Pu_f	17/21/26	17/21/26	20 Pu
Mass of plutonium (t) reloads						7
Assembly renewal rate	70 days	3 months	20% of core every 5 months	80 efpd	160 efpd	100% of core every 3 years
Form	pellet	pellet	pellet	pellet	pellet	pellet
Number of pins per assembly	127	217	169	127	127	271
Assembly geometry	Hexagonal	Hexagonal	Hexagonal	Hexagonal	Hexagonal	Hexagonal
Average linear power (kW/m)			21	36		
Maximum linear power (kW/m)	40	45	36	48	48	48
Maximum clad temperature (°C)	650	700	675	700	620	620
Maximum temperature at centre (°C)	2 500	2 300	2 350	2 200		

* Real thermal capacity is 520 MW.

Figure 4.1. **Fast reactor programmes: start-up of reactors (first criticality)**



4.2.1.1 Fuels for FRs

From the outset, many types of fuel were tested: enriched uranium or plutonium in metallic, nitride, oxide or carbide form, or a mixture of plutonium and uranium oxides. It is worth noting that the past fifty years of fast reactor fuel development have witnessed changes in popularity of the various fuel types from the initial use of metal to emphasis on oxide fuels, then to ceramics (mostly oxide) and finally back to both oxide and metal, as performance demands and priorities have changed.

Because of fuel swelling at high burn-up and for compatibility with the cladding, pure metallic Pu/U alloy is no longer used. Today, fuels are essentially composed of ceramics obtained by sintering. The fuel most widely used at present is in the form of a mixture of plutonium and uranium oxides, $(U,Pu)O_2$ with up to 30% Pu-content (higher Pu-contents being problematic for aqueous reprocessing) (See Table 4.2). The fertile material is natural or depleted uranium oxide. It is located in the matrix of the fuel itself, and, as a breeder, in axial and radial blankets surrounding the core.

FR-MOX fuel manufacture is similar to the manufacture of MOX fuel for LWRs, including high-temperature sintering where the fuel is in the form of solid pellets, annular pellets or vibro-fuel. Major differences are that the fuel stack is housed in a steel cladding tube, and that pin clusters are placed within a hexagonal steel wrapper tube. Since the clad of the pins acts as a first barrier, it must be compatible with the fuel and the coolant (molten sodium) to guarantee mechanical strength and tightness for as long as possible. The fuel material has presented few limiting factors, even when performance targets have been extended by a factor of three. It has been the cladding material rather than the fuel itself which has had the greater influence. Stainless steel is the material that best meets these requirements today and, as demonstrated by various tests, allows to reach burn-ups of between 100 000 and 200 000 MWd/t.

Table 4.2. **Irradiation performance of MOX fuel in fast reactors (major achievements)**

Country or group of countries	Standard MOX fuel[1]		Experimental fuel		
	N°. of pins irradiated	Burn-up reached MWd/t	Maximum burn-up MWd/t	Main reactors[3]	Type of fuel[3]
Western Europe	265 000	135 000	200 000[2]	Phénix, PFR, KNK-II	Solid and annular pellets
United States	64 000	130 000	200 000	FFTF	Leading pins
Japan	50 000	100 000	120 000	Joyo	Solid pellets
CIS	13 000	135 000	240 000	BOR-60	Vibro-pac fuel
	1 800	100 000	–	BN-350	Solid and annular pellets
	1 500	100 000	–	BN-600	Solid and annular pellets

1. The distinction between "standard" and "experimental" fuel is not obvious. "Standard" refers to the bulk of fuel pins comprised in full sub-assemblies and irradiated without special management measures.
2. The figure of (approximately) 200 000 MWd/t of heavy metal corresponds to pins loaded in PFR.
3. This summary is not comprehensive. Neither all reactors, nor all fuel types are listed.

Table 4.3 lists the main manufacturing facilities, as well as the reactors that they have supplied. Manufacturing capacity was relatively low, and only the Cadarache plant in France had been designed for an industrial-scale capacity, i.e. that required for the Superphénix cores.

Table 4.3. **Fuel manufacturing facilities for fast reactors**

Country	Manufacturing plant	Capacity (tHM/y)	FR supplied by the plant
Belgium	Dessel, Belgonucléaire	5	SNR-300
France	Cadarache, COGEMA	20	Rapsodie, Phénix, Superphénix
Germany	Hanau, SIEMENS	10 (b)	KNK-II, SNR-300
Japan	Tokai-mura, PNC	10	Joyo, Monju
United-Kingdom	Windscale, BNFL	5	PFR
USA	Apollo, Babcock-Wilcox (ex: NUMEC)	5 (a)	FFTF
Russia	Chelabinsk, Paket at Mayak	0.3	BN-350, BN-600
	Dimitrovgrad, RIAR	1	BN-600

(a) Now dismantled.
(b) Now permanently shutdown.

Today, technological maturity has been attained based on mixed oxide for the fuel, fertile blanket of depleted uranium oxide (in the breeder mode), and stainless steel for the pin cladding and the assembly wrapper tube.

However, other avenues are still being explored, such as carbide or nitride fuels, as will be discussed below.

4.2.1.2. FR-fuel reprocessing

While the reprocessing of FR fuels makes use of the same process as is used for thermal reactor fuel (the PUREX process), a number of special factors must be taken into account: the presence of sodium, use of stainless steel cladding and structural components, high residual power, and high plutonium content.

While the experience gained is much smaller than that for reprocessing of thermal reactor fuels, France and the United Kingdom possess experience in reprocessing FR assemblies. In addition, lots of fuel from BOR-60 were also recycled. The FR fuel has been reprocessed either in specialised installations (e.g. the AEA plant at Dounreay in the United Kingdom, or the Marcoule site in France), or diluted with fuel from thermal power plants (La Hague, France). Part of the fuel from the Phénix and PFR prototypes was reloaded into the core after two successive reprocessing operations, thus demonstrating the complete fuel cycle.

4.2.1.3. Reactor coolant choices

Given the high power density, molten metals selected for their high thermal conductivity are used as coolant. This type of coolant allows the reactor to be operated at low pressure, thus reducing the probability of a loss-of-coolant accident. The liquid metal used in the first test reactors was mercury, but this was soon replaced by sodium (Na) which is common and cheap. Sodium melts at 98°C and boils at 880°C, giving it a wide service range. Its density at these temperatures is comparable to that of water, so that well-known pumping technology can be used. Yet, sodium presents certain drawbacks: at operating temperature, it ignites spontaneously in contact with air; also, it reacts violently with water, meaning that sodium-water reactions must be considered in designing the steam generators. Today, the technology for controlling these problems is well developed.

Another unfavourable effect is the activation of the sodium in the core. This entails the construction of a secondary, inactive sodium circuit which separates the active sodium from the steam generator, and hence a cost disadvantage. The primary circuit may be of the loop type (e.g. Monju) or the pool type, i.e. fully integrated in the reactor vessel (e.g. Superphénix). The latter concept allows all the active sodium to be confined in the main reactor vessel.

4.2.2 Current trends in FR technology development

4.2.2.1 Alternative coolant choices for fast reactor systems

Sodium has so far been universally adopted as the coolant for prototype and demonstration reactors; it has the desired, favourable heat transport and neutronics characteristics, is compatible with steel structural materials, and is available at a relatively low cost. On the other hand, it forms a radioactive activation product, ^{24}Na, and reacts chemically with water and air.

More recently, the difficulties with sodium reactions experienced at prototype and demonstration reactors have led to a renewed interest in alternative liquid metals which may have a number of advantages such as [63]:

- Increased economic competitiveness, if the plant can be simplified, e.g. by suppressing the intermediate heat transfer circuit.
- Increased inherent safety.
- Resistance to proliferation, e.g. through a very long reactor lifetime without refuelling.

Three developments are presently stimulating research on alternate liquid metal coolants:

- The opening of the nuclear sector of Russia, which gave access to information on the lead-bismuth eutectic (LBE) technology used in reactors for submarine propulsion [64,65], and projects for lead-cooled fast reactors [66,67].
- The programme on new and innovative technologies supporting the nuclear option in the 21st century launched by the US DOE ("Generation IV") [68,69] and also a similar programme in Japan [70,71], both including sodium, lead and lead-bismuth cooled fast reactors.
- The research on transmutation of nuclear waste in sub-critical, accelerator-driven reactors (the subject of this report): consideration of lead or lead alloy for spallation targets prompted their consideration as the reactor coolant, too.

Table 4.4. lists the advantages and disadvantages of Pb and Pb-Bi as compared with sodium.

Table 4.4. **Comparison of Pb(-Bi) versus sodium as reactor coolant**

Advantages of Pb and Pb-Bi	Disadvantages of Pb and Pb-Bi
Lead- and lead-bismuth-cooled cores have a **smaller positive void reactivity effect** (the positive void reactivity effect is a much-criticised feature of sodium-cooled fast reactors). A smaller positive void reactivity effect allows an ADS core to be operated at a higher k_{eff} and correspondingly **lower proton current**. **Lead**: In contrast to sodium, Pb is **not activated** in critical reactors (does not apply to an ADS in which Pb is activated by high-energy (n,p) reactions). High boiling temperature (1 743°C for Pb and 1 670°C for Pb-Bi vs. 880°C for sodium) implies **reduced potential for boiling-induced accidents**. Hot liquid Pb and Pb-Bi does not violately react with air. The **absence of a chemical reaction with water** may allow loop-type reactors to be designed with a simplified heat transport system. **Lead**: In the event of a hypothetical fuel melting accident, frozen lead may provide an **effective barrier against radiation and a radioactivity release** due to the high melting temperature (328°C for Pb vs. 98°C for sodium and 123°C for Pb-Bi).	**Lead-bismuth**: Neutron capture in ^{209}Bi produces the alpha emitter **^{210}Po**. **Lead**: The high melting temperature (328°C for Pb vs. 98°C for sodium and 123°C for Pb-Bi eutectic) implies an **increased potential for coolant blockage accidents**. Important functions in Pb and Pb-Bi cooled systems are jeopardised by **erosion and corrosion** (this item has been solved for sodium). In combination with the poor inspectability of liquid-metal cooled systems, this could pose significant **safety problems** (structure failures, blockages by sludge). The **high density** of Pb and Pb-Bi (or the associated high static pressure) – complicates the accelerator-reactor interface design; – calls for design measures against floating of core structures; – complicates the seismic design of the plant; – increases the probability of loss-of-primary-coolant accidents; – increases pumping power needs. In primary systems with natural circulation, the properties of Pb and Pb-Bi may make the **response of the system to heat balance disturbances very sluggish** (favours a start-up accident). **Less experience exists** than for Na as most FRs are sodium-cooled (Pb-Bi used in submarine reactors, Pb not used in any operating reactor).

Lead, as a coolant, has three important advantages over sodium: it boils at high temperature (1 743°C), does not react with water or air, and is not activated by fission neutrons. However, lead has

the disadvantages of high density and low heat conductivity, is corrosive for steels and has a high melting temperature (328°C) with the risk of freezing.

The lead-bismuth eutectic (LBE), which has been used as coolant for submarine reactors in Russia [64,65], can be operated at lower temperature (melting temperature 123°C, close to that of sodium), which improves, in comparison with lead, the compatibility with structural materials and reduces coolant freezing risks. However, bismuth produces the radioactive, volatile nuclide ^{210}Po ($t_{1/2}$ = 138 days) which becomes a radiological hazard in case of coolant leakage.

Other liquid metals are also possible coolants, but are less suited for large-scale application (e.g. mercury which is used as a target in pulsed neutron sources) or have never been used in reactors (e.g. tin).

Fast reactor concepts using gas coolants (helium, carbon dioxide) were studied in the past [72,73]. Such concepts are now being revisited, mainly because the transparent gas atmosphere facilitates in-service inspection and maintenance and the heat transport circuits can be simplified.

Table 4.5 lists the advantages and disadvantages of He and CO_2 as compared with liquid metals.

Table 4.5 **Comparison of gas versus liquid metals as reactor coolant**

Advantages of He and CO_2	**Disadvantages of He and CO_2**
The **steam entry reactivity effect** of gas-cooled fast reactors, which can be positive with normal fuels, is of **less concern** than the coolant void reactivity effect of liquid-metal cooled fast reactors. Gas-cooled fast reactors are **neutronically suited for transmutation** because minor actinides in the fuel have a beneficial influence on the steam entry reactivity effect which can be positive with normal fuels (the opposite is true for the coolant void reactivity effect of liquid-metal cooled fast reactors). Transparency of gas coolants **simplifies inspection** of internal structures. **Helium**: Is **inert and not corrosive**. **Heat transport system can be simplified** (no need for intermediate circuits, possibility to use a helium turbine). **Simplified handling of irradiated fuel** (no cleaning from rests of frozen coolants). **Experience available** from CO_2-cooled commercial thermal reactors (like Na, He has only been used in prototype reactors).	High coolant pressure implies **potential for loss-of-coolant accidents**. High pressure difference across reactor-accelerator interface constrains **design options** regarding e.g. the pressure vessel (choice of a steel pressure vessel of moderate size rather than the proven large pre-stressed concrete pressure vessel, which limits the power of the core). Lacking thermal inertia of unmoderated cores implies **fast transients** in off-normal conditions. Less favourable heat transfer characteristics imply a **limitation in fuel power density** and hence suitability for transmutation. High flow rates of hot dense gas induce dynamic loads and hence **noise and vibration**. **CO_2**: May require **gas chemistry and corrosion control**.

Helium is particularly attractive as it is a chemically and neutronically inert single-phase gas which is not activated. Disadvantages of gas coolants are the less favourable heat transfer characteristics and the higher operating pressure which requires depressurisation accidents to be dealt with in the safety analysis.

Finally, attention should also be drawn to new initiatives for using water as an advanced reactor coolant:

- In Japan [73], the usual BWR concept is being adapted towards a tight lattice fuel bundle and a higher void fraction, to obtain a breeding ratio of unity or slightly above.

- In Europe [74], an LWR operating in a thermodynamically supercritical regime, i.e. water enters the reactor as liquid and exits as high pressure steam without a phase change, is being studied.

These recent developments, based on LWR technology, are not especially aimed at actinide transmutation, but rather at improving resource utilisation and plant optimisation.

4.2.2.2 Perspectives for fast reactors

In essence, a major slow-down in FR development occurred in almost all OECD countries since the mid-1980s. In addition, in the past five years, R&D on fast-spectrum systems has shifted from new FR to ADS concepts. Nevertheless, FR concepts have been continuously studied and developed as, for example:

- The Self-consistent Nuclear Energy System (SCNES) in Japan.
- The Integral Fast Reactor system (IFR) in the USA.
- The European Fast Reactor (EFR) in Europe.

The following paragraphs will give an overview of the FR activities in some Member countries and briefly describe the respective developments.

Japan

Test and prototype reactors

The development of fast reactors in Japan has been based on Joyo and Monju. The experimental reactor Joyo, which went critical for the first time in 1977 with a Mark-I core at an initial power of 50 MWth (later increased to 75 MWth), was equipped in 1982 with a Mark-II core of 100 MWth. This core, in which the blankets are replaced by stainless steel reflectors (to become a burner), has been operated for 35 duty cycles to test fuels and materials. Since June 2000, the reactor is being upgraded to the Mark-III core, that will provide enhanced irradiation capabilities (about 30% higher neutron flux, increased plant availability, upgraded irradiation technology). The maximum fuel burn-up achieved so far in Joyo is 71 000 MWd/t (fuel pin average).

Monju, a prototype reactor of 714 MWth (280 MWe), went critical for the first time in April 1994, and supplied electricity to the grid for the first time in August 1995. The first core of Monju was loaded with MOX fuel at a plutonium enrichment of 20 and 30% in the inner and outer core zones, respectively. At equilibrium, one fifth of the core fuel is to be discharged at each refuelling; the

discharged burn-up will reach 80 000 MWd/t (average). The successive core loads of Monju are planned to consist of the same MOX type fuel. Higher burn-up values will progressively be attempted.

In December 1995, during pre-operational testing at 40% power, Monju experienced a sodium leak in the secondary, inactive sodium circuit, caused by a rupture of a temperature detector. Since then, the reactor is shut down for repair work. After authorisation to proceed with the design modifications, at least 4 years are needed before plant operation can be resumed.

Self-Consistent Nuclear Energy System – SCNES

In Japan, a self-consistent nuclear energy system (SCNES) [75-80] has been defined as a system satisfying four objectives, i.e. energy generation, (fissile) fuel breeding, confinement of minor actinides and radioactive fission products and, last but not least, guaranteeing nuclear safety. The term "self-consistent" means that even if the system produces materials dangerous to human beings and the environment, it can eliminate these materials within the system itself. The potential of large fast breeder reactors with MOX, nitride and metallic alloy fuels has been studied in relation to nuclear safety and their ability to transmute radioactive nuclides .

Some long-lived fission products (LLFP) with relatively large capture cross-sections, namely ^{79}Se, ^{99}Tc, ^{107}Pd, ^{129}I, ^{135}Cs and ^{151}Sm, are selected for transmutation, based on their having effective half-lives of under 10 years, and the others are confined in the system. The LLFPs can be transmuted in the radial blanket and part of the axial blanket regions. In order to enhance transmutation efficiency, neutrons in the radial blanket region are moderated by solid hydride ZrH1.6. Minor actinides are recycled and transmuted as a fuel and can be confined to the system without any significant impact on nuclear and safety characteristics. The hazard index level of the LLFPs per ton of spent fuel from the SCNES after 1 000 years is as small as that of a typical uranium ore.

In case of a metallic fuel, a slug of U-TRU-10%Zr alloy with a few percent MA would be used in a homogeneous recycling scheme. The proposed reprocessing method for this metallic fuel would be a sequence of electro-refining, retorting, filtration, electro-migration and finally injection casting. Metallic technetium would be precipitated at the bottom of the cadmium pool in the electro-refining cell while metallic palladium is dissolved in the pool. Selenium would be recovered from the off-gas system in the pre-treatment process. Finally, iodine could be recovered as NaI in the pre-treatment process before electro-refining where caesium remains as chloride in the electro-migration process for salts.

A nitride fuel actinide recycle system coupled with nitride FBRs and pyrochemical reprocessing was also investigated in order to establish a confinement and transmutation system for long-lived radioactive nuclides. The results of these studies are summarised as follows:

- The use of nitride fuel permits an excellent fast reactor core performance; i.e. small or negative void reactivity and nearly zero burn-up reactivity changes.
- A transmuter with 5 wt% MA-content can support more than 6-10 units of PWR spent fuel a year. After several recyclings, the MA compositions almost reach equilibrium, except for ^{246}Cm.
- The toxicity of ^{14}C produced by using ^{14}N (natural nitride) becomes almost equivalent to that of americium and curium after 5 000 years, and the toxicity for 90% enriched ^{15}N is similar to that of FPs after 1 000 years. Therefore, the recovery of ^{14}C becomes a very important issue to which pyrochemical reprocessing could be a solution.

- The pyrochemical process can be adapted to reprocess nitride spent fuel. Evaluation of the process shows that actinides are reasonably well separated from fission products, and that the high level wastes are nearly actinide-free.
- Preliminary studies for the plant design also showed that the fuel cycle cost of this coupled system could be substantially reduced by employing pyrochemical reprocessing, owing to its simplicity and compactness.

France and Western Europe

Prototype and demonstration reactors

Research work has been on the way for nearly 30 years, not only in France, but also in the neighbouring European countries Belgium, the Netherlands, Germany, Italy, and the United Kingdom. Work, centred on DFR, KNK-II, SNR-300 and PFR has now been terminated, while more generic R&D work is still being pursued in some laboratories (especially linked to plutonium incineration). The plan is to build a European Fast Reactor (EFR) as a successor to Superphénix, but the respective decision has now been postponed to 2010 or later.

As for the 250-MWe Phénix prototype, plans are to restart it and to operate it for another 6 irradiation cycles. Phénix had been shut down from 1990 to 1994, and again from 1995 up to now, first to take measures to avoid unintended reactor shut-downs as observed in 1989 and 1990, and secondly to refurbish the secondary sodium circuits. However, the maintenance, inspection, and repair work proved to be much more extensive than originally planned, with the safety authority asking, among other requests, for seismic upgrade measures, repairs to all the steam generators, visual inspection of the upper internal structures of the reactor block, and ultrasonic inspections of the welds of the reactor core shell. Upon completion of the renovation work, resumption of operation at 2/3 nominal power is planned for the summer of 2002. At this power level, the planned 6 irradiation cycles would correspond to a period of approximately 5 years which would keep the reactor in operation until 2008.

The major achievements on MOX fuel in the prototypes Phénix and PFR can be summarised as follows:

- Pin cladding materials consisting of austenitic steels and nimonic alloys have been optimised and tested in high burn-up irradiations at PFR to more than 135 000 MWd/t for full sub-assemblies. Average burn-ups above 180 000 MWd/t have been achieved with experimental fuel.
- Ferritic wrapper tubes, virtually non-swelling under the impact of fast neutrons, have been developed to accommodate these high burn-ups.

The 1 200 MWe Superphénix (SPX) plant, owned by the European electricity utility NERSA, a joint venture between utilities from six countries, had been restarted in 1994 after a four year interruption initiated by a pollution of the primary sodium, which was followed by a public hearing. It was then re-licensed to be progressively converted from a plutonium breeder to a burner, following the recommendation of a governmental commission (the so-called Curien Commission). To that end, steel reflector assemblies have been fabricated to replace the radial, fertile blanket. Three test assemblies have also been manufactured: two of CAPRA type, differing in the origin of their plutonium, either from first or second generation, and one NACRE assembly containing 2% neptunium added to the usual MOX, as part of the SPIN programme.

During 1996, Superphénix went critical for 266 days, i.e. 95% of the scheduled operation time. The reactor was shut down at the end of December, having reached 320 equivalent full power days, which corresponds to a burn-up of 35 000 MWd/t (maximum) for the core.

In 1997, the decision by the French government, confirmed in February 1998, to definitely shut down Superphénix, lead to a marked reorganisation of the fast reactor programme in France. Concerning the investigations on enhanced TRU burning, a partial redeployment in Phénix of former Superphénix experiments is planned.

The European Fast Reactor collaboration

The European Fast Reactor (EFR) collaboration was established in 1988 when the participating organisations launched a program of design and validation activities, which was pursued for ten years. During the first two-year phase – the Conceptual Design phase – the best features of the national commercial fast reactor projects were integrated into a compact EFR "first consistent design" along with alternative and fallback options. This was followed by a three year Concept Validation phase in which the system engineering for EFR was completed and the R&D results were integrated into the design.

The initial major objectives set by the utilities for the EFR were:

- An up-to-date safety standard, comparable with that of future LWRs, and licensability in the participating countries without significant design changes.
- Potentially competitive electricity generating costs compared with future LWRs.

These objectives were supplemented by the recommendations which resulted from the 1993 safety and economic assessments, the feedback of Superphénix operating experience, and the prospects for new missions for fast reactors in the nuclear fuel cycle. As a result, emphasis was placed on:

- Demonstration demonstrating that a high load factor could be achieved in combination with ambitious operating and safety standards, notably with consideration of severe accidents in the containment design.
- Progress on provision for in-service inspection and repair.
- Flexibility regarding the missions in the fuel cycle and in particular possible integration of CAPRA core designs into the Reference EFR.

To meet the goal for economics it was essential, in addition to minimising the plant investment cost, to produce a design which would ensure both high plant availability and a lifetime target similar to that for future LWRs (possible extension to 60 years). This necessitated special attention to components and structures where failure would lead to prolonged outage for repair (of which the permanent reactor structures, the heat exchangers and the steam generators are particularly important), and to developing efficient in-service inspection and repair methods. The approach adopted was to use, as far as possible consistently with the other requirements, technologies which were already verified or which could be expected to be fully endorsed by R&D. Considerable attention was given to the development of well founded and validated design rules.

EFR safety approach

A prime feature of the safety design of EFR is the extensive application of "defence-in-depth" principles. The successive protection levels include:

- Careful selection of appropriate materials, sound basic design backed up by extensive R&D, strict application of quality assurance procedures.
- The provision of systems to detect failures or deviations from normal operation and to prevent such failures from escalating into fault conditions.
- Protective systems and engineered safety features incorporated into the design to cope with classical and other initiators.
- Preventing failures of equipment or human error from leading to accidents.
- Providing several sequential physical barriers to prevent any hazardous radioactive release to the environment.
- Ultimate risk minimisation measures to enhance further the reliability of shut-down and decay heat removal and the retention capability of the containment.

Because the reactor is not pressurised and is surrounded by a close-fitting guard vessel, uncovering of the core due to loss of coolant is precluded and measures to prevent core melting are concentrated on enhanced shut-down and decay heat removal. Through these preventive measures the risk of core melting is reduced to an extremely low level and beyond the objectives generally set for future reactors. Nevertheless, according to the most demanding safety requirements, the prevention level is supplemented by a mitigation approach in which consequences of core melting are considered in the design of the containment system.

EFR core design

The EFR core design has been optimised for safety through:

- The choice of fuel pin linear rating, dictated by the prevention of local fuel melting in case of inadvertently withdrawing absorber rods withdrawal.
- The choice of core height to minimise the positive reactivity effect of sodium voiding.
- The Doppler coefficient to provide efficient reactivity feedback in rapid transients.
- The use of annular fuel pellets to prevent escalation of core accident sequences involving fuel melting.

Recent progress has been made, in the framework of international R&D agreements, on developing advanced computer codes for assessing the behaviour of optimised fast reactor cores, like the EFR core, under extreme conditions (core disruptive accidents). Applying these codes has shown that the consequences are benign, and the level of energy liberated is small compared with the strength of the primary system boundary.

Integrating a plutonium and minor actinide burning core can lead to two EFR variants (breeder and burner) to avoid compromising either as an economic power generator, but it is desirable that the differences should not be extensive. In the first CAPRA feasibility studies, this desire to accommodate a breeder or burner core with only minor adaptation of the reactor was demonstrated to be achievable. A burner core with the same power, core envelope and absorber rod arrangement as for EFR was proposed, but with a different pin diameter and fuel subassembly pitch to improve the plutonium burning efficiency. The diagrid and above-core structure therefore have the same overall dimensions for both breeder and burner cores, but the detailed geometry has to be adapted to the sub-assembly pitch, giving an otherwise identical reactor based on the same technology.

United States

Past achievements

Between 1953 and 1994, several experimental fast reactors were operated in the USA (see Table 4.1a). Two of these, the Experimental Breeder Reactor 2 (EBR-2) in Idaho and the Enrico Fermi Fast Breeder Reactor (EFFBR) in Michigan, achieved significant electrical outputs (20 MWe and 66 MWe, respectively). The latter suffered a fuel melting incident and was finally shut down in 1972. The Fast Flux Test Facility (FFTF, 400 MWth) started in 1980 with a mixed oxide fuelled core and reached its burn-up target of 100 000 MWd/t for a full core load in 1987. The US fast reactor programme came to a halt in 1994 when the government decided to shut down EBR-2 permanently, to put FFTF in standby conditions, to cancel the Clinch River Breeder Reactor (CRBR) project, and to abandon the Integral Fast Reactor (IFR) project (see below) [81]. At the end of 2001, the US Department of Energy finally announced its decision to terminate also FFTF.

The IFR concept, a new fast reactor initiative promoted by Argonne National Laboratory, is based on a fuel consisting of a ternary alloy of uranium, plutonium and zirconium. This ternary alloy was conceived as a successor to the electrometallurgically reprocessed alloy which had already been recycled in EBR-2. Such alloys remain compatible with the steel cladding up to high burn-up values. The concept was integrated by General Electric into a full plant design called PRISM (Power Reactor Innovative Small Module), consisting of nine reactor modules contributing 135 MWe each. The PRISM design was subsequently modified to raise the electric output per module to about 300 MWe (PRISM Mod. B). All IFR designs were based on full actinide recycling using an electrometallurgical processing plant co-located with the reactor complex.

The IFR concept

The integral fast reactor (IFR) was under development by the US Department of Energy for the decade between 1984 and 1994 [3,82]. Technology development was carried out at Argonne National Laboratory, and an industrial team led by the General Electric company utilised the technology for a specific nuclear power plant design, the Advanced Liquid Metal Reactor (ALMR).

The IFR fuel cycle[36] consists of a fast-spectrum nuclear reactor using liquid metal (sodium) cooling and metallic alloy fuel, coupled with recycle technology based upon an electrometallurgical partitioning of fission products from actinides. The actinides are combined with make-up uranium feedstock and multiply recycled through the reactor for total consumption by fission while the fission products are stabilised in waste forms suitable for long-term disposal.

IFR recycle technology

The recycle technology development was designed to meet two key requirements, i.e.:

- To retain all TRU intimately admixed during every recycle step back to the reactor (alternately, the TRU recycle product was not required to be free from fission products because the minor actinides already made remote handling a necessity).
- To achieve a fission product waste stream essentially free from TRU.

The first key requirement – that of keeping the TRU always intimately mixed – was to assure that the IFR fuel cycle introduced no diversion or proliferation vulnerabilities not already present from the

36. See fuel cycle scheme 5 in Chapter 3.

LWR once-through fuel cycle. The higher actinides (primarily americium and curium), which are intimately mixed with plutonium throughout the recycle, not only serve as fuel contributing to the efficient use of uranium resources, but together with the residual fission products they and the higher plutonium isotopes render the recycle material compositionally unattractive and radioactively unapproachable as is LWR spent fuel. The second requirement was to assure not only that all actinides were recycled to the reactor for total fission (resource utilisation) but also that the fuel cycle waste stream is comprised solely of fission products that would decay to radiotoxicity levels as small as that of the original ore during a time period of only 300 to 500 years. Such a period would be short enough for engineering and institutional measures to assure waste isolation with a high degree of confidence until the hazard had decayed away (waste, environment).

The electrometallurgical recycle technology utilises an electrolyte bath of lithium and potassium chlorides above a cadmium-pool, both at 500°C. Chopped fuel pins are anodically dissolved out of the steel cladding and into the electrolyte at a few volts; the reactive FPs and bond sodium remain in the electrolyte, the noble metal FPs and cladding hulls remain as solids in the chopped-pin basket or dissolve in the lower cadmium-pool, and the uranium is deposited on a solid cathode. All TRU and some lanthanide fission products deposit in a separate liquid cadmium-cathode. The chemical free energies of the chlorides are such as to guarantee that all TRUs co-deposit to avoid the possibility of a pure plutonium product. The uranium and TRU cathodes are removed from the electro-refiner and separately retorted at about 1 300°C to recover entrained electrolyte salt and cadmium for recycle to the electro-refiner. The resulting actinide ingots are blended with a make-up of depleted uranium and zirconium alloy and are injection cast at about 1 300°C into fuel slugs which are sodium bonded inside steel cladding for recycle to the reactor.

The radioactivity of the recycled products requires that all recycle process steps occur behind heavy shielding. This shielding requirement motivated the selection of simple and compact technologies for recycle and refabrication in order to minimise the volume and associated construction costs of hot cell facilities. Electrometallurgical processing and refabrication of metallic alloy fuel by injection casting were selected in part for the sake of the large reduction in criticality-limited process equipment and hot cell volumes attainable in the absence of a neutron-moderating aqueous matrix that would reduce critical masses by a factor of 50. Sodium bonding of fuel to the clad allows for loose dimensional tolerances on the cast pin slugs, and the fast neutron spectrum allows for loose compositional tolerances on fission product carry-over from electrometallurgical processing for recycle back into the reactor. Such loosening of tolerances facilitated remote processing.

IFR fuel technology

The IFR fuel is a metal alloy of 10 wt% zirconium, 15 to 25 wt% recycled TRU, and the remainder of depleted uranium. The zirconium-content was selected to achieve a high enough solidus (1 180°C) for operation at reactor outlet temperatures of 590 to 600°C but a low enough liquidus (1 300°C) to facilitate the injection casting fabrication process. The fuel slugs are sodium bonded inside austenitic or ferritic stainless steel cladding. Smear density (area ratio of fuel slug to inside clad) is set at 75% so as to allow for fission gas induced porosity in the fuel slugs to interconnect by the time the fuel has swelled radially to contact the clad at about 1.5 atom% burn-up. The interconnected porosity provides an escape route for fission gas to the (upper) gas plenum, terminates radial swelling at 30 v%, and thereby precludes mechanical interaction between fuel and clad driven by fission gas induced swelling.

An extensive fuels irradiation program on uranium-zirconium and uranium-plutonium-zirconium IFR fuel was conducted from 1983 to September 1994; it verified favourable fuel pin performance with peak burn-ups up to 20 atom% (200 MWd/kg) at linear heat rates of up to 500 W/cm. Transient testing – both operational tests and accident tests to fuel pin disruption – established a database of

properties for use in safety and licensing activities. A safety case was developed to “license” the conversion of the EBR-II core loading to IFR recycled fuel assemblies (containing fission product carry-over), but the IFR programme was terminated before electrometallurgically - recycled fuel could be reintroduced into EBR-II.

IFR passive safety technology

The metallic alloy fuel form not only facilitates a compactness and simplicity in the recycle equipment and process, but yields safety benefits as well. The absence of low-mass-number scattering elements in the alloy and its high density combine to facilitate core layout designs of minimal reactivity loss upon burn-up, thereby passively precluding opportunity for transients induced by control-rod-run-out. The high thermal conductivity of the metallic fuel maintains a small temperature increment of the fuel above the coolant, which in turn minimises stored energy and stored (Doppler) reactivity. These effects combine to counter passively transients due to loss of coolant pumping or loss of heat sink – even in the absence of scram action. Finally, the relatively low melting point of the fuel in relation to the clad creep rupture and eutectic interaction temperatures, and in relation to the sodium boiling point, combined with the homogeneously distributed fission gas in the fuel, together provide a fast acting “fuse” fuel dispersing mechanism which squelches reactivity addition in severe accident situations and precludes the possibility of reaching super- prompt-critical conditions or vapour explosions. This results in minimal energy deposition before dispersal, avoids energetic loads on the vessel, and passively promotes retention within the vessel even under severe accident scenarios.

Removing decay heat by passive means in IFR reactor designs rests on retaining the coolant inventory by the use of sodium cooling at atmospheric pressure and double top-entry tanks containing all of the primary heat transport equipment and fluid. Natural convection carries decay heat from the fuel clad to dedicated secondary circuit decay heat removing loops driven by buoyancy flows and operating continuously at <1% rated power. The heat capacity of the primary coolant in the primary circuit tank is large enough to absorb within safe temperatures the initial excess of decay heat over the capacity of the passive secondary heat removal channel.

These passive safety properties make it possible to remove all safety functions from the balance of plant equipment, and thereby to reduce their cost in fabrication, installation, and maintenance.

IFR waste technology

Transmutation and fission products created from the uranium feed stream but unsuited to recycling as fuel are destined ultimately for a geological repository, and the ecological and safeguards implications of waste management are an important element of IFR technology development. The IFR design objective of multiple recycle for total consumption of the uranium feed stream, which results in a waste stream essentially free of TRU, was motivated not only by the goal of resource conservation but also by the desire to minimise the ecological and safeguards risks associated with the waste from the IFR fuel cycle.

The electrometallurgical recycle technology is designed to discharge less than one part per thousand of the recycled TRU fuel into the waste stream destined for the repository. Noble metal fission products from the IFR cycle are to be incorporated as alloying elements in an iron-zirconium metallic alloy. Active FPs are to be captured in a zeolite ion exchange matrix which is subsequently blended and glass bonded into a ceramic/glass monolith. These waste forms for FPs from the IFR fuel cycle had been selected and fabrication technology development and waste form characterisation was underway but uncompleted in September 1994 when the programme was cancelled.

The virtual absence of actinides from the waste totally eliminates long- and short-term risks of proliferation or criticality from IFR waste disposal. Similarly, the approach for minimising the radiotoxicity risk associated with IFR wastes is straightforward: with transuranics eliminated from the waste, the radiotoxicity source term exiting the fuel cycle derives only from long- and short-lived fission products. The hazard from the LLFPs is broadly comparable with that of the radiotoxicity that was removed from the earth with the ore that was mined to make the fuel in the first place [83]; the radiotoxicity hazard from the short-lived FPs decays to a level below that of the LLFPs within 500 years.

IFR development status

At the time the IFR programme was terminated (September 1994), the fuels irradiation program had demonstrated excellent steady-state and transient performance of the metallic alloy fuel, sodium-bonded to either austenitic or ferritic steel clad at heat ratings up to 500 W/cm and for peak burn-up up to 20% (200 MWd/kg) at TRU/heavy metal enrichments up to 25%. Passive safety performance had been demonstrated at the EBR-II power plant, with benign consequences upon loss of pumping action without scram and loss of heat sink without scram; both tests were performed from full power with absolutely benign consequence and immediate reactor restart to full power. The injection casting fuel fabrication technology was fully developed and the electrometallurgical processing technology had been demonstrated at the industrial (10 kg/batch) scale with all actinides and with surrogate (non-radioactive) FPs. The cathode consolidation processing step had been partially demonstrated at industrial scale. A full set of industrial-sized recycle/refabrication equipment had been designed, built, installed in the recycle hot cell, and checked out. The processing technologies for reducing LWR spent fuel were still under bench scale development, while developing fabrication technology and for the waste form and testing its properties were just getting started. The GE-led industrial team had completed advanced conceptual design of all NPP systems and had just initiated conceptual design for the recycle facility. The USNRC had extensively reviewed the safety of the ALMR and had formally issued a pre-licensing safety evaluation report (SER).

After September 1994, EBR-II was de-fuelled and lay-up activities were started. The GE-led industrial work was discontinued. The equipment which had been built and installed in the recycle hot cell was redirected to the task of electrometallurgical treatment of 100 EBR-II spent fuel assemblies so as to demonstrate a capability to produce fission product, uranium, and TRU products suitable for long-term disposal. (Untreated sodium bonded EBR-II spent fuel does not meet acceptance criteria for the geological repository.) The work on FP waste forms was continued for application to the EBR-II spent fuel treatment, and the electrometallurgical process technology development was continued for treating selected DOE-owned spent oxide fuel.

Electrometallurgical treatment activities on spent EBR-II fuel have recently confirmed that the full spectrum of fission product elements present in the EBR-II spent fuel are indeed separable from the uranium in industrial-scale, remotely operated and maintained electrometallurgical and cathode processing equipment.

Other countries

Commonwealth of Independent States

In the Commonwealth of Independent States (the former USSR), BN-600 in Russia is operated successfully, and BN-350 in Kazakhstan was finally shutdown in 1999 after 27 years of service following a government decision. Both reactors have basically been fuelled with enriched UO_2; plutonium test sub-assemblies containing so far more than 3 000 fuel pins have been successfully irradiated up to 100 000 MWd/t. This fuel, based on pellet technology, was produced at the Mayak

plant at Chelyabinsk. The design lifetime of BN-600 (30 years) expires in 2010. The development of a lifetime extension program till 2020 will start in 2001.

In parallel, the BOR-60 experimental fast reactor at RIAR (Research Institute of Atomic Reactors), Dimitrovgrad, has been loaded with, among other advanced fuels, both pellet and vibro-packed MOX fuel, including fuel with a high plutonium content in standard and advanced claddings, manufactured on site up to an annual production capacity of one tonne of granulated fuel. Thus BOR-60 was able to recycle its own plutonium.

Based on this experience, construction has started on:

- One 800 MWe fast reactor, BN-800, at the Beloyarsk site.
- A large MOX fuel manufacturing plant at Mayak RT-1 (Complex-300).
- A new RT-2 facility to store and reprocess spent fuel from civil reactors and to fabricate MOX fuel.

However, financial difficulties have led to delays over the whole construction programme. Nevertheless, according to the "Programme of Nuclear Power Development in the Russian Federation for the 1998-2005 Period and up to 2010" [84], the construction of the BN-800 power plant is to be completed by 2010 at the Beloyarsk site. Meanwhile, the BN-800 core was redesigned in order to increase plutonium consumption, ensure a negative void coefficient, and reduce costs.

The current situation regarding the Russian R&D activities in the field of fast reactors can be summarised as follows:

- Developing the hybrid core design for the BN-600 reactor, and first design studies of a full MOX core (both the traditional fuel pellet, and RIAR's vibro-packed fuel are being studied). Irradiation tests in BN-600 with experimental sub-assemblies including both MOX fuel types are planned.
- Justifying lifetime extension for BR-10, BOR-60 and BN-600.
- Studies advanced, high safety fast reactor designs, including a large (~1 600 MWe) sodium-cooled fast reactor, and designs with alternative coolants (e.g. lead).
- Developing the basic design of the BREST-300 (lead-cooled) demonstration fast reactor with closed fuel cycle facilities located at the reactor site, and its experimental justification (the Beloyarsk site is considered to be a candidate site for BREST-300).
- Design studies for the large reactor BREST-1200.

India

In India, mixed carbide (U-Pu)C fuel was loaded in the Fast Breeder Test Reactor (FBTR) near Bombay, and the first core of a very low power attained criticality in 1985. The second core should reach the power of 40 MWth; it requires about 200 kg of (Pu 0.55, U 0.45)C fuel pellets. The peak burn up in the fuel reached 71 170 MWd/t. Post-irradiation examination of the fuel sub-assembly discharged at 50 000 MWd/t peak burn up indicates that the gap between fuel and clad is still not closed, and that the fuel is in excellent condition. A revised target peak burn up of 100 000 MWd/t has consequently been set. Detailed design, R&D, manufacturing technology development and the safety review for the 500 MWe Prototype Fast Breeder Reactor (PFBR) are ongoing.

China

In China, the Chinese Experimental Fast Reactor (CEFR) is the first step in the development of fast reactor technology. CEFR is a sodium-cooled pool type reactor; it has a thermal power of 65 MW and is equipped with a 25 MWe turbine generator. After completing the conceptual design and preliminary design in 1993 and 1997, respectively, some additional safety analyses were performed (mainly with regard to sodium spray fires). Presently, the CEFR's reactor building is under construction. The main components, including the reactor block, primary and secondary circuits, and the fuel handling system have been ordered. First criticality of CEFR is scheduled for the end of 2005.

4.2.3 P&T-related specific aspects of fuels and coolants

4.2.3.1 Fuels with increased concentrations of minor actinides

The fuel types of interest for P&T are the same as those which have been used or proposed for conventional reactors, i.e. oxide, nitride, and metal. The ranking of these fuels by melting point and heat conductivity is oxide >nitride >metal and metal >nitride >oxide, respectively.

Nitride fuel is particularly suited for transmutation applications because different MAs can coexist in the fuel. Highly concentrated ^{15}N, which does not exist in significant quantities naturally, will be used in order to control formation of long-lived radioactive ^{14}C; ^{15}N must be recovered in the fuel during reprocessing. Basic data on the thermal properties of MA nitrides necessary for fuel design were obtained. It was confirmed that the particle size of TRU nitrides can be adjusted by means of carbothermic reduction and that very fine uranium nitrides can be produced via the sol-gel process. In addition, burn-up tests of mixed uranium-plutonium nitride fuel produced on a trial basis showed that fuel integrity can be maintained up to a burn-up of at least 5.5%. However, it will be necessary to accumulate irradiation data for MA-nitride fuels, devise measures to deal with fuels with very high decay heat, and develop an economical way to produce enriched ^{15}N. For this purpose, MA-nitride fuels will be produced on a trial basis and irradiation experiments will be carried out.

One FR option has chosen fuel with MAs mixed into conventional mixed-oxide fuel. This extension of conventional fuel makes use of what has been learned in past FR studies. In this case, MAs in the fuel are limited to some 5% in order to maintain the integrity of the fuel and avoid adverse effects on the neutronic characteristics of the core. Nuclear data on MA nuclides were measured and evaluated, and design studies were carried out to determine the acceptable amounts of MAs and rare-earth elements in the fuel. Immediate issues include improving the accuracy of the nuclear data and physical properties of MAs, evaluating the behaviour of MA fuel under irradiation, and developing the technology to produce MA fuel industrially.

Another possibility is the use of metallic fuel – a ternary alloy of uranium, plutonium and zirconium (U-Pu-Zr) – which would be suitable for dry reprocessing, and would help simplify the fuel production process. For the electrorefining process, an important step in the dry reprocessing, feasibility has been confirmed through joint international research and is at the stage of engineering experiments, but the feasibility of the process for reducing oxides, and of technology to treat spent salt, has still to be confirmed. Immediate issues include compilation of data on fuel behaviour based on irradiation experiments, and development of injection-casting technology for fuel production.

4.2.3.2 Effect of fuel and coolant choice on transmutation characteristics

As discussed in Chapter 2, the amount of TRU or MAs transmuted in an actinide burner with a closed fuel cycle (FR or ADS) is proportional to the fission energy released by the system. The choice

of coolant and fuel, however, influences the engineering design of the transmuter and the overall feasibility of the system. A study was recently conducted in Japan (see Annex E) to evaluate the effect of the choice of coolant and fuel on the transmutation characteristics (i.e. the minor actinide balance) of different fast reactor cores.

Since it is difficult to make such a comparison on the specific effect of coolant alone, three realistic reactor designs were chosen:

- A sodium-cooled fast reactor of commercial size.
- A lead-cooled reactor of the BREST-300 type.
- A CO_2 gas-cooled reactor of ETGCFR type.

The transmutation characteristics of the respective cores were compared by normalising the MA mass balances of the cores to the thermal power produced.

As gas-cooled reactors have a particularly hard neutron spectrum, it could be speculated that the gas-cooled core might have more favourable MA transmutation characteristics than the sodium- and lead-cooled cores. However, the study showed that, after normalisation, the three cores performed almost identically.

The same study also assessed the influence of the fuel type on the transmutation characteristics. Here, a 1 000 MWe sodium-cooled fast reactor was taken as reference case. The following alternative fuel types were compared with the reference $(U,Pu)O_2$ fuel:

- $(U,Pu)^{15}N$ as nitride fuel.
- U-Pu-10Zr as metal alloy fuel.

The respective analysis indicated that the transmutation characteristics of the nitride and metal cores are similar and slightly better than those of the oxide core. The difference can be attributed to the harder neutron spectrum of the alternative fuel-type cores. However, in terms of MA mass transmuted per year, the difference is rather insignificant.

The study thus confirms that the overall transmutation characteristics are not sensitive to the choice of coolant and fuel-type.

4.3 ADS technology

4.3.1. Introduction

Active projects for ADS systems exist in France, Italy, Japan, Korea, USA, and several other European countries. Research in these countries mainly comprise basic studies on the different aspects of an ADS, although some of these projects aim towards a pre-engineering design phase within the next few years. International collaboration is emerging in Europe (Technical Working Group), in the USA (ANL and LANL) and new co-operative arrangements have been established between France and USA in this domain. An overview of these activities is given in Annex F and will also be mentioned in Chapter 7.

As the R&D on ADS relies essentially on two distinct disciplines, i.e. reactor physics and accelerator physics, some specific efforts have been launched, for instance by OECD/NEA, in order to exchange information between both communities. Today, some discussion is beginning on the

viability of developing a dedicated accelerator for ADS applications whereas there would be some scope to pursue multi-purpose accelerators first (see also Chapter 7 on R&D needs).

In the following, the discussion of the ADS technology is divided into three sections dealing with the sub-critical reactor, the spallation target, and the accelerator.

4.3.2 Sub-critical reactor aspects

Both the evolutionary and innovative transmutation approaches which incorporate accelerator-driven systems call for sub-critical cores with a fast neutron spectrum and fuels dominated by TRU or minor actinides. As pointed out before, these cores are characterised by a very low fraction of delayed neutrons and by a low (or near zero) Doppler reactivity coefficient. In principle, the physics of the ADS and of its sub-critical core is well understood, and there are several publications which deal extensively with the subject [85,86]. However, several concepts are new and their understanding requires experimental validation.

The following sections focus on a description of the basic physics phenomena in the sub-critical multiplying core, with reference to the coupling phenomena and their impact on the sub-critical core (SC), and discuss how the sub-criticality helps to reduce (or to eliminate) the negative consequences of impaired core characteristics on the safety of the multiplying medium. The areas which need particular care for experimental validation will be indicated, and some ongoing experimental programmes will be quoted. Finally, some relevant design-oriented problems of sub-critical cores and their integration into an ADS will briefly be indicated.

4.3.2.1 Neutron flux distribution

In a critical system, the condition of balance of neutron production and consumption at each point of the phase space $(E, \bar{r}, \overline{\Omega})$ is expressed by the Boltzman equation, which can be expressed in matrix form:

$$A\overline{\varnothing} = P\overline{\varnothing} \tag{1}$$

where A is the "consumption" and P the "production" operator, and $\varnothing$ the flux vector.

In the same system, made sub-critical, the condition to have a stationary state is to have an external source $S(E, \bar{r}, \overline{\Omega})$ such that, e.g. the Eq. (1) can be written as:

$$A\varnothing_{in} = P\varnothing_{in} + S \tag{2}$$

$\varnothing_{in}$ is the solution of the inhomogeneous Eq. (2). The distribution in space, energy, angle of $\varnothing_{in}$ is obviously different from that of $\varnothing$. Of course, $\varnothing_{in}$ approaches $\varnothing$ as the level of sub-critically becomes smaller and smaller, approaching the critical configuration.

For an ADS, once defined in material properties, the geometry of the system, the relevant cross-sections and the source intensity (in neutrons per second), the distribution of the inhomogeneous flux is fully determined by Eq. (2).

Relevant integral parameters characterising the sub-critical core (SC), such as reaction rates, can be easily calculated. This allows evaluating the power deposited at each point of the system, the damage rate, the breeding ratio etc. This is done exactly as in critical systems, characterised by $\varnothing$.

4.3.2.2 The reactivity of the sub-critical core

It is formally possible to describe a sub-critical system with the introduction of a parameter k_{eff} which allows to "restore" the balance Eq. (2):

$$A\varnothing = \frac{1}{k_{eff}} P\varnothing \tag{3}$$

Since $\varnothing$ has the same distribution as the "critical" flux, this equation is obviously an approximation of the real case, as described by Eq. (2).

In order to improve the definition of sub-criticality and to take into account the change in distribution of the flux, a different definition of the sub-criticality has been proposed, by means of a "k-source" k_s. The procedure is to apply the formal balance condition (3) to the inhomogeneous flux Eq. (2):

$$A\varnothing = \frac{1}{k_{eff}} P\varnothing_{in} \tag{4}$$

Integrating and recalling that $A\varnothing_{in} = \frac{1}{k_S} P\varnothing_{in}$ one obtains:

$$k_S = \frac{\langle P\varnothing_{in} \rangle}{\langle P\varnothing_{in} \rangle + \langle S \rangle} \tag{5}$$

4.3.2.3 Neutron source importance

Understanding the behaviour of the source-driven sub-critical core depends largely on the evaluation of the relative importance of the source neutrons to the fission neutrons generated in the SC.

One introduces a parameter φ*, which is the ratio of source neutrons and of the average importance of fission neutrons. It can be shown that this parameter φ* is related to k_{eff} as:

$$\frac{\Gamma}{\bar{\nu}} \varphi^* = \frac{1}{k_{eff}} - 1 \tag{6}$$

where $\bar{\nu}$ is the average number of prompt neutrons per fission, and Γ the average number of source neutrons per fission. Relation (6) is given in [87], where the experimental determination of φ* is discussed.

The φ* parameter plays an important role in assessing the ADS performance parameters. In fact in [14], it is shown that the relation between the proton beam current i_p, the power in the SC and its sub-criticality is given by:

$$i_p = \frac{\bar{\nu}\left(\frac{1}{k_{eff}} - 1\right)}{\varphi^* Z} \frac{W}{\varepsilon_f} \text{ (Ampère)} \tag{7}$$

where W is the power of the SC in watts, ε_f the energy per fission (MeV) and Z is the number of neutrons per incident proton.

It can be seen from Eq. (7) that a value of φ* higher than 1 can reduce proportionally the proton beam current requirement for a given sub-criticality level. Measurements of φ* are made in the CEA facility MASURCA in Cadarache, in the framework of the MUSE programme [88], which will be described shortly in sub-section 5.3.2.10.

4.3.2.4 Kinetic behaviour of sub-critical cores

The equations which give the kinetic behaviour of a system driven by an external source are of the type:

$$\begin{cases} \dfrac{dW}{dt} = \dfrac{\rho - \beta}{\ell_{eff}} W + \Sigma \lambda_i C_i + S \\ \dfrac{dC_i}{dt} = \dfrac{\beta_i}{\ell_{eff}} W - \lambda_i C_i \end{cases} \qquad (8)$$

where C_i are the precursors of delayed fission neutrons with decay constant λ_i. β_i is the fraction of the total number of delayed neutrons emitted per fission ($\Sigma \beta_i = \beta$) due to the precursors [89].

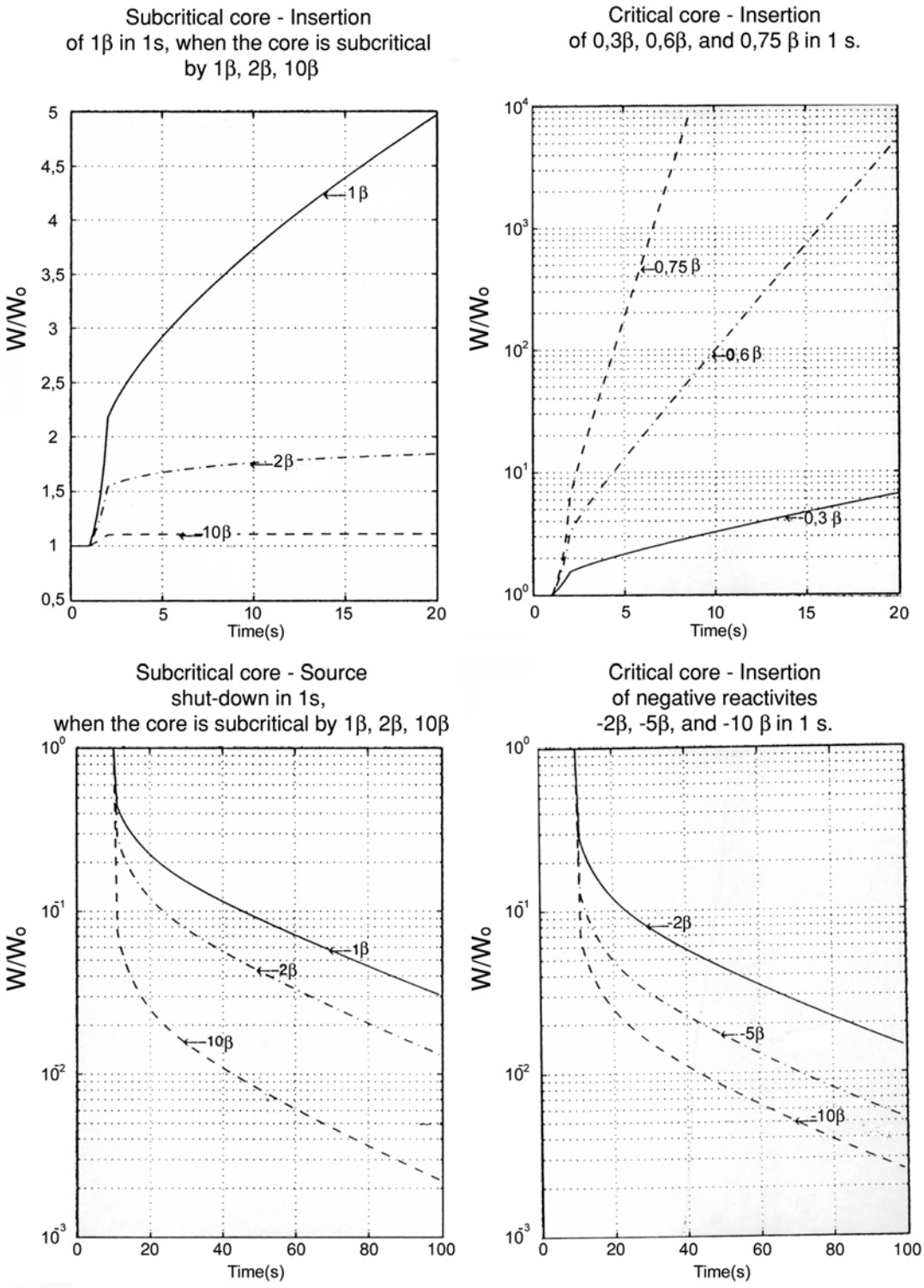
ℓ_{eff} is the neutron generation time and ρ is the reactivity $\left(\rho = \dfrac{1}{k} - 1\right)$.

In steady state (i.e. if $\dfrac{dW}{dt} = 0$ and $\dfrac{dC_i}{dt} = 0$), we have:

$$\rho \frac{W}{\ell_{eff}} = -S \qquad (9)$$

A decrease by a factor h of the reactivity ($\rho' = \rho/h$) or an increase by a factor h of the source ($S' = hS$), induces an instantaneous increase in the power $W' = hW$. For example, if, the system is sub-critical corresponding to -10β, a reactivity insertion of $+5\beta$ causes a doubling of the power (see Figure 4.2). This of course is totally different from the behaviour of a critical system, which becomes prompt critical.

Figure 4.2. **Kinetic behaviour of sub-critical and critical cores**



In more general terms, the kinetic behaviour of a critical system is characterised by delayed neutrons and their time constants (about 10 s.), while the kinetic behaviour of a SC is determined by the time constants related to the external source, in the sense that an instantaneous variation of source has an effect on the time scale of the prompt neutron lifetime (typically of the order of microseconds).

The evolution of the power with time, and the related variation of the temperature, are related to the variation of the reactivity (Doppler reactivity effect, fuel expansion reactivity, reactivity due to the material concentrations in the core, including the coolant etc.). These feed-back reactivity effects are essential for the safety of a critical reactor. In a sub-critical core, the relevance of feedback reactivity effects varies according to the level of sub-criticality. In fact for a deeply sub-critical core, the dynamic behaviour is dominated by the external source and its variation in time. Closer to criticality, the feedback effects become more important and the behaviour of the core is approaching that of the corresponding critical core.

In a very simplified way, if the core is sub-critical by -10β, a feedback reactivity equal to ±1β, induces a ±10% variation of power and a ±50% variation of power if the system is sub-critical by -2β. In a critical reactor +1β reactivity insertion makes the reactor prompt critical and -1β stops the chain reaction. In view of the definition of an "optimal" level of sub-criticality, it is very relevant to verify the transition of the behaviour of the SC from a "source-dominated" to a "feed-back dominated" regime.

4.3.2.5 Reactivity and loss-of-flow accidents

Fast external insertions of reactivity give rise to different consequences in critical or sub-critical cores. Examples have been given in [88,90]. In [90] a 0.55 β/s reactivity insertion in a Phénix fast reactor type core, critical or sub-critical at k_{eff} = 0.95, gives rise (at constant external source level) to the following power and average temperature evolutions:

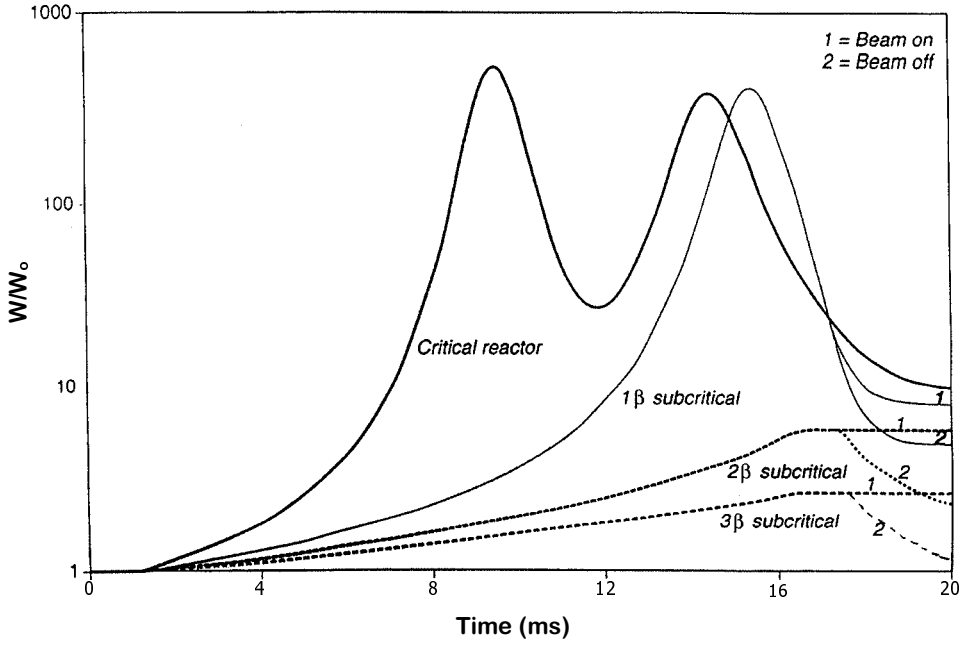
	Critical core	**Sub-critical core (k = 0.95)**
Delay before fuel fusion	2 s.	12 s.
Inserted reactivity	1.1β	6.6β
Power increase W'/W	2.2	1.5

In [88], a reactivity of 170 β/s is injected in a critical core (W_o = 1 GW), or in the same core made sub-critical at -1β, -2β, -3β. The results show that prompt criticality is reached in the critical core after 6 ms with a first power peak of 700 GW at 8.5 ms and a second peak of 500 GW at 13.2 ms. In the sub-critical mode, the peaks are respectively of 530 GW at - 1β, 6 GW at - 2β and 2.2 GW at 3β (t = 16 ms) (See Figure 4.3).

The increase in power is considerably slower in a sub-critical system, and the total energy deployed is much smaller.

In the case of loss-of-coolant-flow accidents, References [88] and [90] give simple examples, which show that, in the case of no shut-down of the source, the behaviour of a -10β sub-critical system is less favourable, since in a critical system the increase of the coolant temperature is slower and lower due to the feed-back effects. Again, the choice of the level of sub-criticality is relevant, if one takes into account the potentially beneficial effects of the intrinsic characteristics of the core.

Figure 4.3. **Impact of external reactivity insertion**



This of course, has to be verified for each type of core and associated fuel and coolant. It is obvious from these considerations that the accelerator beam intensity must be coupled through safety grade scram circuits to the power level of the SC, so that it can immediately be shut down in case of a power excursion.

4.3.2.6 Cores with low Doppler effect

In the case of an ADS dedicated to transmutation, the fuel will be dominated by MA which will have a low Doppler effect, due to the absence of ^{238}U.

The effect on the dynamic behaviour of the core will differ according to the level of sub-criticality (see Figure 4.4). At large sub-criticality, the calculations of the effect of reactivity insertion performed with a "standard" Doppler coefficient k_D, or with a "low" Doppler ($k'_D = 0.1\ k_D$), show no difference in the power or reactivity behaviour. Close to criticality on the contrary, the effect can be significant.

4.3.2.7 Choice of the sub-critical level

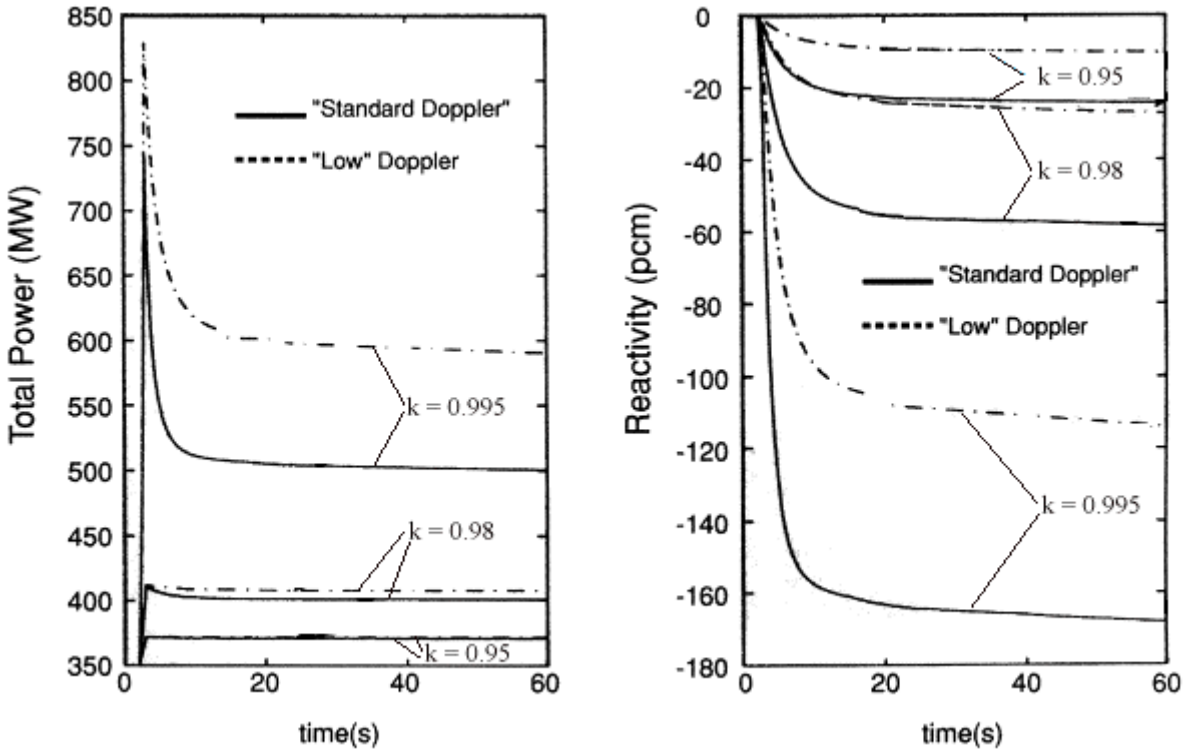
No final criteria have been established up to now in order to define an "optimal" level of sub-criticality. However previous considerations indicate the importance of finding a compromise between the "source-dominated" and the "feed-back dominated" regimes.

More quantitatively, in the case that no control rods are foreseen in the SC, the level of sub-criticality should be such that the core stays sub-critical when going from a "hot" state (i.e. normal operation) to a "cold" state (i.e. reactor shut-down). Since thermal feed-back induces generally (e.g. in standard fast reactors) a positive reactivity effect (Δk_{FB}) on going from "hot" to "cold", one can require that the "cold" core should stay sub-critical even in the case of an accidental reactivity insertion (Δk_{AC}), due for example to coolant voidage.

In that case the required "k_{eff}" should conform to the following relation:

$$k_{eff} + \Delta k_{FB} + \Delta k_{AC} < 1$$

Figure 4.4. **Insertion of 1/3 reactivity in 1 second.**
Behaviour of an ADS "Phénix Type" at three different levels of sub-criticality.



During operation, the maximum reactivity insertion ($\Delta k_{AC}^{M} \langle 1$) can be higher than Δk_{AC}. In that case one has the requirement that:

$$k_{eff} + \Delta k_{AC}^{M} < 1$$

Moreover, during reactor operation, the reactivity varies owing to the irradiation (burn-up) of the fuel and its isotopic evolution. In general this reactivity variation Δk_{BU} is negative, but in some case (e.g. a fuel made essentially of minor actinides, which act as "fertile" materials, since they are transmuted into more "reactive" elements, as it is the case for example of ^{241}Am), Δk_{BU} can be positive. In that case, if the core has no control rods and one does not want to modify the external source, e.g. by changing the current intensity, one should have:

$$k_{eff} + \Delta k_{AC}^{M} + \Delta k_{BU} < 1$$

Looking for a compromise between the different criteria indicated above, one has also to consider that a very large sub-criticality may not be necessarily the optimal solution. In fact, besides obvious considerations on the "cost" of a strong external source, a largely sub-critical core has a peaked power distribution, dominated by the source distribution and therefore very far from flat, as required to optimise the fuel irradiation and, consequently, the fuel transmutation (see also the next chapter on safety issues).

4.3.2.8 Reactivity control and monitoring

The control of reactivity and of the power level in a critical reactor is essentially through control rods. In principle, an ADS can be controlled solely through the external source. As an example, the variation of reactivity with the fuel burn-up can be compensated by an appropriate change of the beam current intensity. A similar system can also be conceived to control the reactivity change between "hot" and "cold" states. However, major variations of the current would be necessary. For example in a SC without control rods, with $k_{eff} = 0.99$ in the "cold" state, $k_{eff} = 0.98$ in the "hot" state at the beginning of an irradiation cycle and $k_{eff} = 0.95$ at the end of the cycle, the source intensity should change by a factor of approximately 5 to account for both the attainment of nominal power and the variation in reactivity during the operational cycle. In this context, it is clear that the use of control rods should be carefully considered to ensure at least some of the functions of reactivity control.

Moreover, if in a SC, in particular in a "source dominated" mode, the shut down of the source has an instantaneous effect to reduce power, the inverse effect, e.g. an "overshoot" due to a sudden increase of the external source, has the consequence of an instantaneous increase in the power. Although more limited than the potential power increase in a critical reactor, such an accidental situation should be examined.

Also, when the reactor is shut down, the consequences of inserting the full "reserve" of beam current should be analysed. In fact, if the insertion of the full "reserve" of beam current cannot be excluded, this accidental event could lead to a power variation given by [91]:

$$\frac{W'}{W} = 1 + \frac{\Delta\rho}{\rho + \beta} = 1 + \frac{\delta_{ip}/i_p}{1 + \beta/\rho}$$

If W_{Max} is the maximum allowable power in a short time interval, one can deduce the maximum allowable sub-criticality level such that $W' < W_{Max}$.

Finally, we should mention that in principle, long term variations of the reactivity can be achieved by an appropriate variation of the φ^* parameter. This can be obtained, for example, by changing the geometrical arrangement of the buffer (or of the buffer material) surrounding the spallation source.

As for monitoring the level of sub-criticality, different methods can be envisaged and experimentally validated. Some examples are as follows:

- Using the source of in a pulse mode. Recording the time evolution of the counting rates of in-core neutron detectors can allow measuring the reactivity. In fact, the point kinetics predicts the prompt decay of the neutron population after a pulse to be of the type $\exp(-\alpha t)$ with $\alpha = (\rho - \beta)/\ell$. For known values of β and ℓ one can deduce ρ from the decay of the neutron population observed experimentally.
- If control rods are foreseen, the modified source multiplication method (MSM, see [92]) can be used provided the calibration of the control rods' reactivity is performed at near-critical level.

4.3.2.9 Beam trips

As far as the coupling of the accelerator to the sub-critical core is concerned, one significant point which has been raised [93] is the effect of frequent beam trips on the SC. Since we have seen that the time scale for power variation (due to source variation) is very short, while the heat transfer time from fuel to coolant is of the order of 0.1 to 1 sec, the heat is stored in the fuel for ~1 s. making high thermal conductivity fuels a possible requirement. In a similar way, thermal stresses in the core structures can

be expected owing to the difference in time constants between power increase and temperatures variations in the structures, and in the case of frequent beam trips, fatigue failures of the structures could occur and arouse safety concerns.

4.3.2.10 Experimental validation

The physics characteristics and the predicted behaviour of a SC, as outlined in previous paragraphs, need an experimental validation, in order to calibrate the calculation tools and to gain confidence in the prediction of the basic safety features of an eventual future ADS, which will be fuelled with very innovative fuels.

The main fields which need experimental validation are:

- The effects of the relative contributions of the source neutrons and of the neutrons generated by fission, φ* measurements should achieve that objective in stationary conditions.
- Experiments performed at different sub-critical levels, with or without feed back effects, can be essential to understand the transition between a "source-dominated" and a "feed-back dominated" regime.
- Spatial and energy distributions of neutrons and their variations close to the external source.
- Assessment and monitoring of the sub-criticality level.
- The relationship between the external source and the power in the core.

A first experiment related to verifying the physical principles of an ADS was performed by C. Rubbia at CERN (FEAT experiment, [94]). A proton beam struck directly a natural uranium block, and the "energy amplification" was experimentally verified.

If the SC is not very sub-critical, i.e. with k_{eff}>0.95, it is possible to study its neutronics using other well-known external neutron sources substituted for a true spallation source, for instance a ^{252}Cf spontaneous fission source or a 14 MeV (d,t) neutron source. Since 1995, such studies have been under way at the MASURCA facility of CEA in CADARACHE, and a series of experiments called "MUSE" (MUltiplication avec Source Externe) has been performed (See Table 4.6) in a collaboration between physicists from Cadarache (CEA) and ISN-Grenoble (IN2P3), now extended to various European partners in the frame of a specific project in the Fifth European Framework Programme. The purpose of these experiments is to separate the effects of the source and of multiplication in the SC [87].

In the present MUSE-4 experiments with a pulsed 14 MeV neutron source called GENEPI, the target is surrounded by a lead buffer, to simulate the neutron diffusion inside an actual lead (or lead-bismuth) target. Numerical simulations have shown the validity of the basic hypothesis of the experiments, namely that using a spallation neutron source or the neutrons issued from the (d,d) or (d,t) reactions, the neutron spectrum in the core close to the buffer region is very much the same, whatever the energy distribution of the neutron source (See Figure 4.5). Additional information on the MUSE experimental programme is given in Chapter 7.

A joint ENEA-CEA working group has recently launched the idea to carry out a pilot experiment to demonstrate the feasibility of stable operation and to analyse the dynamic behaviour as well as to investigate certain safety issues of an ADS. This experiment, actually called TRADE and which would form a first example of ADS component-coupling "at real size" (<1 MWth), would be performed in the TRIGA reactor at the ENEA Casaccia Centre. The reactor would be operated as a sub-critical assembly and externally driven by a proton cyclotron. Additional information on the TRADE experimental programme is given in Chapter 7.

Figure 4.5. **The MASURCA installation for the MUSE programme**

A sub-critical MUSE-4 configuration

The MUSE experiments and other experiments which are planned or have recently been started should give most of the demonstrations needed in order to proceed to a sound design of an experimental ADS, as proposed, e.g. in the European Roadmap towards and ADS demonstration [9].

Table 4.6. **The MUSE experiments at MASURCA**

	Type of source	**Range of sub-criticality**	**Diffusing buffer around the source**
MUSE-1 (1995)	^{252}Cf spontaneous fission neutron source	-1.5% $\frac{\Delta k}{k}$	None
MUSE-2 (1996)	^{252}Cf spontaneous fission neutron source	-3.0÷3.5% $\Delta k/k$	Sodium Steel
MUSE-3 (1998)	Pulsed neutron source from (d,t)	-0.5÷-6.% $\frac{\Delta k}{k}$	Sodium Steel
MUSE-4 (2000-2001)	Pulsed neutron source from (d,d) and (d,t)	-1÷0.4% $\frac{\Delta k}{k}$	Lead

4.3.2.11 Technological problems

All conceptual ADS designs available today are of a preliminary nature and some relevant technological problems are still to be solved in a satisfactory way.

This is the case, for example, of the shielding configurations in the upper part of the systems. The shielding in fact should allow for the potential deep penetration of high-energy neutrons ($E_n \geq 100$ MeV) released by the spallation of protons (Typically $E_p = 0.6 - 1.5$ GeV).

High-energy neutron penetration experimental studies performed in Japan, confirm the very large thickness of material (such as concrete or stainless steel) needed in order to reduce to an acceptable level the doses around the structures.

The beam entrance configuration is also a matter of concern. In fact, a simple vertical entrance of the beam can imply a very complicated system for the fuel loading-unloading system and can also be sub-optimal with respect to the need to guarantee the beam tube free from the intrusion of back-scattered neutrons.

These are just a few examples of technological problems that can have impact on the coupling of the different components of an ADS, and which could need substantial efforts in order to develop a robust ADS design.

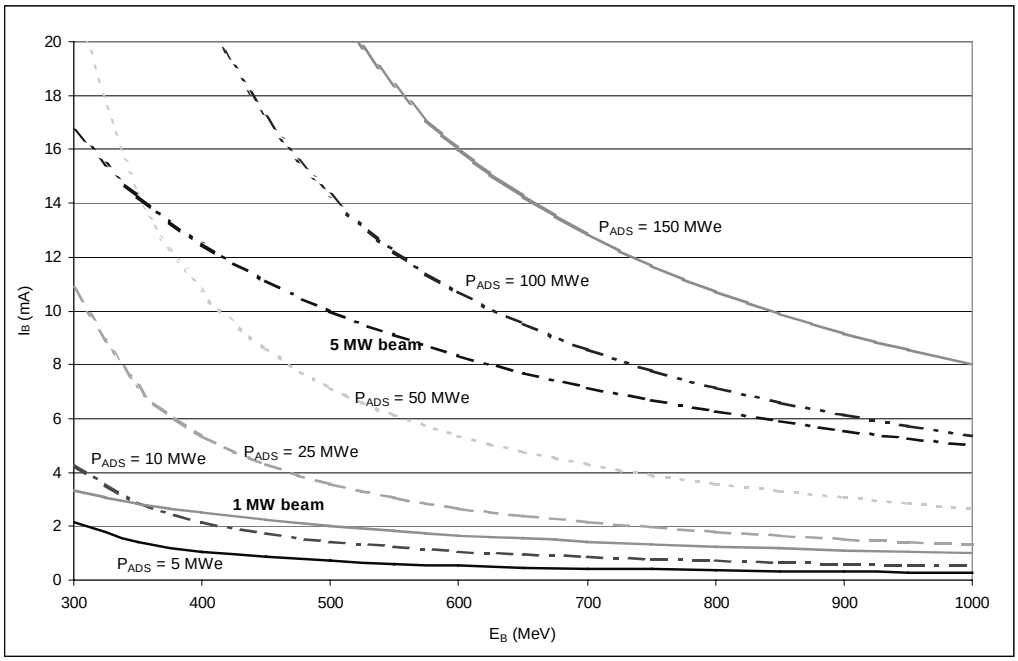
4.3.2.12 Relationship between sub-critical reactor and accelerator

A crucial element in the assessment of an accelerator-driven system is the required beam power, and thus accelerator characteristics, for a given sub-critical reactor configuration, i.e. reactor power, level of sub-criticality (k_{eff}), etc.

Figure 4.6 shows the relation between the beam parameters (beam current I_B and beam energy E_B) and the electric power level of the ADS for a fixed k_{eff} of 0.97. As shown, the total power level of an

ADS is limited to about 100 MWe as currently beam power is limited to at most 5 MW. The chosen ADS characteristics (136 MWe) in the fuel cycle schemes are in that respect already extrapolations of technology to the future.

Figure 4.6. **Beam power for ADS**



4.3.3 Spallation target technology

One of the most stressed components of an ADS is the spallation target which is designed to generate the maximum amount of neutrons while ensuring the removal of the heat released in the spallation process. As the proton beam power being deposited in such a target attains several MW, even up to about 20 MW, very high power densities of several hundreds of kW per litre, occurring in the structure and in the spallation material, need to be safely removed. In addition, the mixed proton-neutron irradiation field in the target imposes very specific conditions on the design and operability of the target and influences strongly all the thermo-mechanical options for such targets. Heavy metals such as tungsten, tantalum, uranium, lead-bismuth, lead, mercury are considered as possible spallation materials for targets. Gas, heavy water or liquid metals are under consideration as coolants for these targets.

Two main options for a spallation target are available, i.e. solid or liquid metal. A wide range of experience with operating facilities exists in the design and use of solid spallation targets, essentially in mostly pulsed spallation neutron sources of lower power. Owing to the extremely high power density, the use of a solid metal target in ADS applications has to be excluded. The main advantages of a liquid metal target in ADS are, first, the perfect cooling of the target and the heat removal capacity which is inherent in the design having a flowing liquid metal coolant, and as a consequence, secondly, the greater achievable and tolerable power densities and the significant reduction in irradiation damage to the target itself and structural materials. However, other problems can arise with liquid metals such as lead or lead-bismuth as spallation material and coolant. These are corrosion and erosion of structural materials which are in direct contact with the liquid metal at high temperatures

and high flow velocities, the behaviour of (volatile) spallation products and the need for a beam window between the evacuated proton beam guide and the spallation region.

Solid targets

At present, only solid targets are used in operating spallation neutron sources. They are usually assemblies of rods or disks fabricated from tungsten, uranium or tantalum and cooled with heavy water. In certain test cases, steel rods containing lead or lead-bismuth have been introduced as spallation material. The following problems and difficulties should be noted in designing and developing these targets:

- Radiation damage to target and structural materials, i.e. swelling, and the degradation of heat-conducting properties associated with helium accumulation.
- Complexity of target cooling, where high energy deposition is observed (several hundreds kW per litre).
- Radiolysis and activation of the cooling water, where experience in SINQ (PSI, Switzerland) showed that contamination of water with ^{7}Be and other spallation products as well as deposition of ^{7}Be on inner surfaces of the circuit can be unpleasant operational problems.
- Cooling of the target after shut-down of the accelerator (proton beam), where residual energy deposition due to decay of spallation products can reach 1-2 kW/L and relatively powerful auxiliary cooling systems are needed.
- Whether sodium (Na) should be used as liquid metal coolant, given that its very favourable thermal and heat-removal characteristics are counterbalanced by its chemical reactivity with air and water.

Solid targets were proposed in some early ADS-projects but this route was rather quickly abandoned in favour of liquid metal targets.

Liquid metal targets

Lead (Pb) and lead-bismuth (Pb-Bi) eutectic have been the two primary candidate liquid metal target materials for the production of spallation neutrons in ADS. Lead would be advantageous over Pb-Bi as it would significantly reduce by a factor of 10^{3}-10^{4} the build-up of the α-emitting ^{210}Po coming from (p,xn)-reactions on Bi in the target. Nevertheless, an essential disadvantage of lead is the higher melting point (327°C compared to 125°C for Pb-Bi eutectic) which causes many engineering and technology complications. Other liquid metals, such as mercury (Hg), have also been proposed for use in advanced spallation neutron sources as well as for ADS. The main advantage of Hg would be the absence of ^{210}Po-activity and the possibility of not having to heat up the system before operation which would simplify many engineering problems. But the high volatility of Hg imposes extremely strict requirements on the primary circuit and the integrity of the cover gas system since radioactive mercury must be prevented from leaking into a working environment. Mercury targets with a boiling point of 356°C would also be difficult to use in ADS as the working temperatures are higher than in spallation neutron sources for other purposes. Finally, the high neutron absorption cross-section of Hg is against its use in ADS.

The use of liquid metal targets does not simplify the design and development of spallation targets as multiple problems do appear. First of all, a liquid metal target needs a container, i.e. a liquid metal circuit integrated into the core of an ADS as well as an interface between the liquid metal target and the proton beam guide. This latter may take the form of a solid beam window or a windowless design

(see later). The choice of structural materials both compatible with the liquid metal and able to resist the thermo-mechanical loads on the target circuit is a prime focus for R&D. The selection of a container material for the liquid target will therefore greatly affect the lifetime and safety of the target system. Because a beam window is an integral part of the target containment structure, it will be exposed both to a significant flux of high-energy protons and neutrons at a temperature of up to 650°C, and to a very corrosive environment. In a full-scale ATW system, for instance, a total proton fluence between 10^{26} and 10^{28} p/m^2.year can be expected on the target window. It is therefore likely that the beam window will have to be replaced at least yearly because of the expected material damage. In conclusion, the material should have good compatibility with the liquid metal target, good machining and welding capabilities, sufficient mechanical strength at the high operating temperatures, a low-neutron-absorption cross-section, and good performance under an intense proton bombardment.

In most of the target designs, temperature-gradient mass transfers will be the most damaging of all materials degradation phenomena in liquid metal because of the large temperature gradients expected in the system. As a result, both the thinning of the window and potential fouling of the heat exchangers are troublesome. The relative resistance of 24 metals and alloys to mass transfer in liquid lead with a temperature difference of 300°C (500-800°C) was measured by Cathcart and Manly [95]. The results indicate that only niobium and molybdenum showed no mass transfer, which in the other materials was slight to heavy. This liquid metal corrosion behaviour can be significantly reduced by controlling the partial oxygen pressure in the liquid Pb or Pb-Bi [96]. Additional measures such as coatings or surface restructuring and alloying are very promising technologies to minimise liquid metal corrosion of critical components such as the beam window [97].

Besides these mass transfer and oxidation phenomena, spallation products, including hydrogen and oxygen, will build up through the interaction of high-energy protons with the target material. These products will behave as alloying or impurity elements in the liquid lead or lead-bismuth target material where they are formed and thus may have detrimental effects on the containment material. While the production of oxygen seems to be negligible or could even be eliminated with an oxygen control system as mentioned above, further studies on the production rate of oxygen and on its interaction with other spallation target products are required.

Specific features of the coolant technology for liquid metal targets are caused by two factors:

- Accumulation of spallation products which could possibly influence the physical-chemical processes in the coolant and destroy protective oxide films on structural surfaces.
- High activity in the coolant and the cover gas causes high activity in the gas mixture which is removed from the circuit. This makes the coolant loop non-repairable and essentially aggravates analysis of gas compositions.

With respect to these spallation products, benchmark calculations showed that long-lived radioactivity accumulates mainly owing to primary nuclear reactions. Secondary reactions are responsible for producing a small number of long-lived isotopes, ^{207}Bi, ^{210}Po and some others generated by radiative capture of low energy neutrons. Neutrons in the energy range 20-800 MeV and protons with energies above 100 MeV make the main contribution to the total activity generation although these parts of the spectra inside the target have a rather small contribution to the total flux. Correctly estimating the activity of short-lived nuclides is the main problem in analysing target behaviour in the case of short accelerator shut-downs. They make the dominant contribution to both activity and heat release in the first moments after the shutdown, creating the intermediate links and additional routes for decay to the long-lived nuclides. The strong dependence of calculated concentrations of short-lived nuclides on the choice of the cross-section data library for determining the reaction rates is to be noted. Recent experiments using tungsten and lead targets indicate that the experimental production rates of

spallation products agree with the computer code calculations within a factor of 2 for about 50% of the observed nuclides but differ significantly more, even by several orders of magnitude, in for the others [98].

Lead targets

In the LANL ATW pre-conceptual design for a liquid lead target, it is estimated that the maximum temperature of the system will be around 900°C at the point where the proton beam impinges on the window. At this temperature, iron-based alloys are inadequate because of their high-creep rates and poor oxidation resistance. Iron-based alloys are usually limited to a maximum of 650-700°C in service. Similarly, nickel-based and cobalt-based super-alloys are only marginally acceptable because they are limited to a maximum of 900-1 000°C. Another disadvantage of these super-alloys is that nickel is incompatible with liquid lead, and cobalt has a high absorption cross-section. Refractory metals such as niobium, tantalum, molybdenum and tungsten are usually used at service temperatures much above 900°C.

The three refractory metals – molybdenum, tantalum, and tungsten – all have their own problems as containment materials for the ATW system. Molybdenum becomes too brittle after low-fluence proton irradiation. Tantalum has an unacceptably high thermal neutron absorption cross-section and poor oxidation resistance. Tungsten also has a high thermal neutron absorption cross-section and low ductility.

Nb-Zr has been selected as a containment material for the ATW liquid lead target. Although this material has desirable properties for contact with the liquid lead target, one major drawback is its low oxidation resistance at high temperatures. Therefore, the ATW lead target must be designed to be used in a vacuum or at very low partial pressures of oxygen. This low oxidation resistance therefore also defines the minimum wall thickness (about 0.2-0.4 cm) of the window if a 10 or 20% loss in wall thickness is permitted during one year of operation [99].

Another possible way to alleviate the materials problem for the liquid lead target would be to use coolant to lower the window temperature at the cost of a more complex window design. There are two design options to lower the maximum temperature in the beam window material, which is at the stagnation point of the window hemisphere with present target design studies. First is a re-shaping of the footprint of the proton beam (e.g. the two-dimensional power distribution in the proton beam) in such a way that the maximum power is moved out of the centre of the beam. Second is by changing the flow field in the spallation volume and around the beam window, e.g. by introducing an additional jet flow across the window as is going to be realised with the MEGAPIE spallation target at SINQ, PSI [100].

As a consequence of reducing the maximum operating temperature of the beam window, more candidate materials would be available. Reducing the liquid metal temperature below 300°C could favour serious liquid metal embrittlement, which needs further considerations.

Lead-bismuth targets

Owing to the lower working temperatures achievable, lead-bismuth eutectic has become the preference in most of the ADS designs with separate targets. A specific feature of a molten lead-bismuth target is the high activity of spallation and fission products in the coolant (approx. 500 Ci/kg). Because of thermal diffusion and evaporation, gaseous (Kr, Xe) and volatile (Hg, Cs, I, Br, Rb) nuclides escape into the cover gas system. The specific activity can reach a few Ci per litre (2.5 Ci/l for the LANSCE conditions), which is 5 orders of magnitude higher than in a reactor with Pb-Bi

coolant under normal conditions. This necessitates special shielding for the cover gas system, complicates repair operations and makes a gas system break hazardous. In addition, an important factor influencing radiation safety is the accumulation of α-active polonium radionuclidesisotopes. Unlike reactors where only ^{210}Po is formed as a result of neutron capture by ^{209}Bi, in spallation targets (p,xn)- and (α,xn)-reactions result in the formation of Po nuclides, among which ^{209}Po ($T_{1/2}$ =102 y) and ^{208}Po ($T_{1/2}$ =2.9 y) are the most important (the specific activity of ^{208,209,210}Po reaches about 1 Ci/kg in the target circuit).

For the reliable operation of liquid metal target coolant technology, it is important to ensure purification of the coolant and corrosion resistance in structural materials in the cooling circuit. This technology was developed for Pb-Bi as a coolant in nuclear reactors. It comprises, in particular, the formation and maintenance of oxide scales perhaps with a very thin, stable aluminium or silicon plating on the surfaces of the structural material to protect them against corrosion, as well as reducing lead oxides by means gaseous mixtures including hydrogen. Such an oxygen control system together with in-situ ceramic oxygen meters has been developed for loop systems in the Russian Federation and in the Karlsruhe Lead Laboratory KALLA [101].

Structural materials for Pb-Bi targets

The high solubility of nickel excludes the use of austenitic stainless steel or nickel-based alloys as containment material for Pb-Bi eutectic. The materials 2-1/4Cr-1Mo steel, modified 9Cr-1Mo steel, 12Cr-1Mo steel or HT-9 are suggested as candidate container materials for the Pb-Bi eutectic targets in ADS systems. The general tendency of these materials is that the higher the chromium content in the alloy, the higher the corrosion resistance to the eutectic and the lower the strengths, and vice versa. However, an effective oxygen control system still has to be applied to the spallation target.

Inhibitors are very effective in reducing the corrosion of steel by forming, for example, carbide and nitride films on the surface. Zirconium is perhaps the most effective inhibitor of low-alloy steels and it has also a lower thermal neutron absorption cross-section than titanium. If inhibitors are used during the ADS operation, a variety of spallation products, built up by the interaction of high-energy protons with the target material, will react with the inhibitors in the Pb-Bi eutectic. These reactions may reduce the beneficial effect of the inhibitors and more research on the interaction between the spallation products and the inhibitors is required.

Radiation dose to structural materials and beam window

The most complicated and unknown issue is the radiation stability of the beam window (and other structural materials) related to degradation of its mechanical properties under conditions of mixed proton-neutron exposure. Experimental data are, in principle, available only to damage doses of about 10 dpa. Damage dose in the end of the TC-1/LANSCE lifetime (irradiation of 7.5 mA.month) is about 40 dpa, helium and hydrogen generation is 2 500-4 000 and 18 000-22 000 appm respectively. Maximum stresses in the window are 340 MPa.

By use of a windowless target but having a separate window in the proton guide tube, the radiation dose to this window outside of the target may be approximately 1.5-2 times lower than in a metal-cooled window since neutron irradiation will be negligible in comparison with proton irradiation.

The evaporation of liquid metal and of the spallation products into the proton guide tube, the additional shielding efforts, and the vacuum system have to be considered. In case of a rupture of the

guide tube the release of radioactive products has to be taken into account. Another issue with a windowless target is the hydraulic stability of the freely falling liquid metal film, and the formation and possible rupture of waves on it. In principle, in a symmetrical solution, the problem of high temperatures in the stagnation point (lowest position in the cone-shaped film) and evaporation of coolant is not solved for high, ADS relevant proton beam powers.An asymmetrical solution demands more space, but could give solution to this issue.

Effect of beam trips

Beam trips have two major consequences for the target: first, thermal shock and considerable pressure waves on target circuit components and, second, the necessity to use on the secondary side of the heat exchanger cooling water at a temperature higher than the melting point of the coolant, in order to prevent coolant in the circuit from solidifying. Applying double-walled heat exchangers with a coupling fluid, which can change the heat transfer surface between primary side and secondary side of the heat exchanger, would be a possible solution and allow direct control of the heat transfer characteristics.

To prevent or mitigate accident consequences it is important to remove the beam quickly from the target in the event of an emergency situation in the heat removal system (pump failure, window rupture, …). For this purpose, obviously, emergency signals should be envisaged using different physical parameters (temperature, energy supply characteristics, …) and duplicate signal channels.

4.3.4 Accelerator technology

To accelerate a high intensity proton beam to an energy of the order of one GeV, two completely different accelerator schemes are possible, a linac or a cyclotron. The choice depends on many factors, but it is important to clarify from the beginning that, to fulfil the beam requirements for ADS applications (specifically a very low frequency of beam interrupts), both machine designs have to be modified and developed, to extend into a new dimension of complexity, cost, and size. The operating mode for ADS will most likely be CW (continuous); although pulsed mode operation could be used for testing, set-up, etc.

A proton linear accelerator (linac) has performance limitations which may be more economic than technological, and provides straightforward solutions to some of the cyclotron’s problem areas.

Strong transverse focusing elements (quadrupoles) placed at frequent intervals along a linac as well as longitudinal focusing, due to the phase stability, set a much higher limit to the charge per bunch that can be accelerated without significant beam loss. Linacs also operate with RF-cavity frequencies typically 10-20 times higher than used in cyclotrons. Taken together, these factors mean that in principle a linac could accelerate a current from one to two orders of magnitude higher than a cyclotron, with no problems at extraction. Given an adequate linac length, the final energy is not limited on dynamical grounds. The electrical efficiency is high at high beam currents, even in a normal-conducting linac. If a superconducting linac is used for the high-energy section, the efficiency is even greater.

The major drawback for a linac is the length, which depends only on the final beam energy and the accelerating gradient, and is independent of the beam current. The length is an important factor in the cost of the facility, since typically most of it will need to be shielded against radiation produced by small beam losses.

Cyclotrons are based on the so called “cyclotron resonance” which states that, in a constant magnetic field, perpendicular to the beam orbit, the particle revolution frequency is fixed and independent of the particle energy; but, in the case of sector focused cyclotrons, at relativistic energies, energy levels beyond 1 GeV become more difficult to obtain. This simple rule, implemented with some clever ideas to improve the transverse focusing, has been the basis of the hundreds of low energy, low beam power, medical cyclotrons, scattered around the world. The main characteristic of such a machine is that just a few accelerating structures, fed by a CW RF generator via a resonant cavity, are required to transfer, step by step, the full energy to the beam. The beam circles isochronously with respect to the RF field in all the hundreds of passages needed to build up the full energy. Cyclotrons generally produce CW beams, since they operate with fixed magnetic fields and a fixed RF frequency.

As the relativistic effect increases the particle mass, transverse focusing has to be effected by spiral shims on the magnet pole with an angle increasing with energy. High intensity cyclotrons use separated magnet sectors and acceleration over two to three cyclotron stages. A proton energy of 1 GeV seems to represent a reasonable limit for a multistage cyclotron design.

A major problem for a high energy, high current cyclotron complex is the beam extraction system. To limit losses and minimise activation, the deflecting system that guides the beam out of the magnetic field, deflecting it with magnetic channels and high voltage electrodes, is permitted to touch only a negligible fraction of the beam. The current limit in the cyclotron is then given by a design requirement to produce a clean beam at the outer radius of the machine, with a radial separation sufficient for a single turn extraction. The latter depends on the voltage capability and number of the RF cavities, because the turn separation is determined by the energy gain per turn.

4.3.4.1 Present status of linear accelerator technology

Most of the existing large proton linacs have been designed as injectors of large synchrotrons [102], and are short pulse machines with relatively low average beam power. The highest power machine is the LANSCE linac at Los Alamos [103], an 800 MeV accelerator that is capable of delivering an average beam power exceeding 1 MW, with a duty factor of about 10%. All the existing machines are built with room-temperature water-cooled accelerating structures, and are pulsed.

In a linac the maximum current that can be accelerated is dictated by the charge per bunch and this value has been for all the past applications much higher than the required average current which has been of order 1 mA or less. For a given energy, the linac length depends on the average accelerating field, and the power required to excite a room temperature RF structure is proportional to the square of this field. As a consequence, to minimise the mains power and the investment cost, in almost all existing facilities pulsed operation has been chosen. CW operation, however, makes sense economically at very high average currents (of the order of 100 mA). In fact, in this case, the power transferred to the beam is so high (100 MW for one GeV energy) that a good efficiency is obtained even if a power of similar magnitude (about 50 MW) is dissipated on the walls of the RF cavities. The early designs of the large and expensive accelerators intended for tritium production in the USA and France, as developed in the early 1990s, were based on this approach.

Proton linacs are now considered fairly competitive in the 10-20 MW beam-power range mainly because of the very impressive results obtained in the last ten years in the fields of superconducting (SC) cavities and related cryogenics. Hundreds of CW superconducting RF cavities are presently in operation at CERN (LEP2) and Jefferson Lab (CEBAF), with an accelerating field exceeding 5 MV/m. Owing to the very low RF losses in the superconducting regime (5 orders of magnitude lower than for room temperature copper), a very small power is required to create a much higher accelerating field and almost all the RF power is then transferred to the beam. This permits a much shorter and more efficient linac design. Including the cryogenic static losses and the cryoplant conversion efficiency, the mains power

required to establish the accelerating field is both at LEP2 (working at 4.5 k) and CEBAF working at 2 k), of the order of a few kW per MeV. This value depends only on the cavity gradient and operating temperature, and not on the linac beam current. The outstanding results recently obtained at DESY in the framework of the International TESLA Project (a superconducting electron-positron linear collider) have demonstrated that much higher accelerating fields can be obtained (up to 25 MV/m) and reliably used. Moreover the improvements to the cryo-module design (and partially in the niobium quality), have greatly reduced the required mains power; as an example the TESLA cryo-module, with cavities operated in CW at 12 MV/m, requires a total mains power for cryogenics, including RF and HOM, of 600 W per MeV.

The linac scheme that is considered in the following is then based on a solution, accepted worldwide, which moves switches to the use of superconducting-cavity technology at an energy of the order of 100 MeV. That means that the low energy part of the linac would be made up of room-temperature copper cavities, while the high- energy part would be a superconducting-cavity accelerator operating at 2 k. The transition energy has been set at 190 MeV rather than 100 MeV in the SNS linac case, because the lowest beta section of the SC linac is the more critical one for the Lorentz force detuning effect, which is an important issue in the case of pulsed operation.

The short schedule of this funded project in the USA and the lack of superconducting cavity prototyping in this beta range also pushed the transition energy in the SNS linac to a higher value.

The chosen reference linac is composed of a sequence of 4 different accelerators. The beam extracted from the last, the superconducting high-energy linac, is directed on to the spallation target. All the discussions on reliability and efficiency have to take into account the particular characteristics of these different accelerator types, individually referred to the present status of the art. The energies chosen for the transition from one accelerator to another should be considered only as a reference case, the precise value being determined by the overall design optimisation. The 4 accelerators are:

- DC injector up to: 100 keV.
- Radio frequency quadrupole (RFQ) up to: 5 MeV.
- Normal conducting linac (DTL, or similar) up to: 100 MeV.
- Super-conducting linac (elliptical cavities) up to: design energy.

The present state of the art of the first two linac components sets a current limit for the accelerator of the order of 100 mA. Higher currents could be obtained by combining outputs from two DTLs if desired, but this process (funneling) has not been demonstrated. It is worthwhile to note that the current limit applies to the peak current if the linac is pulsed, or to the average current if the CW operation is chosen.

Prototypes of 100 mA proton sources are now in operation in various laboratories [104]. Taking as a reference the results from IPHI at Saclay, it appears that this component, for an injector voltage of the order of 80 kV, is well understood and very reliable. In operating the source at a current level 20% below the design value, no beam trips are observed. Extrapolating the preliminary existing data, at the maximum design current, one beam trip (spark on the electrode) per week is expected. With the experience gained and taking a proper margin on the design values this component could be considered as highly reliable.

A working 100 mA CW RFQ at 350 MHz is now in operation at LANL, as part of the LEDA project [105,106]. Other CW RFQs are in the design/construction phase at Saclay, Jaeri, Legnaro (INFN) and LNL. The general impression is that, thank to the LEDA (Low Energy Demonstration Accelerator)

experience and always with a proper margin, this part of an accelerator can now be designed as a very reliable component, limiting the reliability problems to the high power RF components, like klystrons and RF coupler windows.

Prototypes of CW DTL (Drift-tube Linac) and CCDTL (Coupled-cavity Drift-tube Linac) have been recently developed in the framework of the tritium and ADS R&D programs (e.g. the Italian TRASCO and the French IPHI programs). The DTL linac scheme is very old and well established. In fact it has been used since the early fifty in all the high peak current pulsed injectors for proton synchrotrons. What is required by the new projects is the CW operation that implies the dissipation of a very high power from the accelerating electrodes. This engineering problem is quite similar to that solved for the development of CW RFQs. Very reliable 3D computer programs now exist for a joint optimisation of the electromagnetic and thermo-mechanical behaviour. Once the RF structure is developed and tested, the limit of this accelerator is expected to be related, as usual, to the high power, standard RF components.

The super-conducting linac design is derived from the experience gained at CERN, TJLab, and DESY, where high performance super-conducting electron linacs are in reliable operation. The switching of the SNS design to this technology, once funded and in spite of the tight time schedule, is strong proof that the expectations of improved reliability, and reduced capital and operational cost have to be considered as fully realistic. In practice this design uses elliptical-cavity technology developed for electron linacs ($\beta = 1$), but compresses the shapes longitudinally to adapt them to the lower beta appropriate for different sections of a proton machine. Working prototypes of these cavities have been built at several beta values, and they behave as expected from extrapolating the electron cavity performances [107,108]. Cavity efficiency, in terms of cryogenic mains power per MeV and real estate gradient, increases as beta approaches 1. Beta values below 0.5 are not considered because the shape compression of elliptical cavities is too extreme for good efficiency. Other kinds of SC cavities (spoke resonators) may be applicable below beta 0.5 in the future, but until now only single-cell versions have been demonstrated. Three beta families are required for energies above 100 MeV. Roughly speaking, the first section (β ~0.5) is used to accelerate the beam up to ~200 MeV, the optimisation of the second and the third beta sections (and corresponding energy ranges) depends on the required linac output beam energy. Energies up to 2 GeV are compatible with a three-section scheme.

Beam power, component reliability, trip rate and duration

The need to achieve high beam reliability, or a low beam interrupt rate, is a key requirement for ADS applications. High beam power is preferred by an ADS linac in term of efficiency, cost per MW and even reliability. Minimum and maximum current values respectively of 10 mA and 100 mA could be considered as reference numbers. For currents above 50 mA, a duplication of the low energy section (up to ~100MeV) should be considered to achieve high reliability, while a “spares on line” scheme is preferred for the super-conducting linac. A high duty cycle (up to 90%) beam structure is compatible with the present status of the art of the RF controls [109], opening the option to share the beam power among a number of sub-critical reactor experiments. In this case each experiment would see a pulsed beam in the millisecond scale [110].

In principle, modern controls, based on fast digital electronics, make it possible to reduce the duration of most beam trips generated by sparking high voltage components to less than 100 milliseconds. Since most proposed transmuter designs do not undergo significant temperature changes in less than 300 milliseconds, beam trips of this duration or less will have essentially no impact on their integrity. However, beam trips longer than 300 milliseconds, which would be due to real equipment failures, would produce thermal cycling of the transmuters that could cause life-limiting stress damage. The frequency of these longer beam trips depends on the equipment safety

margins used in the accelerator design, and also on the degree of equipment redundancy. These factors will be major cost drivers. The estimated annual number of trips that can be achieved with "high-reliability" accelerator design ranges from a few tens to a few hundreds per year, a large reduction from the 10 000 per year that is the performance level of existing accelerators. Further studies are required to refine the permitted and attainable trip rate and its impact on the project cost.

On the assumption that all the standard accelerator components are well designed and built according to "space-qualified" specifications; that is with the required margin and redundancy, predictable long beam trips (minutes, hours or days) should be just those randomly generated by the lifetime of the high power components. For example, the present lifetime of high-power klystrons is in the order of 25 000 hours. The failure problem of the ceramic RF windows, which at present affects a number of accelerators (but not all), should be solved with a better design. In fact there are no fundamental limits preventing a fully reliable operation of this crucial component.

As a preliminary synthesis of the reliability issue, we can state that at present it should be possible to design a linac having from a few tens to a few hundreds of beam trips per year that are driven by equipment- failure (i.e >100 milliseconds). The trips caused by sparking and similar (non-failure) events can be reduced to a time scale <100 milliseconds, and would have practically zero impact on the transmuter. In the case of a multi-user pulsed beam these short beam trips could be practically undetectable.

The problem of the few long beam trips per year that are expected can be solved with equipment redundancy, that is extra money (second low energy linac and spares on line for the super-conducting part). For a discussion of classes (duration & cause), and frequency of beam trips, see [111].

Beam losses, conversion efficiency

In a 100-mA CW RFQ, where the continuous beam from the source is bunched and accelerated, LEDA measurements show that the total beam losses can be limited to less than 5% [105,106]. At lower current, 80 mA, 1% to 2% beam losses were measured. For the IPHI design, based on the LEDA design with some improvements, improvement by a factor of two is expected. Because the beam energy is low in the RFQ, such losses do not cause a significant activation problem.

From the LANSCE linac experience, low beam losses are expected in the medium energy part of the ADS linac (DTL or CCDTL) and unrestricted hands-on maintenance should be guaranteed. In addition the longitudinal beam dynamics in the ADS low-energy linac will be much superior to those in LANSCE, because of the replacement of the Cockroft-Walton injector-plus-buncher with a modern RFQ. This step eliminates longitudinal mismatches and greatly reduces the tails in longitudinal phase space.

All the multi-particle beam simulations (up to a few million particles) performed so far by LANL, CEA and INFN, using different codes especially implemented for this purpose, have shown that, with proper optical matching and reasonable error tolerances, no particles are lost in the high-energy accelerator and beam transport systems. In practice, this means that the operational beam loss limit desired for hands-on maintenance (<0.1 nA/m) beam along the high-energy part of the accelerator seems straightforward to attain. Given reasonable matching, the problem of beam halo formation, very crucial in circular machines, should be negligible in a short linac within a few tens of lattice periods.

The conversion power efficiency, defined as the ratio between the beam power and the mains power, increases strongly with beam current. Neglecting the marginal power required for magnets, efficiency is determined by the klystron efficiency, the Joule losses in the normal-conducting accelerating structures and by the cryogenic-losses in the super-conducting linac, RF and static. The

last two are current independent so that their proportion of the total required power increases as the beam power decreases.

On the basis of the reference 1 GeV linac design and taking 67% for the klystron efficiency, the estimated required mains power, P_{mains}, as a function of the beam power, P_{beam}, is approximately given by the simple formula:

$$P_{mains} = 1.9 \times P_{beam} + 10\text{-}15 \text{ MW}$$

About half of the 10-15 MW is the power deposited in the walls of the NC low-energy linac plus its water-cooling pumps, and the other half is due to the liquid helium refrigerator. As a consequence, for a beam power of 1 MW, the efficiency is between 6-8%, while for a beam power of 30 MW the efficiency is 42-45%.

Operation and maintenance aspects

In the existing large accelerator complexes, a short (one day or less) maintenance time is scheduled on a weekly or monthly basis and a long one every year. This maintenance scheme reduces the number of unscheduled long beam trips induced by component failures. In the ADS case, with a proper redundancy in the linac design, the short maintenance periods should be suppressed and a single long maintenance period per year (of the order of one month) could be sufficient. This is however a matter of opinion at present, since the required RAMI analysis has not yet been done, but it is believed to be very difficult to operate an ADS linac for a year without the need for significant maintenance. The number of maintenance periods, however, could probably be reduced from one per week to one per month. The transmuter will have to be "refuelled" about once every three months, an activity that takes 10-12 days. Thus it would be easy to obtain about one month's worth of accelerator maintenance, split into three periods over the year.

To obtain this result, one could use the following design criteria:

- All standard power components designed with a suitable margin.
- Two parallel low energy linacs, up to 100 MeV, in two separated tunnels, to maintain one while the other is running.
- 10% of extra modules in the SC linac, to switch off failed component while running the accelerator with a different parameter set. Klystrons have to be in a shielded area for replacement.

Modern fast electronics should guarantee a linac retuning time in the 100-millisecond region, to compensate for failed elements in the acceleration chain, that is compatible with the transmuter thermal response times. A detailed analysis is required to evaluate the optimum compromise between cost and reliability of the accelerator. A similar and parallel analysis should be carried out for the transmuter, in terms of the trade-offs between tolerance to thermal cycling, neutronic performance, and cost.

4.3.4.2 Present status of cyclotron technology

The concept of a cyclotron-based accelerator for ADS is, like the linac-scheme, a multi-stage accelerator facility with a final energy of 1 GeV.

A proposed scheme for such a three-stage 1 GeV design would probably employ the following energy ranges and machine types for the individual accelerator stages:

• DC proton source	at about:	60 keV
• DC-Pre-accelerator, Cockcroft-Walton or Radiofrequency Quadrupole (RFQ)	up to:	0.8 to 4 MeV
• Injector-cyclotron, 4 to 6 sectors	up to:	80 to 120 MeV
• Final stage ring cyclotron, between 8 and 12 sectors	up to:	1 GeV

The accelerator facility at PSI can be seen as a "proof of principle" facility for the generation of high power proton beams using cyclotrons. Since an upgrade program in the years 1990-1995 the 590 MeV Ring cyclotron at PSI routinely produces beam currents of 1.5 mA to 1.7 mA; the highest beam current extracted so far is 2.0 mA. The facility was operated at a beam power of about 1 MW over more than 6 000 h/y in 1999 with the beam being available during 91% of the scheduled beam time. This is considered excellent for the research projects to which the beams are applied. Thus the PSI cyclotron facility is among the accelerators that produce the highest beam power and probably leading in respect to the annual accelerated beam charge. It indicates that a 1 GeV-machine can probably be built based on today's knowledge, experience and technology. The performance, efficiency and costs of such a project can be predicted with fairly high accuracy.

The problem with beam losses at extraction is solved with a design that guarantees well separated turns at extraction. In the case of the PSI ring cyclotron the turn separation equals 8σ of the beam profile at extraction. As an alternative solution to achieve low beam losses at extraction M. Craddock [112] considers extraction by stripping H^--ions, while L. Calabretta *et al.* [113] propose accelerating H^2 ions and also extract the beam by stripping.

The main stage of the PSI facility is a separated sector cyclotron (SSC) with an energy of 590 MeV. The concept of separated magnet sectors was introduced by H. Willax in 1963 [114] in order to provide the high energy gain per turn required to minimise extraction losses. Compared to the "classical" cyclotron layout, which employs "Dees" (RF acceleration electrodes) inserted between the magnet pole gaps, in a SSC, the magnets and acceleration structures (the cavities) use separate sectors, such that there is more room available for the RF structures. Such acceleration cavities can be built much larger and are therefore more efficient. In practice up to 10 times higher Q-values and acceleration voltages compared to the classical "Dee" design can be achieved. The high acceleration voltage results in a high-energy gain per turn, which is the most important parameter in order to generate separated turns and hence avoid beam losses at extraction. At the same time it helps to raise the limit on the beam current imposed by space charge forces. By increasing the number of sectors more RF cavities can also be inserted, which further increases the energy gain per turn. Hence using the highest possible acceleration voltage and adjusting the number of cavities allows a cyclotron design with the energy gain per turn necessary to reach any desired beam current.

Several conceptual design studies on cyclotrons to be used as a final stage in an ADS facility have been published [115-119]. Most of the proposals use an energy of 1 GeV and a beam current around 10 mA (P_{beam} >10 MW), and are essentially based on the design of the PSI ring cyclotron (SSC), which at present delivers up to 2 mA at 590 MeV (P_{beam} >1 MW) using 8 sector magnets and 4 cavities with an RF voltage of 730 kV peak. For the 10 MW cyclotron 10 or 12 magnet sectors are proposed, arranged in a ring with about 15 m diameter; and 6 to 8 cavities with a peak voltage of 1 MV. Higher beam currents could be achieved in larger rings with more cavities and hence a higher energy gain per turn [119]. Sector magnets in an SSC can be of super-conducting design; such magnets – comparable to the ones to be used in a 1 GeV cyclotron – are under construction at present at the RIKEN

laboratory in Tokyo (Super-conducting Ring Cyclotron for the RI Beam Factory at RIKEN) [120]. This concept allows building particularly compact and energy-efficient SSC's.

The limit on the beam current due to space charge effects has been shown to depend on the cube of the energy gain per turn [121,122]. This law has been used to extrapolate properties and the beam performance of cyclotrons for higher beam power levels. The beam current limit is reached when the beam losses at extraction increase since the broadening of the beam diameter due to space charge forces exceeds the turn separation given by the radius and the energy gain per turn. From experience gained in the upgrade of the PSI facility, in which the peak RF voltage in the cavities was raised from 450 kV to 730 kV, it seems indeed, that such extrapolation is feasible [123,124]. Recent results on a 1:3 scale full power model cavity (measured field gradient: 4.2 MV/m at 150 MHz) show that voltages in excess of 1 MV can indeed be expected for the new cavities now under construction at PSI [124]. No technological breakthrough is required, but some challenging (but rewarding and interesting) R&D will still be called for.

The injector cyclotron at PSI, the Injector 2, accelerates a 72 MeV proton beam up to 2 mA for injection into the main stage cyclotron. Again the concept of a separated sector machine is employed, specially designed for high beam intensities with 4 magnet sectors and 2 RF resonators with a RF voltage of 250 kV peak and 2 acceleration gaps each. The same proven technology is used for beam injection and extraction as in the main stage cyclotron.

Interestingly, this cyclotron is operated in a very special, new scheme where the injected beam bunch is matched into a phase space volume that is stable under high space charge forces. In this matched condition the beam bunch is self-focused in the longitudinal and radial directions and kept together by the space charge forces. In contrast to the phase stability in linear accelerators, halo particles are not spread out over the whole bucket, but return to the matched bunch [125-127].

The injector cyclotron for a future ADS facility should preferably also be operated in this matched condition in order to reduce beam losses in the main stage cyclotron through better beam quality and the fact that particles are well confined in a compact phase space volume with little tailing. A final design of such a cyclotron has not been worked out in detail. Alternative solutions have been proposed by Mandrillion *et al.* [117] and at lower energies by Y. Jongen [118].

The DC proton source and pre-accelerator for an ADS facility are similar to the linac design discussed above. Prototypes of 100 mA ion sources exist. The acceptance of the CW beam into the injector cyclotron is, however, much lower than for the combination RFQ and linac. The pre-accelerator for the PSI Inj.2 is a 870 keV Cockcroft-Walton generator. For the acceleration of a 2 mA beam, a CW proton beam of 10 to 12 mA is bunched into the matched phase space volume of the Inj.2 mentioned above. Using a simple code for calculations of on bunched beams under space charge conditions it may be shown that a beam of 50 mA can also be bunched so that the charge corresponding to 10 mA beam current is contained in the same phase space volume [122]. Presumably an RFQ could also be employed as pre-accelerator, but a design study and prototype work as for the linac case have not been made for the lower RF frequency range (around 50 MHz) used in cyclotrons. The tools for such a study exist.

In the following key fields further development and prototype work might be needed:

- Radio-frequency (RF) systems (used for acceleration), with special emphasis on high power CW amplifiers, high power coupling loops, cavities with high acceleration voltages and low spark rates; and generally, "flat-topping systems". Flat-top systems are used to permit a wider particle phase acceptance during the acceleration process. They decelerate the beam, and

stability in voltage and phase becomes difficult to achieve as soon as the power absorbed from the beam exceeds the wall power in the flat-top cavities.

- PSI is pursuing a project to develop and build a RF cavity for >1 MV peak voltage. Relatively simple conditioning will suffice; fortunately, considering the size of cavity, no chemical cleaning, high temperature baking procedures, etc. is needed to reach the relatively modest electric field gradient of ~3.5 MV/m [128].
- Beam collimation at high power, the design of local shielding, as well as installations for remote handling and replacement of highly activated parts.
- Simulation of beam behaviour and longitudinal matching under strong space charge forces for the PSI Inj.2 and an injector cyclotron for an ADS facility, and especially simulating the performance of an RFQ as pre-accelerator. An advanced computer code for this task has been developed in collaboration between CERN, LANL and PSI.
- Injection and extraction systems, now consisting of a combination of electromagnetic and electrostatic (high DC voltage) components, will have to be optimised to handle increased beam losses besides being designed explicitly for low spark discharge rates in electrostatic devices.

Beam power, trip rate and duration, component reliability

The success of the PSI cyclotron at beam currents up to 2 mA demonstrate that it is feasible to obtain the desired performance at high beam intensities with a cycloton-based accelerator provided oncesatisfied with today's reliability.

Operating cyclotrons at high beam power and, at the same time, requiring very few beam trips of short duration and pushing the time lost to unscheduled beam interruptions to negligible levels, poses a relatively recent challenge in the development of cyclotron technology. The priorities in accelerators used in nuclear and particle physics were clearly set to push the technological performance envelope to higher currents, higher precision in terms of energy resolution or higher yields in the acceleration of exotic particles and charge states (radioactive beams). Cyclotrons designed for medical applications (isotope production and irradiation therapy) were for the first time faced with extreme demands for availability (>95% of the scheduled beam time, low unscheduled down times) and a high annual beam time (>7 000 h/y; that is: low scheduled maintenance time). An ADS makes even tougher demands requiring no more than a few 10s to a few hundreds of beam interruptions per year [127], i.e. a beam availability of better than 98%. The demand corresponds more to the conditions typical of a nuclear power plant than those in a research facility. Hence existing facilities are generally not well suited to evaluate what can be achieved with respect to reliability, as can be seen in recent summaries for linacs [111], or cyclotrons [127], which report at best ≈100 trips/week.

In statistical terminology, Mean Time Before Failure is the critical parameter, failure in this context meaning a beam interruption (beam trip) This MTBF in cyclotrons is dominated by sparking in RF and electrostatic deflection devices.

Beam interruptions due to sparking are generally short (duration <1 mn), but still too long with respect to the thermal time constant of transmuters or sub-critical multiplying assemblies. In the PSI facility the trip rate due to sparking is as high as 8500/y under good conditions [129,130]. This, however, is accounted for in the design of the 1 MW spallation target and not considered to be a problem. For cavities in general, further studies are needed, both on the mechanisms that cause a discharge and on measures for fast recovery, so that the beam can be maintained uninterrupted.

Redesigning critical components in terms of geometries that optimise field gradients, and methods of conditioning and surface treatment, are really the only measures available.

The Mean Down Time of the accelerators is generally dominated by unscheduled interruptions of longer duration lasting >1 h, usually due to component failures. To reduce their contribution one has to focus on the different types of systems separately. Preventative maintenance has to be performed, in many cases increasing operating costs. Furthermore, high voltage power devices, as used in beam deflectors (DC), or RF power amplifiers employing vacuum tubes, are more susceptible to failure than their low voltage counterparts. Important in any case are means to assist in quick fault diagnostics, ready-to-operate replacement units, fast interchangeability in all critical components and devices. Increasing the lifetime of critical components, like the electrodes of beam deflection devices, RF amplifiers (tubes) and RF couplers (windows) is an important aspect of reducing unscheduled down-time and operating costs.

Overall operating costs, including power consumption, maintainability and maintenance costs, etc., became an issue when cost effectiveness was being analysed for commercial rather than research applications of cyclotrons. Existing large facilities, however, have up to now not been optimised with these considerations in mind, so there seems to be considerable potential for improvement in this respect [131,132].

Total MTBF and MDT will always depend on the total number of critical components in an accelerator, because failure rates cannot be lowered below a certain reasonably attainable number, and total component redundancy will be precluded by costs. Ultimately, since critical components like RF cavities, high voltage power supplies, RF power amplifiers, etc will always show comparable reliability characteristics, whether used in circular or linear accelerators, a critical component count as low as possible will be an important factor in overall accelerator performance (MTBF and MDT) for ADS systems.

Beam losses

Concerning beam loss, the extraction of the beam from the 1 GeV cyclotron is the most critical point. A good separation between the orbits at the extraction radius is mandatory in order to achieve good extraction efficiency. It is given by the average radius and the number of turns. In the 10 MW facility these parameters have been selected so that the separation of turns is larger than in the 590 MeV cyclotron at PSI. The yearly averaged extraction efficiency achieved in routine operation of the 590 MeV cyclotron is as high as 99.98% and the same extraction efficiency can be expected in a 10 MW facility.

The truly limiting factor is the radiation dose imposed on the personnel involved in repair and maintenance. This is difficult to predict, because the dose depends not only on the beam loss in the cyclotron and beam lines, but to a larger degree on the design of the equipment, the installation of local shielding, on provision for quick and remote removal of activated components into shielded boxes and the use of manipulators [133,134]. It also depends on preventive measures like concentration of activation products in specially designed beam catchers, optimised material selection and, last but not least, the attitude of the personnel themselves in handling of activated components. The serviceability after irradiation is related rather to the design than to the amount of beam produced or lost. In the PSI facility the annual beam production has been upgraded by three orders of magnitude over the past 25 years while the dose to the personnel could be halved in the same time. The conclusion is that, with proper design strategies as mentioned above, a 10 MW facility can be handled, provided the beam transmission remains comparable to that in the PSI cyclotrons.

Power conversion efficiency

The power efficiency of the facility depends very much on the type of accelerator, on the size of the cyclotron and on the amount of beam loading. It is highest if the facility is operated close to the intensity limit, i.e. at the highest possible beam power for a given accelerating voltage. It can, therefore, only be known after a final design has been finished.

The PSI facility is operated at a low beam loading factor. The power efficiency of 10% is, therefore, rather low. The beam power is 1 MW, the RF power needed to produce the high acceleration voltage is about 1.7 MW and if we assume 67% conversion from mains to RF this results in another 1.35 MW lost. The pre-accelerator, injector cyclotron and beam lines need 2.6 MW and the whole infrastructure demand amounts to about 2.5 MW.

For the proposed 1 GeV cyclotron facility the power efficiency has been estimated to be about 36% [129,130]. The beam power is 10 MW, the RF power needed to produce the high acceleration voltage is about 4 MW and if we conversion from mains to RF amounts to 7 MW lost. The pre-accelerator, injector cyclotron and beam lines need 3 to 4 MW and the whole infrastructure demand has been taken as about 3 MW. The figures given are based on extrapolation from the existing facility without any consideration of power-saving technology.

4.3.4.3 Multiplexed modular accelerator concepts

A future ADS facility for transmutation or energy production must have a beam power specification of several tens of MW. If at the same time beam interruptions are restricted to the extremely low 10 to 40 trips/y, then new strategies in accelerator design will be necessary for linacs and cyclotrons. A beam availability so high can only be reached with highly redundant systems, which significantly add to the cost of a future facility. No conclusive judgement on the cost of a possible facility can therefore be formed at present in the absence of detailed projects.

A thorough investigation into the possibility of redundant subsystems has not, to our knowledge, been made. In view of the large investment involved in such a facility a redundancy in the largest sub-system, which is the accelerator itself, has to be considered.

Two scenarios can be thought of and have been proposed:

- Use of for instance three 100 MW linacs to drive four transmutation targets, as proposed by G. Bauer, with splitting of all three beams, on a pulse by pulse basis, into identical portions directed to all four target systems. If one accelerator stops operation the power to the four target systems is reduced by only 33% each [124]. For such a scenario the accelerators most probably would be linacs.
- Use of for instance three 10 MW cyclotrons driving one 30 MW target [129,130]. Again outage of one cyclotron reduces the power on the target system by only 33%. In this scenario one could even think of having a fourth cyclotron as a back-up. In this case the beam could be brought back to full power quickly if one were willing to add to the operation cost the electricity bill for keeping the fourth cyclotron running.

4.3.4.4 Status of current accelerator projects

JAERI and KEK (High Energy Accelerator Research Organisation) have been jointly proposing the multi-purpose complex facilities in the High-Intensity Proton Accelerator Project [135], and the Phase 1 Project was approved for construction. Phase 1 includes: 1) 400 MeV NC linac; 2) 3 GeV Proton Synchrotron (PS) at 1 MW; 3) 50 GeV PS at 0.75 MW; 4) the major part of the 1MW SNS facility; and

5) a portion of the 50 GeV experimental facility. The total budget of Phase 1 is 133.5 billion yen. The phase 1 will be completed within 6 years. Phase 2 will comprise construction of an ADS experimental facility including 400 MeV to 600 MeV SC linac, upgrade of SNS to 5 MW, construction of a neutrino beam line and upgrade of the 50 GeV experimental facility. R&D of super-conducting cavities for a proton linac has been performed since 1995 at JAERI. The vertical tests of 5 cell cavities of $\beta = 0.5$ and $\beta = 0.89$ have been carried out with surface electric fields of 23 MV/m and 31 MV/m, respectively, at 2 k [136,137]. Fabrication of a prototype cryo-module, which includes two 5 cell cavities of $\beta = 0.60$, is in progress and performance test will be made in 2001.

Two projects, IPHI (Injecteur de Proton Haute Intensité) in France and TRASCO (TRAsmutazione SCOrie) in Italy, are also of particular interest in view of the design and construction of an experimental ADS. A collaboration (CEA-CNRS-INFN) between the two projects has been formally established in such a way that, even though each project has its own programme, many important choices are common in order to obtain the maximum profit from the investments made by the two teams.

IPHI is a 1 MW, 10 MeV demonstrator accelerator, that could be used as front end for a high power proton linac. It consists of:

- An ECR source (SILHI, Source d'Ion Légers Haute Intensité), operated at 2.45 GHz with an ECR axial magnetic field of 875 Gauss, able to deliver a 95 keV, 100 mA proton beam.
- A normal conductive radio-frequency quadrupole (RFQ) able to provide a 500 kW, 5 MeV CW beam.
- A drift tube linac (DTL) tank that brings the proton energy up to about 11 MeV.

The SILHI source has already been built, as well as the low energy beam transport (LEBT) line. The design of the RFQ has been completed; its construction is now going on and should be completed by 2002. The construction of a short DTL tank is in progress while the definition of the high-energy beam transport (HEBT) has almost been done.

The objectives of the first part of the Italian TRASCO research programme, leaded by INFN, are:

- A conceptual design of a 1 GeV, 30 mA proton linear accelerator (linac).
- The design and construction of the TRIPS proton source and of the 5 MeV, 352 MHz CW RFQ.
- The study of possible alternatives for the linac part from 5 MeV (the output of the RFQ) up to about 100 MeV.
- The design of the high-energy section of the linac, based on super-conductive elliptical type accelerating structures, as well as the construction of some prototypical super-conducting RF cavities.

A reference conceptual design of the proton source and medium energy section – the 352.2 MHz RFQ and a DTL – has been determined, for a nominal accelerated current of more than 30 mA. The TRIPS proton source has been built and is under commissioning. A detailed design and engineering work of the 352 MHz RFQ has started and a 3 m long aluminium model of the RFQ has been built and measured for RF field stabilisation tests. Technological tests on a short copper section have been done and the first section of the RFQ is in construction. Preliminary studies of an ISCL (Independently phased Superconducting Cavity Linac) – to be used instead of the traditional DTL – have been also done. The conceptual design of the 352 MHz super-conducting LINAC, able to bring the 30 mA proton beam from 100 MeV up to 1 700 MeV, has already been worked out and is mostly based on the LEPII technology. The construction and the tests of the Nb-sputtered copper $\beta = 0.85$ single-cell and multi-cell prototypes cavities has been done at CERN, under a collaboration agreement between CERN and INFN.

4.3.4.5 Concluding remarks

As can be guessed from the preceding paragraphs, a qualitative comparison of the two accelerator concepts cannot be made at present with any reasonable degree of confidence; too many aspects still depend on further R&D in various disciplines of accelerator science and engineering.

4.4 Conclusions

This chapter started by asking if ADS-technology differs from the already developed FR-technology, if there may be synergy in future development and if there are significant bottlenecks that might be foreseen. The previous description has highlighted the main differences especially in the level of development of reactor technology. Synergy is possible and has to be sought in the future in fuel and materials development where particular focus is necessary on the accelerator-reactor coupling, the dynamic behaviour of ADS and the spallation target technology. In general, this chapter has indicated that:

- On the whole, the development status of accelerators is well advanced, and beam-powers of up to 10 MW for cyclotrons and 100 MW for linacs now appear to be feasible. However, further development is required with respect to the beam losses and especially the beam trips to avoid fast temperature and mechanical stress transients in the reactor.
- Various problems related to the accelerator-reactor coupling have still to be investigated. Thereby, special attention has to be given to the target, and especially the beam-window, which is subjected to highly damaging spallation particles and nuclides and corrosive environments which are not encountered in normal reactors. To this end, research programmes have been initiated in Europe and elsewhere.
- While the reactor physics of sub-critical systems is well understood, the issues regarding the dynamic response to reactivity and source transients require investigation because they are the area of greatest difference between critical and sub-critical systems.

5. FAST REACTOR (FR) AND ACCELERATOR-DRIVEN SYSTEM (ADS) SAFETY

5.1 Safety functions and strategies for fissioning systems

5.1.1 Cardinal safety functions for fissioning systems

At a basic level, there are five safety functions to be fulfilled when deriving energy from actinide fission. First, the nuclear fuel must remain contained within a controlled space because of its radiotoxicity; this is traditionally accomplished by use of multiple containment barriers. Second, shielding must be kept in place between humans and the fissioning and fissioned fuel to avoid suffering radiation damage. Third, a heat-transport path must be in place to carry energy away from the fission chain reacting medium to a heat sink; usually an energy conversion plant. Fourth, the rate of release of fission energy in the chain reacting medium must be regulated to remain in balance with the rate of energy delivery to the heat sink, so as not to overheat the containment barriers around the fuel and challenge their integrity; a capacity to store heat in the reacting medium and the heat-transport channel will buffer mismatches of short duration or small amplitude. Fifth, since some 5% of the 200 MeV from each fission event is initially retained in nuclear bonds of unstable fission products, and since these fission products subsequently decay at their natural rates, a means must be provided for transporting heat from the fission products and transuranics in the fuel for all times after the fission event. Failure to satisfy the latter two safety functions could lead to overheating of the fuel with the potential to compromise the integrity of the containment and shielding. Finally, operation of the fissioning devise in a quasi steady state mode requires a balance of neutron production and destruction rates from one generation to the next in a fission chain even as the composition changes owing to transmutation and as the absorption, leakage, and neutron production properties of the fissioning assembly change with changes in composition and temperature.

5.1.2 Safety strategies

Strategies to fulfil the basic safety functions have been developed and refined over many years for conventional (critical) reactors. The strategy employs defence in depth so that no single failure can result in unacceptable release of radiotoxicity; multiple barriers (fuel cladding, primary coolant boundary, and reactor containment building) are used to prevent release of radiation even in accident conditions. Highly reliable systems for controlling and terminating the chain reaction are used to match heat production to removal. Highly reliable, redundant and diverse systems for removing decay heat are provided. High quality construction minimises manufacturing flaws and rigorous maintenance, formal procedures and exhaustively trained and certified operators are used to minimise human error, which could subvert the achievement of the safety functions. Once safety is “designed into” the system, its efficacy is judged by an independent safety regulating authority.

In recent years, the FR design strategy for safety has gone beyond those traditional measures: the system consisting of the reactor heat source coupled to the balance-of-plant heat engine is configured to achieve the safety functions by exploiting the natural laws of physics to the maximum degree

achievable. This safety approach partially supplants the traditional engineered devices by implementing passive systems or using inherent characteristics that play the role of "functional redundancies": in case the upstream line of defence (LOD) should fail, they can achieve the same purpose. The approach is so implemented as to ensure safe response[37] even if the engineered systems which require assured sources of power and highly reliable "active" sensing and switching equipment were to fail, or if multiple, compounding failures and human errors were to occur simultaneously. The passive safety approach can be valuable for all the levels of defence-in-depth, i.e. accident prevention, accident management and consequences mitigation; for instance, the passive concepts can employ inherent reactivity feedbacks to keep heat production and removal in balance. Designs with minimal loss of reactivity upon burn-up and minimal reactivity vested in control rods preclude accidents due to reactivity. Designs having large margins to damaging temperatures and large thermal mass provide the feedback loops with room to operate safely. Designs using buoyancy-driven flows and uninterruptible heat transport paths to the environment remove decay heat without systematic[38] reliance on operating valves or active monitoring. These passive safety approaches for FRs have been demonstrated in full scale tests at EBR-II, RAPSODIE, FFTF, BOR-60, etc. [138].

Given that the safety approaches for FRs are well known, the plan for this chapter is first to describe the chain of logic that gives rise to the salient differences between FRs and that class of ADS studied here. Then the ADS design is broadly compared with a FR so as to identify which of the basic safety functions might be affected by each of these particular salient differences. This is done in Section 6.2.

Following that, in the Sections 6.3 and 6.4 we go through each case having an identified difference; describe how the safety function is provided for a FR; and (if they are different) discuss potential strategies for fulfilling the function for an ADS.

5.1.3 Definition of the subset of ADS considered

This study is considering the roles of fast reactors (FR) and of accelerator-driven systems (ADS) to serve as transmuters in applications of Partitioning and Transmutation (P&T) for nuclear waste

37. A passive system should theoretically be more reliable than an active one. The reasons are that it does not need any external input or energy to operate and it relies only upon natural physical laws (e.g. gravity, natural convection, conduction, etc.) and/or on inherent characteristics (properties of materials, internally stored energy, etc.) and/or "intelligent" use of the energy that is inherently available in the system (e.g. decay heat, chemical reactions, etc.). Nevertheless passive devices can be subject to specific kinds of failure, such as structural failure, physical degradation, blocking, etc. Generally speaking, the reliability of passive systems depends upon:
    - Insensitivity to external interference with the expected performance.
    - Accurate prediction of relevant physical phenomena.
    - The reliability of individual components.

    This is why the need for reliability assessment, even for passive devices, remain a key concern for future reactors.

38. According to IAEA definitions a passive component does not need any external input to operate. The term "passive" identifies a system which is composed entirely of passive components and structures or a system which uses active components in a very limited way to initiate subsequent passive operation. A categorisation has been developed by the IAEA (mainly on the background of thermal hydraulic systems) distinguishing:
    A: physical barriers and static structures.
    B: moving working fluids.
    C: moving mechanical parts.
    D: external signals and stored energy (passive execution and active actuation).

management. A FR serving as a transmuter would be designed as a net burner of some or all transuranic isotopes, i.e. having a breeding ratio less than one. Similarly, for this function the ADS would be designed as a net burner of some or all transuranic isotopes. The general term, ADS, comprehensively includes all manner of non-self-sustaining, fissioning, neutron-multiplying assemblies driven by an external neutron source that is provided by a charged particle accelerator and a neutron-producing target. ADS systems under current worldwide study include both thermal and fast neutron-multiplying media comprising either liquid or solid (lattice) fuel and driven by either cyclotron or linear proton accelerators. The underlying missions targeted for ADS systems span the range from nuclear waste incineration with incidental power production, through power production with integral waste self-incineration, to finally, excess neutron production for the purpose of generating artificial elements by neutron capture reactions in targets.

The Expert Group has confined this study to a subset of ADS configurations: those targeted for nuclear waste incineration with incidental power production, and specifically those which operate on a fast neutron spectrum with an array of solid fuel pins. The scope of this chapter on ADS safety strategy is similarly confined.

Even within this limited scope, a broad range of possibilities is considered. For example, the ADS in scheme 4 is a minor actinide (MA) burners whereas in scheme 3b the ADS is a TRU burner; the physics and safety characteristics of these cases differ because of differences in their values of β_{eff} (which helps to set the degree of sub-criticality of the ADS) and in their swing in reactivity upon burn-up (which helps to set the control strategy). Moreover, the choice of coolant (liquid metal or gas) also distinguishes members of the ADS class considered here.

This class of fast spectrum, solid fuel, waste-incinerating ADS shares with all others a distinction from critical reactors in relying on an external neutron source rather than self-generated delayed neutrons for maintaining the neutron population in balance, with attendant changes in dynamic response and in control strategy. However, the class considered here differs from others in offering unique design and safety challenges in the areas of compensation for burn-up and reactivity feedback characteristics. These unique challenges are traceable to a small number of salient design features, which derive directly from the requirements of the target TRU or MA incineration mission, with incidental power production.[39] The origin of these salient features is discussed next.

5.2 ADS design features that affect safety

5.2.1 Design principles for an ADS burning minor actinide or transuranics and resulting features

Overall purpose; support ratio, and fertile-free fuel

First, the overall purpose of this class of ADS is to function as one element of an integrated nuclear power enterprise comprising conventional and advanced power reactors for energy production and ADSs for reducing the radiotoxicity of the nuclear waste produced by these power reactors before entombment in a geologic repository. The radiotoxic materials targeted for incineration may be minor actinides or may be transuranics, depending on the configuration of the overall enterprise. The ADS may also incinerate selected fission products.

The transuranics are fissioned in the ADS to transmute them to fission products with shorter radiotoxic half-life. A fission of one TRU atom releases approximately 200 MeV of energy. Expressed

39. Not only ADS, but also FRs for pure burner missions (i.e. no fertile material in the fuel) differ dramatically in behaviour from standard FR's and require addressing new safety challenges.

in different units, 200 MeV/fission corresponds to about 1 g TRU incinerated per MWth day energy release. For a fissioning device, the incineration rate of TRU depends on the power rating of the heat removal equipment and nothing else, whether in ADS or a reactor. While the ADS will probably use the heat liberated in the transmutation of transuranics for power production to offset the cost of its operation, its primary function is to reduce the transuranic and long-lived fission product inventories emanating from the power reactors deployed in the nuclear enterprise.

The "support ratio" of the integrated power producing enterprise is the ratio of the power of the reactors to that of the ADS in the enterprise. A large support ratio is targeted for the ADS designed for waste incineration with incidental power production because it relaxes the demands on ADS cost and energy conversion efficiency, inasmuch as the ADS represents only a small segment of the overall enterprise. The primary purpose of the ADS is then to maximise incineration rate per unit of heat that has to be removed from the device so as to minimise its cost. This leads to a need to avoid in-situ production of new transuranic elements and thus to avoid use of fertile atoms (U^{238} or Th^{232}) in the ADS fuel composition.

A 3 000 MWth ADS plant operating for 300 days per year transmutes about 900 kg of TRU into nearly 900 kg of fission products, and releases 9×10^5 MWth days of energy together with an excess of neutrons (~2.5 neutrons/fission). If the fuel were to contain fertile atoms, some of the excess neutrons which are not required to sustain the chain reaction would unavoidably be absorbed in the fertile material and would produce new TRU atoms in-situ. To avoid this, fuel for the class of ADS considered here will contain no fertile (U^{238} or Th^{232}) atoms so as to maximise the net rate of TRU incineration per unit of heat which has to dispelled (and per unit cost of equipment to dispel it). Fertile-free fuel is the first salient design feature shared by proposed ADS systems of the class considered here.

Multiple recycling

Given the goal of totally consuming the TRU or MA fuel by fission and a lattice of solid (rather than fluid) fuel, it is evident that multiple recycling of the fuel will be required. The ADS will operate on a closed fuel cycle with a feedstock of TRU or minor actinides arriving from the power producing reactors of the overall enterprise, and the system will discharge waste containing fission products but hardly any actinides in a form destined for a geologic repository. Internal multiple recycling the ADS fuel will be required to reconstitute the fuel with fresh cladding, because the fluence required for total fission consumption exceeds the neutron damage endurance of any known cladding. Recycle is also required to inject new feedstock into the ADS lattice to sustain the neutron multiplication within its design range as well as to extract the fission products destined for geologic disposal. Although not unique to ADS, this need for multiple recycle constitutes a second salient feature of the ADS considered here.

Except for the "once-through cycle" (scheme 1), the recycle step in the overall complex is where the waste stream to the geologic repository is generated. It is composed of fission products and trace losses TRU or MA which escape the recycle and refabrication processes for return to the ADS or FR. These trace losses waste must be minimised if the ADS is to achieve its assigned mission. It is clear that both the trace loss per recycle pass and the number of recycle passes control the ADS contribution to the complex's total loss, and that therefore a high average discharge burn-up from the ADS is desirable. Moreover, since the radiotoxicity per gram and half-life of the various TRU or MA isotopes vary, it is desirable that the transuranic isotopic spectrum achieved upon multiple recycle should be

favourable in terms of long-term toxicity (including that of all post-emplacement decay daughters).[40] The ADS neutron spectrum determines this.

Fast neutron spectrum

Upon multiple recycling to achieve total fission incineration, the TRU or the MA isotopic composition of the LWR spent fuel feedstock evolves to a different asymptotic composition depending on the neutron spectrum to which it is subjected. The ADS of the class considered here is designed to operate in the fast neutron range so that all transuranic elements stand a good chance of fission upon a single neutron absorption, minimising the production of heavier transuranic isotopes with a less favourable long-term radiotoxicity burden per unit lost to the waste stream. Table 5.1 (see also Table 2.1 and respective discussion in Chapter 2) shows that a fast neutron spectrum is essential for total consumption of MA and is preferable to a thermal spectrum for burning TRU. Moreover, it will become clear later that per unit of energy released, the fractional reduction in TRU content upon irradiation, and its effect on reactivity, are smaller and can be more easily compensated if the in-core fuel inventory is large, as is the case when the spectrum is fast. A fast neutron spectrum is the third salient design feature of the class of ADS systems considered here.

Table 5.1. **Values of D (neutron "consumption" per fission) for ^{238}U and different fuel types (-D: neutron production)**

Top-up fuel	**Thermal TRU burner (ADS)**		**Fast TRU burner (ADS)**		**MA burner (ADS)**		**Critical fast reactor**	
	η_{ec}	-D	η_{ec}	-D	η_{ec}	-D	η_{ec}	-D
Uranium-238	0.92	-0.24	1.28	0.64	1.28	0.64	1.41	0.85
Plutonium	1.15	0.40	1.80	1.34	1.74	1.28	2.03	1.53
Minor actinides	0.89	-0.37	1.37	0.86	1.33	0.79	1.52	1.10
Transuranics	1.11	0.30	1.75	1.29	1.69	1.23	1.96	1.48

Choice of coolant

Although not a design feature which distinguishes ADS from FR concepts, the choice of coolant, plays a strong role in both core design and safety strategy for FRs and ADS alike. It is useful for clarifying the following discussions, therefore, to explicitly include coolant choice among the ADS distinguishing features. Since the neutron spectrum is to be fast, the candidate coolants are sodium, heavy liquid metals (e.g. Pb or Pb-Bi) and gas.

Features shared with fast reactors

As indicated in Figure 2.4, fast reactors are themselves employed to consume TRU or MA in several of the fuel cycle schemes studied by the Expert Group. However, whereas the features discussed above (fast neutron spectrum, multiple recycling and alternative coolant choices) are shared by those FRs with the ADS, the fast reactors do not employ fertile-free fuel. The neutronic properties of fertile-free fuel dictated by the a-priori requirement to maximise the support ratio motivate the features of the ADS which most clearly distinguish it from a fast reactor.

40. For example, in the proposed US geologic repository with an oxidising environment, ^{237}Np (a post emplacement daughter in the ^{241}Pu$\rightarrow$^{241}Am decay chain) dominates the long-term toxicity.

Features that are unique to the ADS

Fertile-free fuel is prescribed for the ADS in order to maximise the ADS support ratio in the power producing energy complex. The neutronic properties of fertile-free TRU or MA fuel – its η value and its delayed neutron fraction – give rise to the remaining distinguishing ADS features: specifically a sub-critical operating state driven by a spallation neutron source.

Sub-delayed critical operating state

Transuranic fuel containing no fertile atoms exhibits a delayed neutron fraction for fast fission in the range of 0.0015 to 0.0020 i.e. about half the value for a conventional FR and about a sixth the value for a conventional LWR. Table 5.2 displays ν_d for fast fission of various actinide isotopes and shows that even at only ~10% contribution to fissions, as is typical for a FR, fertile U^{238} or Th^{232} would contribute very significantly to delayed neutron fraction. The delayed neutron fraction is remarkably small for fertile-free fuel compositions, and therefore the margin to prompt criticality is correspondingly small. This feature, when combined with considerations of reactivity feedback discussed next, leads to another salient design feature of ADS specifically on grounds of safety.

Table 5.2. **Delayed neutron fractions**

Isotope	$\mathbf{Y_d/Y_{total}}$
^{238}U	0.0151
^{232}Th	0.0209
^{235}U	0.00673
^{239}Pu	0.00187
^{241}Pu	0.00462
^{242}Pu	0.00573
^{237}Np	0.00334
^{241}Am	0.00114
^{243}Am	0.00198
^{242}Cm	0.00033

$\Rightarrow$ <u>10% Fertile fission raises β in fertile containing fast reactor fuel</u>

$$\beta(^{238}\text{U}) \qquad \beta(^{239}\text{Pu})$$
$$0.10 \times 0.0151 \quad + \quad 0.90 \times 0.00187$$
$$= 0.00151 \quad + \quad 0.00168$$
$$= 0.00319$$

(Nearly twice the β of fertile-free fuel)

In a fast spectrum, with a fertile-free fuel and no parasitic neutron absorption, the k_∞ of the lattice will be determined by the transuranic η values:

$$k_\infty = \frac{\left(\nu \sum_f\right)_{TRU}}{\left(\sum_a\right)_{TRU}\left[1 + \frac{\sum_{a\ \text{parasitic absorber}}}{\sum_{a\ TRU}}\right]} \qquad (1)$$

$$\simeq \eta_{TRU}$$

A reactor fuelled purely with transuranics will exhibit $k_\infty \cong \eta \cong 1.5$ to 1.8 in a fission spectrum. A critical mass of such fuel composition is small, in the range 5 to 20 kg.

ADS designs, even with fertile-free fuel, are not without parasitic absorption. Calculations for Pb-Bi cooled TRU incinerator ADS indicate that structure, fuel diluent (Zr in this case), coolant and fission products will absorb about half as many neutrons as the TRU itself, reducing the k_∞ by a third [26,139].

Given that the neutron reproduction ratio per fission chain generation, k, is specified by k_∞ and the leakage probability, LP:

$$k = k_\infty (1\text{-}LP) \qquad (2)$$

a fertile-free fast spectrum lattice will experience a leakage probability in the region of 20%:

$$(1 - LP) \simeq \frac{0.98}{1.2} \Rightarrow LP \simeq 1/5$$

The neutron leakage in a fast neutron lattice is sensitive to the assembly geometry because of the long neutron mean free path. Subtle geometry changes induced by thermo-structural effects dependent on power to flow ratio, such as fuel bowing, grid plate expansion, etc., will change the neutron leakage fraction in response to power and flow changes. With 20% of the neutrons leaking, changes in these structurally dependent leakage rates would have to be kept within 1% of their values to avoid exceeding the value of β_{eff}:

$$0.20 \times x = 0.0015$$

$$x = \frac{15}{2\,000} < 3/4\,\%$$

But thermo/structural power feedback cannot be designed for nor can it be controlled to a degree of precision less than 10% [140,141]. Even if parasitic absorbers were purposely added to the fuel to consume the majority of the excess neutrons and reduce the leakage fraction to 5%, it would not change the situation; reactivity changes due to leakage fraction could still not be held reliably to less than a dollar.

Taken by itself, a power-dependent reactivity feedback which exceeds a dollar is not uncommon in FR designs. For example, in an oxide-fuelled fast reactor, the Doppler reactivity vested in the temperature difference between fuel and coolant is in the region of several dollars.

But variability as well as controllability is the issue here. In an ADS functioning as a waste burner, the fuel composition itself and its η value and β value can be expected to vary from loading to loading as the source is spent fuel from LWR or FR, differing in burn-up, cooling times and origins. These feedstock variabilities change not only k_∞, and thermo-structural feedback but also the delayed neutron fraction and even the offset from prompt criticality.

Taken all together, the variability and uncontrollability of the reactivity state in an ADS lattice relative to the reduced offset between delayed criticality and prompt criticality leads to the fourth and dominant design feature of ADS systems, the use of an external source to drive a sub-critical assembly. To avoid any potential for unintended feedback of reactivity, induced by fluctuations in power/flow ratio, to carry the system into the super-prompt-critical regime, the geometry and composition of the ADS assembly are configured so that the operating margin to prompt criticality will always substantially exceed the maximum power/flow reactivity feedback, allowing for the expected variability in the values of η and β_{eff} due to differing feedstock compositions. But the resulting offset then exceeds the value of the delayed neutron fraction itself, so it makes the operating point of the ADS lattice sub-delayed-critical. An external source is required, therefore, to drive a continuing fission reaction with the fissioning system multiplying the externally supplied neutron source. A sub-delayed-critical operating state driven by an external neutron source is the fourth and dominant salient design feature of all ADS.

Spallation neutron source

The size of the neutron source required to drive a sub-delayed-critical ADS depends on both the desired heat rating and on the degree of neutron self multiplication of the lattice, which depends on the degree of sub-criticality. The classical derivation of the asymptotic neutron population resulting from injecting a sequence of source neutrons into a neutron multiplying medium provides for superposing an infinite number[41] of sub-critical fission chains following each source injection:

$$\text{Power a}\left\{\begin{matrix}\text{asympotic}\\ \text{neutron}\\ \text{population}\end{matrix}\right\} = S\Lambda\begin{cases}1\\ +k & \text{for those multiplied once}\\ +k^2 & \text{for those multiplied twice}\\ \vdots\\ +k^n\\ +\\ \vdots\end{cases} \qquad (3)$$

$$= \frac{S\,\Lambda}{1-k} \quad \text{when } k < 1$$

Here, $S = \frac{\text{neuts}}{\text{sec}}$ and Λ = prompt neutron generation time (s.) $\simeq 10^{-7}$ s. Allowing for the fact that energy is released in the generation of fission-multiplied neutrons but not of the external source neutrons (subtracting the external source term, $S\Lambda$, in the power series above), and with appropriate conversion factors, the total fission rate (Power) and the power density are related to the external source and neutron reproduction factor k as:

$$P \simeq \frac{S}{\frac{1}{k_0}-1} \equiv \frac{S}{-\rho_0} \text{ where reactivity, } \rho_0 \equiv \frac{k_0-1}{k_0} \qquad (4)$$

With 1 g of TRU or MA incinerated per MWth day, ADS facility heat ratings must lie in the range of 1 000 MWth or more to support any reasonably sized energy complex. With the required offset from prompt criticality no less than 2 or 3% $\Delta k/k$ (i.e. source neutrons multiplied in the fission chain by no more than 30 to 50), it is clear that no passive neutron-emitting source is strong enough to meet the requirement for ~1 000 MWth power rating. However, plausible extensions in proton beam current capability targeted for linear accelerators (i.e. beams of multi megawatt levels), could achieve the required neutron source strength by driving a heavy metal spallation target.

This leads to the fifth salient design feature of an ADS; namely the external source must derive from a spallation neutron target driven by a high power proton accelerator.

5.2.2 Summary of salient features for ADS TRU and MA burners

The distinguishing features of the type of ADS considered by the Expert Group derive directly from:

- The mission assigned to it in the energy complex, namely TRU or MA (and LLFP) incineration for waste management in the integrated energy complex with power generation only to offset cost; combined with,

41. The power series is extended only for a finite but large number of terms in the light of the discrete lower bound on neutrons equal to one.

- The *a priori* assumptions on scope of cases considered by the Expert Group, namely fast spectrum, solid fuel, and maximised support ratio.

The resulting distinguishing features are:

- Those shared with FR:
  - Fast neutron spectrum.
  - Solid fuel lattice.
  - Multiple recycling.
  - Choice of coolant: Na, Pb-Bi, or gas.

- Those unique to ADS:
  - Fertile-free fuel.
  - Sub-critical operating state.
  - Spallation neutron source driven by a high power proton beam.

5.2.3 Optimising the support ratio

Since the ADS is considered as an element of the overall energy supply enterprise, the support ratio should be optimised. Along with the several schemes considered in this study (single strata, double strata, etc.), another optimisation considers trading off the removal of all fertile isotopes from the fuel, thereby increasing the support ratio of the overall enterprise against adding some fertile isotope content to the fuel and perhaps lowering the cost of an ADS owing to potentially improved safety characteristics deriving from the fertile content's increased β and Doppler coefficient of reactivity. That optimisation is beyond the scope of this study and in any case it would be conducted country-by-country on the basis of their individual policies, existing infrastructure and financing situation. Hence, in this chapter we discuss only the specific case of fertile-free fuel – an extreme but perhaps not ultimate optimisation.

5.2.4 Overview of safety-related issues attendant specifically to ADS design features

The salient design features of ADS give rise, in some cases, to different safety-related issues and different approaches to fulfilling the six cardinal safety functions for fissioning systems as compared with the issues and safety strategy which apply for a FR. Table 5.3, which tabulates salient feature versus required safety function, identifies where these differences exist. In Table 5.3, the effect of these features on strategy for meeting safety functions is indicated for both normal operational and abnormal situations. The rows of Table 5.3 are briefly overviewed here. The entries in Table 5.3 indicate where in the subsequent sections of this chapter the safety strategies to accommodate these new issues are discussed in more detail.

Table 5.3. **Areas where ADS features provide unique differences from FRs***

	Neutron balance		**Heat removal**		**Regulation of power/ Flow & Reactivity feedback**
	Normal	Abnormal	Normal	Abnormal	Normal
Fast spectrum	5.3.1.1 Neutron balance 5.3.1.2 Compensating burn-up loss				
Choice of coolant			5.3.2.2 Fuel & clad compatibility clad breach sludge 5.3.2.2 Na burning Pb-Bi Po issue	5.3.2.2 Freezing potential of liquid metals 5.3.2.2 Depressurisation/ LOCA potential of gases	
Inert matrix (fertile-free) fuel	5.3.1.1 Neutron balance 5.3.1.2 Compensating burn-up loss				
Sub-critical state	5.3.3.1 Asymptotic response of neutron density to source and to reactivity changes		5.3 Operational safety related strategies for FR & ADS systems		5.3.3.2 Dynamics of response to source or to reactivity changes 5.3.3.3 Thermo/structural response to abrupt source changes 5.3.3.4 Trim control options
Spallation neutron source	5.3.1.2 Source importance as a control mechanism		5.3.2.1 Power peaking effect on heat removal		
Multi-recycle		Criticality limited equipment & batch size			

Table 5.3. **Areas where ADS features provide unique differences from FRs* (cont'd)**

	Regulation of power/ Flow & Reactivity feedback	**Containment**		**Shielding**		**Decay heat removal**	
	Abnormal	Normal	Abnormal	Normal	Abnormal	Normal	Abnormal
Fast spectrum							
Choice of coolant	5.3.2.2 Void coeff. of liquid metals 5.4.4 • Pb tamping in HCDA • Coolant channel blockage in HCDA			5.3.2.2 Choice of coolant 5.4.6 Coolant activation products			Loss of pressure in gas systems
Inert (fertile-free) fuel							
Sub-critical state							
Spallation neutron source		5.4.1 Beam tube penetration through containment & vessel	5.4.2 Entry alignment Dropping hazards power density asymmetry hazard	5.4.2 Entry alignment Refuelling activation of magnets 5.4.5 Beam tube activation 5.4.1 Bending magnet activation			5.4.3 LOCA for gas or for side entry beam
Multi-recycle	5.4.4 • HCDA energetics w/no Doppler • Pb tamping in HCDA • Gas coolant blockage in HCDA			Shielding, pyrophoricity, volatility of MAs feedstreams		Heat load of Mas	

* Not only ADS, but also FRs which are targeted for pure burner missions (i.e. no fertile material in the fuel) differ dramatically in behaviour from standard FRs and require solution to new safety challenges.

Spallation neutron source effects

The most readily obvious physical difference is due to introducing the proton beam tube. First is its topological effect on the strategy of multiple containment to provide defence-in-depth. In standard FRs, the fuel is contained first by its cladding (or by multiple layer ceramic barriers in particle fuel), then by the primary cooling circuit boundary and lastly by the containment building. In the ADS, if driven by linacs, the proton beam tube penetrates the last of these and employs a metallic beam window[42] as a topological continuation of the primary coolant boundary. The safety issue pertains to the preservation of defence in depth for the containment and shielding functions. In a FR, similar topologies result from steam lines, which penetrate the containment, and from intermediate heat exchanger (IHX) tubes which represent a topological extension of the primary coolant boundary. In BWRs, the steam lines penetrate both the containment and the reactor vessel. Fast acting valves at the containment boundary of steam pipes and robust heat exchanger tube walls are the means of safety strategies used in standard reactors. For the ADS, the window operates in an environment especially hostile in its temperature and the proton and neutron bombardment that it experiences, and the hazard due to the multi-megawatt proton beam potentially impinging on these barrier boundaries is unique to an ADS.

The beam tube also introduces new issues in the area of shielding, by offering a streaming path from the fissioning lattice to the exterior of the vessel. Finally, being several tens of centimetres in diameter, the evacuated beam tube presents a new issue in the form of a potential increase in reactivity should the beam tube flood and decrease the neutron leakage. The degree of reactivity offset from prompt critical must be sufficient to accommodate such potential flooding safely.

The presence of an external neutron source also has an effect on power density peaking factor [142] in the transmuter core and on the change in power peaking as k_{∞} of the lattice changes with burn-up and as the ratio of source to fission multiplied neutrons is altered by changes in source strength. Also, depending on the geometry of the beam tube entry, the fuel loading pattern may be azimuthally asymmetric, again affecting the power density profile. A design strategy which relies on increased margins so as to accommodate local shifts in power/flow ratio, while undesirable for a dedicated power producer, is quite consistent with the ADS mission wherein power production is only an supplementary function.

Fertile-free fuel effects

A second obvious safety related difference derives from fertile-free fuel, which excludes the traditional Doppler contribution to prompt negative feedback of reactivity in a FR. Small, but not zero, Doppler feedback has been accommodated (and beneficially exploited as a passive safety mechanism in metal-fuelled FRs where a low fuel melting point provides a fast-acting mechanism to terminate HCDA), but FRs with high melting point oxide-fuels rely heavily on prompt Doppler feedback to limit the severity of HCDAs. A mechanisms to terminate HCDA will have to be devised for an ADS with fertile-free fuel.

Pure TRU or MA fuel also presents issues in recycle batch sizes and processing geometries because of a small critical mass. Experience does exist with pyrochemical recycling of metal-fuel in small, discrete batches, limited by criticality constraints. This issue would require much greater care in the case of continuous aqueous reprocessing.

42. In some cases, a fluidic “windowless” target design is considered.

The absence of internal conversion of fertile to fissile species with burn-up will also place demands for reactivity compensation on other design strategies, such as changes to source strength or source effectiveness, batch refuelling, or moving absorber control rods. For minor actinide burners, *in situ* isotopic transmutations mitigate but do not eliminate this issue.

Coolant choice effects

The distinguishing characteristics of the coolant choices relate to system pressure, lattice power density, effect on the neutron spectrum, and chemical activities as tabulated in Table 5.4.

Table 5.4. **Coolant characteristic features**

	Na	**Pb-Bi**	**He**
System pressure	Low	Low	High
Lattice power density	High	Low	Lower
Neutron spectrum	Hard	Harder	Harder
Chemical activity	High	Low	None

These distinguishing features permeate the entire approach to ADS design and the resulting safety strategies. High pressure gas cooling introduces a loss-of-coolant vulnerability but eliminates issues of chemical compatibility. Gas cooling shares with Pb-Bi cooling the need for a low power density in an open fuel pin lattice (which leads to a potential for reactivity additions should hypothetical pin disruption lead to compaction and reduced leakage). The potential for blockage from foreign objects, sludge, or re-freezing fuel debris remains an issue even in an open lattice. The list goes on and on for ADS and FR alike, and is addressed in detail in the later sections as indicated in Table 5.3.

Sub-critical operating state effects

A fundamental distinction arises between ADS and FR in the differences in dynamic response between critical reactors and sub-critical source-driven neutron-multiplying lattices. In a source-driven system, a change in strength or effectiveness of the source or a change in reactivity will cause the neutron population and power level to adjust to a new asymptotic level; whereas in a critical reactor a change in reactivity leads (in the absence of reactivity feedback) to an asymptotic or exponential evolution in the neutron population. While a favourable ADS safety feature derives from its asymptotic rather than exponential response to a positive reactivity insertion [143], a safety challenge still remains in assuring that increases in strength or effectiveness of the source cannot take the ADS to damaging over-power conditions. Eq. (4) indicates that for instance at a Beginning of Cycle offset of $-\rho_o$ equal to 3% $\Delta k/k$ and a reactivity loss of 6% $\Delta k/k$ on burn-up, the source to maintain the End of Cycle power level would have to exceed the Beginning of Cycle requirement by 100%, leading to a potential over-power by a factor of two should the full source strength be introduced prematurely. Options to minimise burn-up reactivity loss include multi-batch fuel loading [139] and optimal mixes of plutonium and minor actinides [144] to flatten the reactivity change with burn-up. However, given fertile-free fuel, it has proved impossible for ADS designers to achieve small burn-up reactivity loss, so that compensation must be by external changes in reactivity (control rods) or in strength or effectiveness of the source. In every case then, a potential for over-power exists, and a highly assured beam trip capability is essential. If heat removal were to fail (loss of flow or loss of heat sink), then

the beam would likewise have to trip off promptly to avoid overheating and melting of the fuel [145,146].

For an ADS, the operating point is offset from prompt criticality by $(\beta + \Delta_o)$ where Δ_o is the sub-criticality operating point. This is compared to an offset of only β for a critical reactor. The effect of this is a lower sensitivity to reactivity feedbacks in the ADS than in a FR. This difference gives rise to a need for different strategies in passive safety concepts to keep heat production and removal in balance.

Of equal consequence relevant to safety and controllability is that the time constant of dynamic response of an ADS is the lifetime of prompt neutrons ($\sim 10^{-6}$ s.) rather than of delayed neutrons (effectively ~10 s.) which forms the experience base for a FR.

In summary, the dependence on source neutrons rather than on delayed neutrons to maintain the fission chain reaction leads to more abrupt responses to control changes, and reduced benefit from power/flow dependent reactivity feedbacks, but it provides a new degree of design flexibility in the ability to control the offset from prompt criticality of the operating state.

The issues of dynamic response to reactivity and source changes are the area of greatest difference between FR and ADS in safety-related characteristics and are an area where few precedents exist in the FR experience base.

Accelerator safety

The accelerator brings with it the traditional accelerator safety issues (high-voltage, control of worker dose owing to components activated by beam divergence, etc.). Since these issues are not peculiar to ADS applications, they are left to be handled in accelerator-specific publications.

Recycle facility safety

The recycling and refabrication processes for TRU and MA fuel introduce issues of criticality, pyrophoricity and atmosphere control; these are discussed briefly in Section 5.5 and do not differ in character from those in FRs intended for TRU or MA. In either the ADS or FR case, however, the small critical mass of fertile-free fuel and the demands on shielding and atmosphere control when working with high concentrations of minor actinides (displaying characteristics of spontaneous fission, neutron emission, and low temperature volatility) raise new challenges compared with current practice.

5.2.5 Guide to location of detailed discussions of safety approach

Table 5.3 indicates the top-level correlation between the salient design features of an ADS and the basic safety functions, which must be provided in any fissioning system. In the table, each safety function pertains to two situations, normal operations and abnormal events, which in general are discussed in different sections.

The order of discussion presented below follows a general pattern covering safety strategy primarily for normal operation in Section 5.3 and then primarily for abnormal conditions in Section 5.4:

5.3.1 Managing the neutron balance and burn-up-induced decrease in source multiplication.

- Fast spectrum and fertile-free fuel effects.
- Effects of source effectiveness.

5.3.2 Removal of heat.

- Effects of coolant choice and source-induced power-peaking effects.

5.3.3 Dynamic control

- Effects of sub-critical operating state dynamics.
- Value of reactivity feedback vs. adjustment of source strength.

5.4 Containment, shielding and removal of decay heat.

- Implications of the source beam transport tube for containment, shielding and refuelling.
- HCDA termination strategy.
- Passive safety response.

5.5 Fuel cycle facility safety.

- Effects of fertile-free fuel.

5.6 Summary.

The entries in Table 5.3 provide a guide for locating the discussion.

5.3 Strategies related to operational safety for FR & ADS systems

5.3.1 Effects of fertile-free fuel and fast neutron spectrum in ADS

5.3.1.1 Neutron balance and choices for disposing excess neutrons

As shown in Eqs. (1) and (2), a multiplying lattice of fertile-free fuel operating on fast neutron chains produces a vast excess of neutrons upon each fission. Only one neutron per fission is required to produce the next fission in the chain reaction. Thus, as many as 50% of the released neutrons are discarded either by leakage or by absorption in non-fuel material. The external source is not needed to produce sufficient neutrons for incineration, but rather because of the approach chosen to accommodate a changing fuel composition and thermo-structural reactivity feedback that may exceed the delayed neutron fraction.

Structure, coolant, fission products and inert diluent in the fuel will parasitically absorb around half of these excess neutrons; the question is how best to dispose of the rest; whether by absorption in

the fuel itself by admixing a parasitic neutron absorbing material with the TRU, absorption in other structures added to the lattice but outside the fuel, or leakage?

The following comments concern the trade-off between leakage and parasitic capture in the lattice:

- When the probability of leakage is large, even small changes in it due to thermo-structural effects will cause changes in reactivity feedback. This has two undesirable effects. First it requires an increased offset from prompt-criticality, thereby requiring a larger spallation source strength (larger accelerator) for a given power level. Second, it requires constant adjustments of the source strength or of a reactivity trimmer to compensate and hold power constant as feedback relaxes following a change in power level (discussed in 5.3.3.3).
- A design strategy requiring most of the excess neutrons to leak from a fast spectrum lattice, whether FR or ADS, would create an unnecessary risk of re-criticality upon compaction under hypothetical severe accident conditions, even given that the fuel might float in the coolant (in the case of Pb-Bi case).

These considerations favour parasitic absorption over leakage as the means to dispose of excess neutrons. Should it be in the fuel pin itself, or in separate discrete absorber pins dispersed in the lattice? Several observations include:

- Pure TRU or MA fuel without an absorbing diluent has a small critical mass, requiring small batches in fabrication and recycling processes with a consequent impact on costs.
- Using fixed absorbers, separate from the fuel, would provide an opportunity to "zone" the k_∞ radial distribution without varying the composition of fuel. Such zoning will be highly desirable to overcome power peaking, and shifts in it with burn-up, which are inherent in source-driven lattices.
- On the other hand, fixed absorbers separate from the fuel present a vulnerability should some abnormal event remove the absorbers from the lattice and thereby add reactivity.
- These considerations suggest that absorbing diluent in the fuel itself is the preferred choice. Many considerations will affect the choice of material for the purpose:
  - The absorber composition should be chemically similar to TRU so that it will naturally follow the TRU or MA during chemical separations on recycle (e.g. consider choices already made for CAPRA).
  - The diluent should preferably be a resonance absorber so as to achieve a measure of prompt Doppler feedback.

It may be desirable to poison the core even further with diluent absorbers as a way to increase the critical mass of the core despite a still smaller leakage fraction. As discussed the following section, this will reduce the fractional decrease of fissile content per unit of energy released and will therefore reduce the fractional increment in reactivity or source required to compensate for burn-up. An opportunity for neutronics optimisation occurs here – to balance discharge burn-up and fluence at their respective limits so as to minimise the number of recycle passes required to achieve complete transmutation.

In summary, safety and other design issues all favour the use of absorbing diluents in the fuel rather than leakage as the means to dispose of excess neutrons in a lattice of fertile-free TRU or MA fuel.

5.3.1.2 Compensating burn-up reactivity loss

Countervailing cost saving goals exist in ADS design. The required source strength can be made smaller (less expensive and requiring less electricity to operate) by making the fission multiplying assembly less sub-critical so that it produces more fissions per unit source. To minimise the size and cost of recycling equipment and to minimise TRU losses to the waste steam, it is desirable to maximise burn-up at discharge. These two goals work in opposite directions because each additional fission (to increase discharge burn-up) increases the sub-criticality, which means the source may have to be increased in order to maintain power at a constant level over the refuelling cycle.[43] Thus, if we hold the power constant by adjusting source strength, the source will be oversized for most of the cycle. Alternately, if we hold the source constant and let the power fall with burn-up, the heat removing equipment will be oversized for most of the cycle.

If we reduce discharge burn-up so as to mitigate reactivity loss, the recycle equipment will be larger than it could have been, and the unavoidable trace losses of TRU to the waste stream will be larger than otherwise. Alternatively, if we refuel partial batches frequently (to approximate continuous refuelling) we will reduce plant capacity factor and all plant equipment will be idle for a greater percentage of the year.

In light of these trade-offs, the ADS design must certainly find a way to reduce burn-up reactivity loss per unit energy release, and then as complementary measures:

- Compensate for declining reactivity with frequent partial core refuelling as burn-up occurs.
- Load excess fissile material and then compensate for loss of reactivity by withdrawing external neutron-absorbing control rods.
- Increase the source strength as burn-up occurs.
- Increase the source effectiveness as burn-up occurs.
- Some combination of the above.

The first step – which clearly should be taken – is to increase the critical mass so that each TRU atom fissioned is a smaller fraction of the total fissile mass and will lead therefore to a smaller percentage increase in sub-criticality thus requiring a smaller percentage change in compensating absorber reactivity or source strength.

- This is one additional reason to employ a fast spectrum system where critical mass is larger than in a thermal spectrum system.
- It is reason also to increase neutron wastage by diluent absorption, lowering k_∞ to nearly unity and requiring a larger and less leaky lattice of maximum fissile inventory.

The second approach is to refuel the core in parts and provide for an adjustable parasitic neutron absorber (a control rod) which can be moved so as to hold sub-critical reactivity constant as fissile content is burned out. This is well-established and reliable technology from FR experience.[44]

43. Alternative burn-up compensation options are discussed next.
44. Semi continuous refuelling, as in a CANDU, has received some consideration, but is not currently being pursued.

The third potential approach is to adjust the proton beam current and resulting source strength. While widely discussed, this approach requires an accelerator substantially oversized for all but the end-of-cycle conditions. Besides cost considerations, a safety vulnerability is introduced should the full beam power be applied at the beginning of a cycle. An alternative approach of operationally running a constant proton beam but adjusting the effectiveness of the spallation neutron source can be considered. The spatial dependence of source effectiveness could possibly be exploited by moving the spallation target from top of core to core centre, making it more effective. Alternatively, the energy spectral dependence of source effectiveness could be exploited by introducing the spallation neutrons into the core at an ever-increasing energy. As shown in Figure 5.1, the spallation neutron emissions spectrum is much harder than the fisison emission spectrum, with a very substantial tail above 6 MeV. Figure 5.2 also shows that η of the TRU isotopes rises dramatically above 6 MeV. Thus, increasing the energy of the source neutrons from 3 MeV to 10 MeV would increase υ (and η) of the TRU or MA fuel by as much as 33 to 50%.

Figure 5.1. **Comparison of spallation and fission neutron source spectrum**

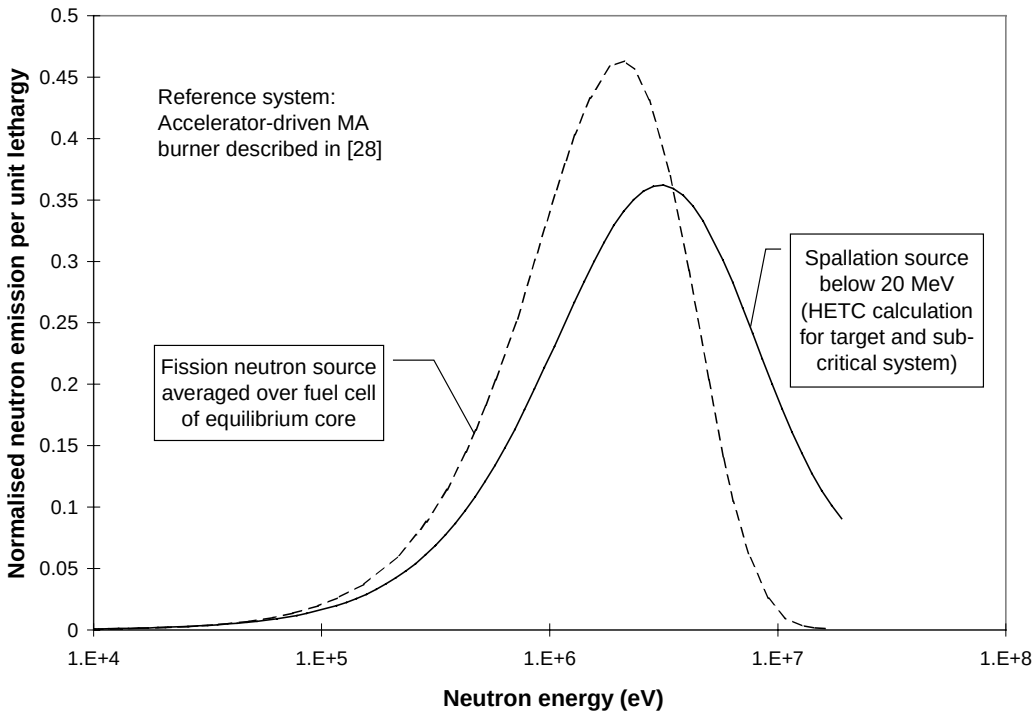
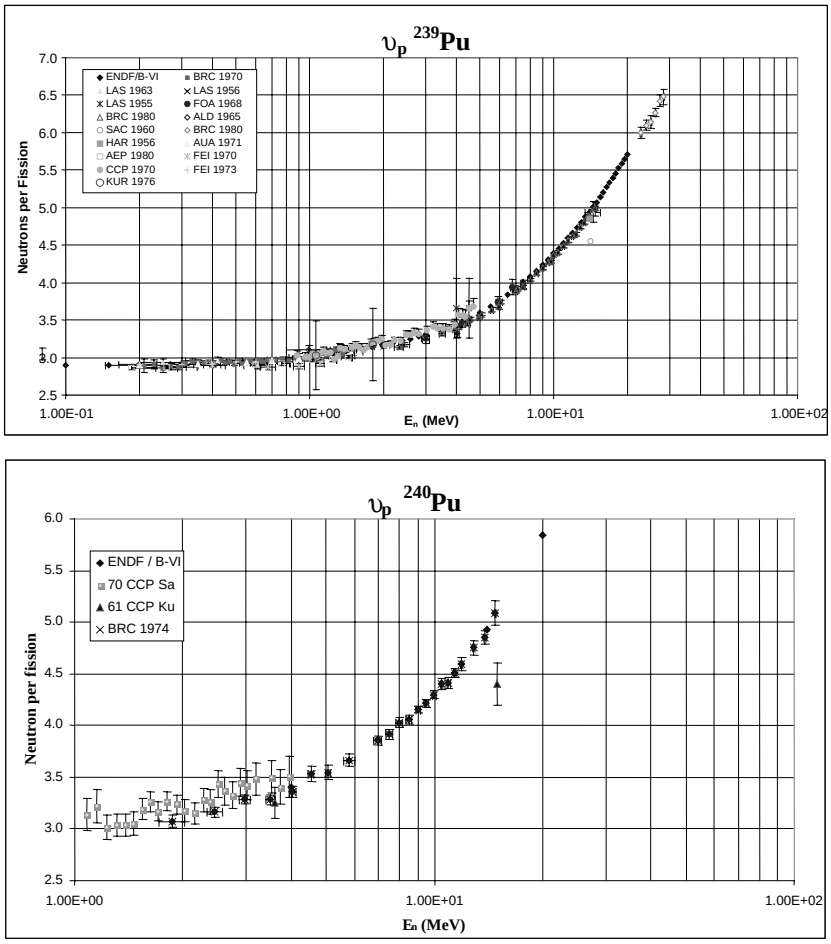


Figure 5.2. **Average prompt neutrons per fission for ^{239}Pu and ^{240}Pu**



Assume for example the first collision of a 10 MeV spallation neutron with TRU and it causes fission. Whereas k for a fission spectrum neutron is 0.98, the k for a ~10 MeV neutron is 50% larger because υ has increased by 50%. All subsequent multiplications in each prompt chain will be sustained by fission neutrons where υ and η return to their fission-spectrum average values, but that first fission with a 10 MeV neutron has already amplified the source by the ratio [υ(10MeV)/υ(2MeV)]. Repeating the derivation which led to Eq. (3) gives:

$$\text{Power} \sim \begin{pmatrix} \text{asymptotic} \\ \text{neutron} \\ \text{population} \end{pmatrix} = S\Lambda \begin{pmatrix} 1 \\ +k' \\ +k'k \\ +k'k^2 \\ +\ . \\ \quad . \\ \quad . \end{pmatrix}$$

$$= \left(S + k'S\left[1 + k + k^2 + ...\right]\right)\Lambda$$

$$= S\Lambda \frac{1+(k'-k)}{1-k}$$

So with k'/k = 1.5 and k = 0.98 we get a 49% increase in source strength. This can compensate for a 49% increase in -ρ, the sub-criticality of the lattice, due to burn-up. Such changes in first flight spallation neutron energy could potentially be achieved by mechanical or pneumatic adjustments of the optical thickness of the buffer around the spallation target – quite sufficient to compensate significant burn-up loss.

Some comments can be made regarding the relative safety advantages of the several options:

- First, increasing the working inventory by reducing k_∞ with an absorbing diluent in the fuel would appear to impair no safety function.

- All the other complementary options for burn-up compensation create a vulnerability to transient over-power because compensating for the reactivity loss upon fuel burn-up, requires either added reactivity or more (or more effective) source neutrons; there is no other choice. Since the option to do this must be designed in, then the risk of doing it prematurely by inadvertence cannot be avoided: each option has a potential to initiate an over-power transient.

- Adjustments to the depth of control rod insertion, location of the spallation target or optical thickness of the spallation neutron buffer are all mechanical in nature; are subject to inherent speed limitations; and (for control rods) are within the experience base of FRs, all attractive features from a safety viewpoint.

- Design efforts to optimise the control strategy for burn-up will have to await some quantification of the relative capital and operational costs of the source, the recycle equipment and the heat removal equipment. However, several observations about cost are obvious without detailed optimisation studies; they pertain to the relative costs of fissile material, neutrons, and protons. The ADS mission is to incinerate fissile TRU or MA which is viewed as a waste; in this mission, fissile material has no commercial value and is cheap. As discussed above, the neutrons exist in excess and must be discarded to parasitic capture; in this mission, they too are cheap. On the other hand the protons are costly, produced in expensive equipment at high operating cost. Finally, as is easily seen by differentiating Eq. 4, to the first order, power scales linearly with changes to source strength or to sub-criticality but with very different factors. Multi-megawatt changes in the proton beam are required to achieve the same power increment that only a few tenths of a percent change in sub-critical reactivity can produce. These observations suggest that a cost effective approach for holding power constant during the ADS fuel burn cycle is to load cheap excess fissile material and discard a variable number of cheap excess neutrons in an adjustable parasitic absorber (control rod) so as to avoid sizing the accelerator for the neutron multiplication level otherwise needed at the end of cycle.

5.3.2 Heat removal in ADS; effects of coolant choice

An important objective is to maintain adequate cooling of all fuel pins throughout their lifetime while avoiding cost penalties involved in over-designed pumps, over-cooled pins and the consequently low coolant outlet temperature with reduced efficiency in energy conversion. This is more challenging for an ADS than for FRs because of inherently greater power peaking in the ADS. Both the source and the beam entry geometries are contributors; source effects are discussed here and those of beam entry geometry in 5.4.2.

5.3.2.1 Power density peaking and its changes with burn-up

In a source-driven sub-critical multiplying lattice, the classical convex fundamental mode neutron flux distribution of a critical reactor is not retained; instead, a concave flux shape peaks at the source and decays with distance from the source prevails. Power-peaking factors are controlled by radially "zoning" the k_∞ distribution of reactivity, whether by fuel shuffling or by grading the radial distribution of fuel volume fraction or of absorber distribution.[45]

A strategy of uniform flow through either FR or ADS cores would require sizing the pumps for the pins of highest power, with consequent pump over-sizing and with substantial over-cooling of most fuel pins. The traditional way to address this issue in FRs of high power density is to use ducted assemblies (i.e. preventing cross flow) with flow metered to individual assemblies by means of orifice plates. This works well when the radial power density profile changes little with burn-up. Such invariance of radial power profile may not be achievable to the same degree in an ADS, depending on the approach taken to compensate for declining reactivity. For example, adjusting source amplitude or the spatial distribution of its effectiveness will necessarily exacerbate the shifts in neutron flux profile by changing the ratio of source to fission multiplied neutrons; it will exacerbate the effects of changes in the radial k_∞ distribution as the fuel preferentially burns out in regions of higher power density.

Assurance of adequate cooling under normal and abnormal conditions is a significant safety issue for ADS and FR alike, and the higher power peaking and its change with fuel burn-up in an ADS will demand careful attention during design optimisation. A general strategy cannot be formulated before the approach to controlling the decline in reactivity is decided. However, an avenue for consideration is to design the ADS with low power density in a loose lattice of ductless assemblies and to assist pumped flow by natural circulation, which tends inherently to distribute itself radially so as to maintain a temperature rise in the coolant that is relatively uniform across the core. This option is available to liquid (but not to gas coolants) and it carries a cost penalty owing to increased vessel size for a given power rating when the power density is decreased.

5.3.2.2 Choice of coolant

Several different coolants are under current consideration for fast neutron ADS designs. Sodium-cooled designs comprise the least extensive departure from the traditional fast reactor experience base. Lead-bismuth eutectic alloy designs are considered, owing especially to the excellent proton-induced neutron spallation properties of lead and the possibility of incorporating a common database of materials properties and design experience for both liquid metal spallation target and reactor coolant. Gas cooling (He or CO_2) of a fast spectrum ADS is also under consideration. Taking a sodium-cooled fast reactor as the reference, the safety-relevant differences to be addressed by coolant choice are discussed below.

One of these is activation of the coolant and of the impurities which are either initially contained in it or dissolved or suspended into it through corrosion and erosion of structural materials. This issue, which is common to FR and ADS, is significant both throughout operational lifetime and later, during D&D and ultimate disposal of the coolant. Neither He nor CO_2 is activated by neutron irradiation. Sodium is activated, producing a 15-hour β-emitter (^{24}Na), but owing to excellent chemical compatibility with structural material, it picks up essentially no activated impurities. Lead-bismuth is activated producing a 138-day α-emitter (^{210}Po) and it tends to pick up scale and dissolved material from chemical interactions with structural materials.

45. Enrichment zoning is precluded by the choice of fertile free fuel.

A second significant safety issue concerns chemical compatibility between coolant and structural material, ease and practicality of controlling structural corrosion, mass transport and sludge formation. The rank order of most to least favourable chemical compatibility at temperatures contemplated for ADS operations (450 to 550°C) is gas, sodium, and lead alloy. Especially in the case of lead alloy, extreme diligence is required to avoid building up of a sludge of corrosion products which could block coolant flow and lead to damage by overheating fuel pins. In the case of lead alloys, an oxide film is maintained on the cladding surface to prelude chemical attack by the coolant. Experience outside Russia is limited on two issues, the increase in impedance to heat transfer due to the oxide film, and the film's robustness during rapid power density changes attendant on beam trips, as will be discussed later in Section 6.3.3. In all cases, in-service inspection of structures with core support or heat transport function will be required; both liquid metal coolants present challenges here.

Liquid metal coolants freeze above ambient temperature and may require trace heating to maintain liquidity in the event of a long shut-down. The issue is of special relevance for lead which freezes at about 325°C, so the flow and temperature of feed water to the heat transfer equipment must be designed and controlled with this high temperature in mind. The high boiling point of lead and lead-bismuth, on the other hand provides more than sufficient margin to boiling.

The coolant voiding coefficient of reactivity owing to the energy dependence of the η of TRU fuel is a significant safety issue in liquid-metal cooled FR systems. Reducing the density of a liquid metal coolant, or creating voids in it, causes two opposing reactivity changes: increased neutron leakage (reducing reactivity) and reduced neutron moderation with a resulting increase in the energy-averaged value of η (increasing reactivity). The rank order of most to least favourable net feedback effect occurs with gas, lead alloy, and sodium; gas has little effect on either leakage or moderation, and lead alloy has less than sodium. Steam entry in a gas-cooled system driving a Rankine cycle through a steam generator is similar in effect to positive void, it adds reactivity. Change of phase is not an issue for gas or Pb-Bi but is for Na. In every case however, an ADS is purposefully designed to reduce the safety significance of reactivity effects as compared with a FR by increasing the offset to prompt criticality and operating below delayed criticality. The coolant void effect will do no more than influence the degree of sub-criticality in an ADS [147].

The high density of lead-bismuth introduces two issues on which little experience exists. First is the structural support and the seismic structural response of large reactor vessels when filled with dense lead alloy. Second is the design of refuelling equipment and fuel assembly hold-down devices for the case where the fuel and the structures are less dense than the coolant and they tend to float in it.

The larger thermal mass of liquid metals compared to gas provides a mitigating feature in loss-of-cooling accidents, providing longer periods of grace for removing decay heat.

A significant safety issue is the consequence of leaks in the primary coolant system. Rank ordering of coolant for ADS and FR favours liquids over gas for this issue because only gas operates at above-ambient pressure. However, each coolant displays a vulnerability which is unique to itself. Since gas cooled systems operate at high pressure, a loss of integrity anywhere in the gas circuit could lead to a loss-of-coolant accident (while thermal neutron spectrum, graphite moderated gas systems with TRISO fuel particles have a huge thermal storage capacity and a very high temperature threshold for damage and can ride this event out, it remains to be shown that the same could be accomplished in a gas cooled ADS design). Loss-of-coolant accidents have extremely low probability in liquid-metal cooled systems in a pool layout (e.g. 10^{-4} to 10^{-6} for the EFR primary vessel leakage). Each liquid metal, however, displays a safety vulnerability upon loss of integrity in the primary boundary. Sodium burns in air, creating an aerosol containing (24 hr β-emitting) radioactive ^{24}Na. Lead-bismuth alloy does not burn but none-the-less releases 138-day (α-emitting) ^{210}Po. Safety approaches have been developed in the fast reactor communities to mitigate and recover from leakage of Na and Pb-Bi and

are more mature than in a gas cooled fast reactor. However, in-service inspection and repair are much more difficult with cooling by opaque liquid metal rather than by gas.

For fast spectrum ADS applications safety-related issues upon loss of primary boundary integrity should be evaluated first at the particular point of vulnerability inherent to ADS: the single thin wall between the transmuter coolant and the evacuated extension of the proton beam tube leading into the spallation target at the centre of the core. That window operates in a hostile environment of proton and neutron damage and it alone lies between the centre of the fissioning lattice and the proton accelerating structures outside the containment building. Melting through the beam tube presents a similar vulnerability to losing containment.

5.3.3 Sub-delayed critical operating state; dynamics

5.3.3.1 Asymptotic response of power to changes in source and sub-critical reactivity changes

The neutron multiplication of an external source in a sub-critical lattice relates asymptotic neutron population to steady-state source strength as shown in Eq. (2).

With an appropriate conversion factor, the asymptotic neutron population is proportional to the total fission rate (Power) or the power density as shown in Eq. (4) and repeated here:

$$P \simeq \frac{\Lambda S}{\frac{1}{k_0} - 1} \equiv \frac{\Lambda S}{-\rho_0} \quad \text{where } \rho \equiv \frac{k-1}{k}$$

The asymptotic fractional change of power or of power density to fractional changes in source strength, δS, or in reactivity, $\delta\rho$, is given, to first order, by differentiation:[46]

$$\frac{\delta P}{P_0} = \frac{\delta S}{S_0} - \frac{\delta\rho}{\rho_0} \tag{5}$$

and, for example:

- 10% source change causes 10% asymptotic power change.
- 10% reactivity change causes 10% asymptotic power change.

Changes in power are directly proportional to those in source or sub-critical reactivity at sufficiently small values, but the baseline values of source and of sub-critical reactivity are very different in magnitude: the source is very large, requiring proton beams in the range of tens of megawatts, whereas the sub-critical reactivity is only a few percent in the neutron reproduction factor, k. Because hundreds of megawatts of fission power are required to incinerate, but accelerator can deliver only tens of megawatts of proton beam, source multiplication of 50 or more are needed and require a sub-criticality, ρ_o, no more than a few percent.[47]

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46. For large changes, ΔS, or $\Delta\rho$, the non linear terms may be important: $\frac{\Delta P}{P_0} = \frac{\Delta S}{S_0} - \frac{\Delta\rho}{\rho_0\left(1+\frac{\Delta_0}{\rho_0}\right)}$

47. Additionally, the rate of generating radiotoxic spallation products, an unavoidable consequence of ADS operations, is minimised by operating near criticality so that spallation products do not outweigh TRU incineration in the net balance of radiotoxicity.

This has profound practical relevance to controllability because when ρ_o is near zero and the source is strongly multiplied, any small changes in contributions by reactivity feedback to the value of k translate into big changes in sub-criticality, ρ, and to big changes in source multiplication. For example, each source neutron is multiplied 50 times if k = 0.98.

$$\rho_0 \approx -0.02$$

In this case, a change Δk of only 0.002 represents a substantial 10% change in ρ_o (and in power when the source remains constant). Although the change in k seems minute, it is the range of thermo-structural feedback.

Thus, in an ADS, even slight changes to thermal or structural reactivity feedback would require multi-megawatt changes in proton beam strength (or in source effectiveness) to compensate for them and hold power level constant. But such thermo-structural reactivity feedbacks will unavoidably occur and will relax over several minutes following every power change, suggesting the consideration of a compensating control rod in the ADS to avoid the need for continual large adjustments of the multi-megawatt proton accelerator.

Reactivity changes due to fuel burn-up will be very much larger than thermo-structural feedback, causing increases in sub-criticality in the region of several hundred percent. However they occur on a longer time scale. Design options accommodating them were discussed in Section 6.3.1.2 but again are profoundly affected by the implications of Eq. (5).

Whatever the strategy chosen for adjusting power level in the face of reactivity changes, the degree of sub-criticality, ρ_o, takes on a safety significance in the ADS because it is the basis for assuring that no plausible reactivity variation can take the chain reaction into the super-prompt-critical range. Thus ADS systems will require a safety-standard means of monitoring the level of sub-criticality to ensure that it never falls below the specified minimal value.

5.3.3.2 Dynamics of power response to changes in source and to changes in reactivity

In a critical FR system, a reactivity change leads to an asymptotic[48] change in power over a period controlled by a time constant, $1/\lambda$, of about ten seconds as determined by the delayed neutron half lives $1/\lambda_i$.

$$1/\lambda = \Lambda_0 + \sum_i \frac{\beta_i}{\beta}\frac{1}{\lambda_i} \simeq 10 \text{ s.}$$

The initial prompt jump step change in power (which takes place on the 10^{-6} second scale of the prompt neutron generation time, Λ), is very small at about 0.003, and is followed by the slower period response which takes off from the post-prompt-jump power level, the rate of which depends on the magnitude of the inserted reactivity. The rate of power adjustment is chosen to match the thermal and structural time constants, which are in the range of 0.1 to 100 seconds (see Figure 6.1).

Sub-critical operating state and dynamics effects

A fundamental distinction between ADS and critical reactor in safety control arises from the dramatic differences in dynamic response between critical and sub-critical source-driven lattices. In a source-driven system, a change in strength or in effectiveness of the source, or a change in reactivity,

48. Feedback effects were discussed in the previous section.

will cause the neutron population and power level to adjust promptly[49] to a new asymptotic level in accordance with Eq. (4). By running the neutron multiplying lattice below delayed criticality and making-up the deficiency in neutrons by supplying them from an external source, an increased margin between the operating state and prompt criticality can be achieved, and this strategy has the beneficial effect of allowing for greater variations in reactivity without entering the range of a prompt-critical abnormal accident where power would increase exponentially with a time constant of microseconds. Moreover, in the sub-prompt critical range of normal operations, it leads to bounded rather than exponential power density responses to reactivity changes. These are desirable effects for the reasons discussed previously. However, the price that is paid is that the microsecond time constant of the prompt neutron dynamic response now prevails even in the normal operating range.

The dynamics and control challenges can be illustrated under the realistic assumption that the neutron population, n(t) is in prompt quasi-static equilibrium with the source.

For a reactor it is the delayed neutron source; for the ADS it is the external spallation source plus the delayed neutron source:

$$\frac{dn}{dt} = 0 = \frac{\rho - \beta}{\Lambda} n + \lambda C + S \tag{6}$$

$$n(t) = \frac{\Lambda}{\beta - \rho(t)} [\lambda C(t) + S(t)]; \; [\text{units}] = \left[\frac{\text{neutrons}}{\text{cm}^3} \times \text{vol. of core} \right]$$

where:

Λ = prompt neutron generation time ~ 10^{-7} s.
$1/\lambda$ = delayed neutron precursor lifetime ~ 10 s.
β = delayed neutron fraction ~0.002.
$\beta-\rho(t)$ = $\beta - \rho_0 - \Delta\rho(t)$.
$\beta-\rho_0$ = reactivity offset from prompt critical $\left[\frac{\Delta k}{k} \right]$.
$\Delta\rho(t)$ = feedback + external control reactivity.

The prompt neutron population establishes equilibrium immediately (~10^{-6} s.) after any change in:

- External source change S(t)
- Delayed neutron precursor source change λ C (t)
- Reactivity change $\rho(t) = -\rho_0 + \Delta\rho(t)$

The responsive rates of the two sources which drive the neutron population are very different: S(t) is fast and can change by 100% in 10^{-7} sec. while the delayed neutron source, λ C (t) is sluggish with a time constant of $1/\lambda$ ~10 sec.

49. The adjustment will occur within several prompt neutron generation times for a sub-criticality of 2 to 3% $\Delta k/k$. Given a generation time of ~10^{-7} s., prompt means adjustment times of no more than a few microseconds for an ADS.

Moreover, the delayed source has a memory of previous history of n(t):

$$\frac{d}{dt}C(t) = 0 = \frac{\beta n(t)}{\Lambda} - \lambda C(t) \tag{7}$$

$$C(t) = \int_{-\infty}^{t} e^{-\lambda(t-\tau)} \frac{\beta}{\Lambda} n(\tau) d\tau$$

Finally, whereas for a critical reactor the delayed neutron source is the only source present, for the ADS the delayed source is but a very small fraction of the total source and it depends on the level of sub-criticality.

From Eqs. (6) and (7):

$$\frac{S_0}{n_0} = \frac{-\rho_0}{\Lambda}; \frac{\lambda C_0}{n_0} = \frac{\beta}{\Lambda} \tag{8}$$

So, for the ADS:

$$\frac{\text{delayed source}}{\text{total source}} = \frac{\lambda C_0}{\lambda C_0 + S_0} = \frac{\beta}{\beta - \rho_0} = \frac{\begin{pmatrix}\text{delayed critical offset}\\ \text{from prompt critical}\end{pmatrix}}{\begin{pmatrix}\text{total offset from}\\ \text{prompt critical}\end{pmatrix}}$$

$$\text{For example, given a } 3\%\ \Delta k/k \text{ sub - critical state and } \beta = .0015, \text{ then} = \frac{1}{21}$$

Because it is only a small fraction of the total source, to changes in which the neutron population adjusts promptly, the delayed source cannot be counted on in the ADS to slow down the dynamic response of the neutron population (and concomitant power density) even though such a reaction is highly desirable because the time constants of heat removal, relaxation of thermal stresses, and relaxation of reactivity feedback all lie in the range of 0.1 to 100 seconds (See Figure 5.1). Since delayed neutrons cannot buffer the differences between the prompt neutron power adjustment time and the slow thermo-structural relaxation times, new control challenges arise for the ADS; specifically, the controller and actuator must themselves perform this function so that the control actuator (whether acting on source strength, source effectiveness, or reactivity) must achieve:

- Very gradual adjustments.
- Very precise changes.
- High reliability.

Moreover, the fuel is where neutronic and heat removal time constants clash continually; giving rise to new requirements on the fuel, as well; specifically it must be structurally resistant to thermal shocks, and must have heat storage capacity to slow down heat release transients.

Controller options include traditional control rod actuators as well as actuators controlling source strength or source effectiveness (either spatial or spectral dependencies). As in compensating for reactivity swing with burn-up, the control actuator will probably be required to have a “nuclear safety grade” level of reliability.

In summary, the dependence on spallation source neutrons rather than on delayed neutrons to maintain the fission chain reaction in balance from one fission chain generation to the next leads to an extremely abrupt response to control actions, reduced influence of reactivity feedback from changes in power or flow-rate, and adds to the importance of the fuel and of the control actuator itself in reconciling the vastly different time constants of nuclear and heat removal processes.

5.3.3.3 Thermo-structural time constants and ADS tolerance to abrupt power density changes

The time constant for the response of power to changes in source or reactivity is evidently very much shorter than that of heat transfer from fuel to coolant (~0.1 to 1.0 s.) so that all incremental heat from a change of source or reactivity is initially deposited in the fuel and remains there for up to a second. As an example, for pin linear heat rates typical of FRs, the temperature difference between coolant and centre line in oxide-fuel is ~1 500°C; accordingly, a 10% source increase leads to a 10% step change in power density and so to a 150°C increase in ΔT across the fuel pin radius, occurring over the several hundred millisecond time constant of thermal diffusivity in the fuel. For metal or nitride, the fuel temperature rise is ~200°C from coolant to fuel centre line so that a 10% source change gives rise to a 20°C increase in ΔT across the fuel pin radius in several hundred milliseconds. To prevent the resulting thermal shocks from reducing the fuel to rubble, the potential ADS design approach is to employ fuel of high thermal conductivity and heat capacity but especially with a high degree of structural toughness.

The time constant of power change is also very much shorter than the transit time for coolant through the lattice and than the time constant for heat transfer from the coolant to the heat sink across heat exchanger tubes. Structural members in the transmuter are of heavy gauge metal for which thermal stress resulting from rapid changes in surface temperature is serious degradation; thermal shocks to structural members having safety related functions in core support or heat removal are clearly to be avoided.

Proton beam trips occur frequently in current linear accelerators [127], and they provide an ADS-specific safety issue in that power density has a time constant much shorter than that of structural thermal response. Upon a proton beam trip, the coolant temperature rise will promptly collapse to zero and all structures downstream from the core outlet will be bathed by coolant which is much cooler than it was just a few seconds previously. Since the time constant for structural temperature equilibration is longer than for temperature change in the coolant, thermal stresses will exist, and if the beam trip events occur frequently, low-cycle fatigue failure of the structures becomes a safety and an operating issue [148,149]. (Because it is a poorer heat transfer medium than liquid metals, gas coolant leads to reduced thermal shocks on downstream structures, but does not eliminate the phenomenon as a safety issue).

Any of numerous design approaches could be taken. First, it is clear that intentional power changes would have to be controlled so as to be gradual. Second, the fuel has to be capable of adiabatic heat storage to buffer any sharp changes, (i.e. to have very large margins to damage temperatures, structural toughness, and high heat capacity). Finally, since temperature changes upon power change are proportional to the nominal temperature rise across the core; de-rating power density to reduce core temperature rise can be considered, at the expense of increase in size and cost of all capital equipment; alternatively, flow could be rapid forced circulation to yield a smaller temperature rise across the core. Either way, abrupt changes in power would yield smaller changes in the outlet temperature of coolant.

5.3.3.4 Options for trim control of power to compensate for thermo-structural reactivity feedback

Thermo-structural mechanisms of reactivity feedback have amplitudes which are individually significant in relation to ADS operating sub-critical set-points in the range of $\rho_0 \cong -0.02$. the following mechanisms adjust to power changes with time constants which vary from a few seconds to several hundred seconds:

- Fuel axial expansion (~5 s.)
- Fuel assembly radial bowing (~5 s.)
- Grid plate radial dilation (~200 s.)
- Core support structure axial expansion (~400 s.)
- Control rod driveline expansion (~60 s.)

Experience in FRs indicates (See Figure 6.3) that these thermo/structural feedback mechanisms each have values in the range of 20 to 50 β (on β of 0.003), i.e. in the range of 0.0006 to 0.0015 $\Delta k/k$. Therefore, following any power change in an ADS, the net level of sub-criticality, ($\rho_0 + \Sigma$ feedbacks) will fluctuate by up to 50% of its value with net positive and negative effects relaxing over several minutes. How should these variations in net sub-criticality be trimmed to hold power constant?

To avoid constantly adjusting the multi megawatt proton beam strength to trim feedback, it would be preferable to hold the proton beam current constant and use a mechanical actuator to trim the reactivity or the source effectiveness to hold power constant as the feedback relaxes; such mechanical adjustments are simple to implement and the technology is within the extensive FR experience base for reliable controllers and actuators. Moreover, from a controllability point of view, since moving absorber actuators (control rods) is naturally slow whereas proton beam changes are naturally fast, it would be preferable to trim control mechanically rather than through beam strength, because, as discussed previously, it is important to increase the time constant of the change in power density to match that of heat removal.

Finally, controlling the power level of an ADS through the accelerator beam current might lead to a "nuclear safety grade" designation for the accelerator equipment and its maintenance, with significantly unfavourable cost implications. Alternately the proton beam could be operated at 100% strength at all times with a safety grade scram circuit, while effects of declining or fluctuating reactivity could be compensated by mechanical actuators of safety grade. Assuming a beginning of cycle $-\delta_o$=3% and a burn-up swing of 6%, a control rod bank worth of 6% $\Delta k/k$ would accomplish the same compensation for burn-up reactivity as a proton accelerator larger by factor of two, probably at a significantly lower cost. Mechanical adjustments of neutron source effectiveness through changes in source location or spectrum may be other options. Even adjustable mixes of various spallation target materials having differing neutron yields per proton might be considered.

In any case, even when controlling with reactivity, adjustments in power density will follow within microseconds and therefore the controller should move the actuators quite gradually, while the fuel should have high thermal storage capability and be mechanically tough.

5.3.4 Spallation neutron source and beam tube effects

The spallation target and associated proton beam tube introduce vulnerabilities to added reactivity and increased source strength unique to an ADS. The most obvious is the potential for positive reactivity insertion upon flooding the beam tube and shutting off the streaming leakage path for

neutrons out of the core. The second is the potential to increase inadvertently the strength or the effectiveness of the source neutrons in a way that increases power density. Penetration of containment is a third issue and will be discussed later in Section 5.4.

5.3.4.1 Beam tube flooding

Should the beam tube wall or window loose integrity, then in the cases of Na or Pb-Bi cooling, the coolant would immediately flood the evacuated beam tube to a level at least the height of the coolant free surface. Although the spallation source would move to the upper regions of the flooded column and its neutronic coupling to the transmuter core (i.e. its effectiveness) would drop to nearly zero, an increase in reactivity would result from extra neutron reflection due to filling the tube with coolant which acts as a reflector more than an absorber. It will be essential to design the ADS so that this event would not take the core into prompt criticality. Scoping calculations for a 840 MWth Pb-Bi cooled system with a 40-cm diameter beam tube indicate that reactivity additions in the range of 0.5% $\Delta k/k$ (i.e. several dollars) are possible.

The beam tube flooding scenario will be one among many of the considerations for setting the sub-criticality level on the transmuter fissile loading.

5.3.4.2 Source importance changes; buffer voiding event

The "effectiveness" of the neutrons which are injected into the sub-critical transmuter lattice depends not only on properties of the transmuter lattice core itself but also on the spatial position and the energy spectrum of the source neutrons. Current ADS design concepts place the spallation target near the centre (axially and radially) of the transmuter lattice where its is most effective. When central placement of the target is the design approach, abnormal events which move the source location off centre (e.g. the beam tube flooding event described above) will reduce the effectiveness of source neutrons, and without changes in reactivity the neutron population and associated ADS power level will tend to decrease.

In addressing the energy dependence of S*(R,E) for source neutrons, it is noted that after the first flight leading to a fission event, multiplication in the subsequent prompt fission chain will be determined by the familiar formula for sub-critical source multiplication shown in Eq. (3), which depends on the properties of the transmuter core geometry and composition ß and particularly on the η value of the transuranic fuel averaged over the fission neutron emission energy spectrum as slowed down by the lattice materials. This fission-multiplied neutron energy spectrum never exceeds the top end of the fission emission spectrum at a few MeV. The fission power is directly proportional to this fission-multiplied neutron source strength as shown in Eq. (4).

However, the spectrum of spallation neutrons has a tail in its distribution which goes well beyond 6 MeV as shown in Figure 5.2. Therefore, spallation target designs incorporate a row of "buffer" assemblies around the target, filled with coolant and other scattering materials to moderate the energy distribution of the neutrons down to the MeV range and to spread their directions of emission so as to produce a more nearly isotropic source. Should this moderator material be somehow removed, the most energetic spallation neutrons would pass through the buffer on their first flight with little or no scattering moderation and undergo their first fission interaction at high neutron energy (e.g. > 6 MeV), with a higher value of η than intended and releasing substantially more fission neutrons. All subsequent events in the fission chain would continue as before. Thus a vulnerability exists in that an abnormal loss of moderation in the buffer would significantly raise the power to flow ratio at a fixed value of sub-criticality.

Since an abnormal buffer voiding event will constitute a vulnerability in any case, this mechanism can perhaps be put to good use and included among the options considered in Section 6.3.1.2 for deliberate control of the transmuter power level in the face of declining reactivity with burn-up.

5.4 Containment, shielding, and decay heat removal

The traditional FR design approach for assuring the containment of radioactivity is based on defence in depth with three containment boundaries: the fuel cladding, the primary coolant system boundary, and the containment structure. Each barrier has provisions for cooling so as to maintain its integrity in both normal and abnormal conditions. The outermost barrier, the containment structure itself, must be designed to ensure containment when the middle barrier is vulnerable during refuelling operations, and it must provide a highly reliable channel for transmitting decay heat to an exterior ultimate heat sink without loss of containment even under severe accident conditions. The strength requirements of the containment derive from considering hypothetical core disruption events and the amount of internal heat, pressure, and missiles that they could conceivably generate. While many containment issues are similar for FR and ADS, several are peculiar to features of the ADS.

5.4.1 Proton beam tube penetration of containment barriers

The presence of a spallation target at the centre of the transmuter core and the arrangements made to direct a high energy proton beam on to it are features totally absent from a fast reactor. Several safety issues related to the containment function, but also pertaining to refuelling, shielding and coolability, are discussed here.

An obvious issue raised by the beam tube of an ADS is that of maintaining multiple containment barriers. For linac-driven ADS, the proton beam tube penetrates the containment-building barrier. (Cyclotron-driven ADS could conceivably place the cyclotron inside the containment structure and avoid penetrating the outermost of the three containment barriers). In both cases, the proton beam tube itself (and the proton window if one is employed) comprise a re-entrant segment of the primary coolant boundary barrier.

The beam tube penetration of the containment boundary is similar in character to the secondary coolant loop penetration of a standard sodium cooled fast reactor or the steam line penetration of a thermal reactor, where safety-grade closure valves can be provided. The multi-megawatt proton beam itself, however, comprises an ADS-specific hazard to the integrity of the tube and the fast acting valve alike, because beam misalignment would promptly melt through the tube wall and if not immediately tripped the beam would melt the fast acting valve.[50]

The beam tube as a re-entrant segment of the primary coolant boundary is topologically similar to an intermediate heat exchanger tube in a FR. In the latter case, the tube contains secondary coolant at ambient pressure whereas in the former it is under vacuum.[51] Again the beam misalignment hazard to proton beam tube integrity and the particularly hostile environment experienced by the beam window are unique to ADS design. Ruptures of either window or tube wall will open up a connection into vacuum so that flows of atmosphere will initially be inward into the accelerator cavities; moreover, loss of vacuum will itself trip the beam as a response to sparking in the HV accelerator cavity. Longer-term containment must be addressed.

50. Upon loss of vacuum, sparking in the acceleration cavity would normally lead to accelerator trip.
51. In the BREST reactor concept an integral steam generator replaces the intermediate heat exchanger, and the tube walls separate high-pressure steam from ambient pressure coolant.

For pressurised gas-cooled ADS, loss of integrity in the beam tube or window represents one of the ubiquitous potential pathways to loss of coolant.

An ADS-specific challenge to the primary coolant boundary barrier may derive from the position of the bending magnet. If, on the one hand, a top-entry beam tube is employed, then the multi-ton bending magnet is placed directly above the transmuter vessel, where it represents a falling hazard in the event of structural flaws or damaging seismic accelerations. If, on the other hand, the beam penetrates the transmuter vessel from the side so as to eliminate this hazard, and penetrates at an elevation below the surface of the liquid metal coolant, then the beam tube provides a vulnerability for coolant draining on failure of nozzle weld or window. With gas cooling, vulnerability to loss of coolant is independent of entry arrangement.

5.4.2 Refuelling and shielding

For the most part, the maintenance of sub-criticality, of containment, and of decay heat removal during refuelling operations present issues common to ADS and FR. The presence of the proton beam tube, however, presents several challenges which are unique to the ADS.

The most obvious is the issue of shielding. Because an unobstructed flight path is essential to deliver the proton beam on target, the opportunity for shielding inside the beam tube is foreclosed. The beam tube comprises a radiation streaming path of significant cross sectional area for gammas and neutrons from the centre of the transmuter core to the exterior of the primary coolant vessel, or even beyond the containment building and into the proton acceleration structures. If the beam enters from the top, the beam tube gives rise also to a straight neutron streaming path from the core centre to the region of the bending magnets, causing their activation by neutron bombardment. If the beam enters from the side, the streaming path extends further into the accelerator segments themselves with the potential for activating them. Bending magnets could provide a labyrinth path, but would be subject to activation of the magnet itself so that shielding and appropriate maintenance procedures become necessary. Both activation of structures outside the vessel and direct radiation streaming present challenges to keep worker dose exposures low during operations and maintenance.

If the beam enters from the top, then bending magnets and their shielding are located directly above the core, and difficulties of access may arise in refuelling, notably through interference with the polar crane and constraints on height. Nonetheless, if top entry beam geometry could accommodate refuelling, it would be beneficial in that the fuel-loading pattern would be azimuthally symmetric, as is not possible with other beam entry orientations.

If the beam enters from the side, interference with refuelling will be avoided, but the fuel-loading pattern will necessarily be azimuthally asymmetrical to accommodate the beam tube. “Teapot” configurations have been considered for conceptual ADS layouts; the beam enters the transmuter vessel at an angle from above, down the spout, and thereby avoids the loss-of-coolant vulnerability, interference with refuelling, and the vulnerability to impact on the magnet. Like the side entry configuration, the teapot approach introduces an asymmetry in core layout and refuelling, giving rise to radial and azimuthal distortions of power density.

The safety relevance of distortions to power density arising from the side and the teapot beam entry configurations is not known without analysis, but will pertain to ensuring cooling, and to accuracy in predicting fissile burn-up. These could affect certain approaches to guaranteeing the specified degree of sub-criticality and in any case will increase uncertainties in the margins from criticality in recycling facilities assumed in sizing process equipment and to control recycle operations.

5.4.3 Decay heat removal

A heat transport pathway sized at 0.1 to 1.0% of rated power must be maintained to the ultimate ambient heat sink from the fuel pins, and must be guaranteed to function under conditions following a severe accident. Enough thermal storage capacity must be provided to absorb the temporary initial excess of decay heat over the capacity of the heat removal channel.

Traditionally in FR plants, the heat transport path to the balance of plant (BOP) heat engine equipment carries a requirement for safety-grade removal of decay heat. Recent fast reactor designs have relied instead on dedicated redundant heat transport circuits from the reactor vessel pool to the air outside the containment structure and often configure them to operate continuously and "passively" on the basis of buoyancy-driven flows. The "pool" category of liquid metal cooled FRs utilises double walled vessels to assure that primary coolant is confined to the pool even if the primary vessel leaks, and the coolant inventory is maintained by this second "guard vessel" to cover the core and the heat exchange surfaces dedicated to removing decay heat (DRACS). With thermal ratings of 1 500 MWth the outer of the two vessels (the guard vessel) is sometimes declared the (close-coupled) containment structure and its outer surface is itself cooled by a natural draught of ambient air (RVACS) as the final link in the decay heat removal channel; (some licensing authority regulations do not accept this close coupled containment).

For liquid metal cooled ADS with top or teapot entry beam tube, a similar approach should apply with no additional issues. For gas cooled ADS or liquid-cooled ADS with a side entry beam tube, the issue of loss-of-coolant accidents would have to be addressed as for LWRs and HTGRs.

Whatever the design for decay heat removal (RVACS, DRACS, or BOP), safety-related testability will be required for ADS as for FR.

5.4.4 Containment loading criteria; HCDA termination

The size (internal volume) and design pressure rating of containment structures for LWRs and FRs have historically been determined by the loadings they must sustain in their role as the final barrier to reactivity release, even in the event of a severe accident which leaves the fuel cladding and primary coolant boundary in tatters. For water-cooled thermal reactors, the determining event is the loss of coolant accident and large hydrogen deflagration (from Zircaloy-water reaction at high temperature); for fast reactors it is the Hypothetical Core Disruptive Accident (HCDA).

Fast neutron lattices of all kinds are not in their most reactive configurations; changes which decrease surface to volume ratio will reduce neutron leakage and increase reactivity, conceivably to reach super-prompt criticality. This unavoidable vulnerability has led to decades of work on severe accident evaluations to determine bounds on energy release resulting from hypothetical core disruption, so as to provide the information for sizing and design of the containment building to contain that release and release rate. Early small prototype sized metal fuelled FRs relied on fuel dispersal to quench the postulated prompt critical burst while later, the larger commercial-sized oxide fuelled FR relied on prompt-acting negative feedback from Doppler absorption in fertile material contained in the fuel to reduce energy release from the burst. The recent modular-sized ALMR relied on melting of the metal fuel and its immediate dispersal by fission gases to preclude a prompt critical burst. In all cases the goal was to quench the chain reaction quickly and thereby limit the energy released, to put lesser demands on the containment structure.

The absence of a fertile Doppler feedback combined with potentially low neutron leakage in an ADS core built of a tough fuel capable of high energy storage will quite obviously necessitate a

changed strategy for terminating a severe accident. If, in response to an abnormal initiator, a large ADS composed of fertile-free fuel were to reach prompt criticality, it would then present an exceptionally severe challenge to containment because it lacks both of the historically employed mechanisms to quench a prompt burst in a fast reactor, i.e. it has neither the Doppler feedback in large cores of robust oxide fuel nor the prompt dispersal achievable in small cores of low-melting fuels [150,151].

Both the lead-cooled and gas-cooled versions of FR and ADS have high volume fractions of coolant in the array with greater vulnerabilities than in sodium-cooled FRs. With the geometry for gas cooling, the disrupted fuel pins would block the neutron streaming paths provided by the coolant channels, and thereby add reactivity.[52] With lead cooling, the high density of the coolant itself would offer inertial resistance to spatial disassembly of the lattice thereby requiring a larger deposition of energy for ultimate disassembly and quenching.

It will be important for the ADS safety strategy to devise means of precluding prompt bursts in HCDA using intrinsic properties of the lattice, as was possible for the modular ALMR. An extra degree of freedom is available in the ADS design to cope with HCDA initiators; it is the initial degree of sub-criticality. If it can be made large enough to overcome the reactivity addition of any plausible compaction or coolant voiding, then the potential for super-prompt criticality can perhaps be foreclosed by design. The need to do so provides one of the strongest incentives for designs which consume the excess neutrons generated in a pure transuranic fuel by using internal neutron absorbers integral with the fissile within the fuel pin, rather than by relying on neutron leakage.

Upon fuel pin disruption – even in the absence of prompt bursts – the issue of re-criticality in the fuel debris must be addressed. The situation might turn out to favour the lead-cooled option where the fuel would float and possibly disperse radially as dross on the lead surface. For the gas and sodium option, the traditional FR re-criticality issues will apply.

5.4.5 ATWS initiators; passive versus engineered safety approach

In design and safety considerations for a FR, transients with scram constitute part of the design basis while Anticipated Transients Without Scram (ATWS) are often considered as Beyond Design Basis and used for sizing the containment structure and its pressure rating. Rod run-out (transient over-power) without scram (TOPWS), loss of heat sink without scram (LOHSWS), and loss of pumping action (flow) without scram (LOFWS) are considered whereas loss of coolant is not considered credible for double-vessel pool designs with liquid metal cooling.[53]

For FRs, the term “scram” refers to inserting the bank of safety rods, always with a single rod assumed to be stuck. Depending on specific licensing requirements, simultaneous tripping of primary and secondary pumps may also be assumed. For ADS the term “scram” has not yet been defined, but must certainly mean that at least the proton beam is turned off.

In some FR designs, completely passive accommodation without damage of LOFWS and LOHSWS events has been designed in and even demonstrated in pilot-scale plants such as RAPSODIE and EBR-II [138]. This FR passive safety approach has relied on thermo-structural

52. Particle-fuel gas-cooled fast reactors require careful scrutiny to identify their potential strengths and vulnerability.
53. The leakage through the double vessel has been considered for Superphenix to represent the “Ultimate (BDB) for which public evacuation procedures must be defined. US licensing procedures for CRBR used the HCDA resulting from a transient over-power driven by loss of flow (positive sodium void worth) as the basis for public evacuation procedures.

reactivity feedback to self-regulate heat production to match the available heat removal rate [150,151]. And in the case of TOPWS, a favourable passive safety response can be demonstrated for FRs designed to have near-zero reactivity loss upon burn-up, so that very little excess reactivity is vested in the control rods.

The favourable performance of ADS in reactivity insertion (TOPWS) events has been well documented for reactivity additions which do not take the system super prompt critical and where a beam trip occurs in time to avoid fuel damage [145,146]. Loss-of-heat-removal events in an ADS lead quickly to overheating if the beam remains on as has also been well documented [145,146]. Even with the beam off, it is useful to suppress multiplication of delayed fission neutrons by reducing reactivity when heat removal has failed.

In keeping with the trend in FRs to place increased emphasis on passive means to reinforce active engineered safety systems, several passive safety features affecting cooling rates and source strength are currently being considered for ADS:

- Natural convection at full or significant power levels (to accommodate LOFWS) [146].
- Passive beam interrupts or relocation to a position of lower effectiveness upon overheating of coolant (to accommodate LOHSWS) [146].
- Electricity to drive the accelerator derived not from the grid, but instead fed back from the ADS itself (to accommodate LOHSWS, LOFWS) [151].

Passive power self-regulation based on thermo-structural reactivity feedback, as has been exploited for fast reactors, is precluded by the fundamental characteristic of sub-critical source-driven systems. For an ADS, the operating point is offset from prompt criticality by $(\beta - \rho_o)$ where $-\rho_o$ is the sub-critical operating point. The offset is only β for a critical reactor. As is evident from the denominator in Eq. (6), the effect is that the power level in an ADS is less sensitive than in a reactor to reactivity feedback. Moreover, as is also evident from the inhomogeneous source term in Eqs. (6) and (7), the power can never be driven to zero by reactivity changes as long as the spallation source is non-zero. The ADS must therefore adopt different strategies for employing passive concepts to keep heat production and removal in balance. Specifically, it needs some means for passively adjusting the strength or effectiveness of the source in response to power changes. Options include powering the accelerator with ADS-generated electricity[54] [152], or source-transmuter coupling dependent on coolant temperature or density. Absorber or moderator curtains in the buffer surrounding the source, or spatial relocation of the target (all activated by temperature or density changes in the coolant) affect coupling and might offer opportunities to apply passive source feedback analogous to the passive reactivity feedback successfully exploited and demonstrated in fast reactors as the passive means to self-regulate the rate of heat production to match removal.

Among research efforts on safety in ADS, applications of passive safety approaches to accommodating ATWS should be stressed, because the efforts for the past decade on FRs have shown significant potential for benefits.

5.4.6 Activation products

In both FRs and ADSs, the activation of materials of construction affects safety, both by exposure to workers during operations, maintenance and decommissioning, and later as a long-term toxicity hazard attendant on waste disposition from the decommissioned plant. Since the function of the class

54. The exceedingly long time constant for feedback presents a major challenge with this option.

of ADS discussed here is to reduce the long-term radiotoxic legacy of nuclear energy, it is especially important that they should not add to it.

The issues of coolant activation and its effect on operational safety issues are similar for ADS and FR and were discussed in Section 5.3.2.2. Activation effects on long term waste management were studied by Oussanov *et al*. [153], who found considerable differences in the character of long-term residual activity arising from sodium, lead, and lead-bismuth coolants. For Na, 50 to 80 years of storage should be sufficient to allow unrestricted further use, for lead perhaps 1 000 years, and re-use is not feasible for Pb-Bi.

Spallation products and activated proton accelerator structures and beam tubes in an ADS are features not shared by a FR. The production rate of radiotoxic species depends directly of course on source strength, providing an incentive for small levels of sub-criticality. The study of long-lived toxicity generated in spallation reactions is at an early stage [154,155]. The mass spectrum of spallation products spans the range from tritium up to the mass number of the target material, and the relative yields depend on the energy of the incident proton beam. Notably, long-lived alpha-emitting rare earth spallation products (e.g. Gd, Sm, and Dy isotopes) could be avoided through use of a target of mass number less than 145 amu – e.g. tin. Preliminary model studies on yields of alpha-emitting rare earths in heavier targets suggest [156] their significance relative to polonium (in Pb-Bi targets) and generally vis-à-vis toxicity reductions obtained by transmuting technetium and iodine.

Beam loss is one of the crucial design factors in the high-current accelerators required for ADS; it also activates accelerator structures, affecting both operations and ultimate disposition of equipment. Detailed calculations were made [157] for the 100 MeV to 1 GeV section of a normal-conducting linac assuming a loss of 0.48 nano A/m (i.e. 1.2×10^{-8} proton/meter) in a 40 mA machine based on the TRISPAL design. Ordinary concrete shielding of between 1 to 4 metres would be required to limit surface dose rate to 1 mrem/h. Misalignment of the beam (40 mA) into the structures for 50 μ s. was also studied and found to produce activation which has largely decayed away after about 15 min. Like the spallation product issue, structural activation in high-current proton beams is at an early stage of investigation.

5.4.7 Propagation of local faults

Issues of local faults (such as breached fuel clad or plugging of coolant channels) which could be propagated and exacerbated into full core events are common to FR and ADS. They have been extensively studied for sodium cooling with oxide and metal alloy fuels where it is shown that chemical interactions between coolant and fuel should preferably avoid forming low-density products. Also, chemistry control of Pb-Bi alloy coolant to avoid both cladding attack and sludge formation has been thoroughly studied in Russian military experience, but scant experience exists outside Russia. It is clear that extensive, multi-year in-pile irradiation testing campaigns will be required for any new combination of fuel, cladding and coolant, as in every one of the inert matrix (non-fertile) fuels considered for TRU/MA ADS burners.

5.5 Safety in fuel cycle facilities

Complete consumption of the transuranic feedstock requires multiple recycling because the neutron fluence required to fission all the transuranic atoms exceeds the neutron damage endurance of the cladding material. Three of the five cardinal safety functions (containment, shielding, and removing decay heat) are identical whether the fuel is in core, out of core in transfer casks, or out of core in a recycle facility; thus the discussions of such issues already given for the transmuter core carry over to the fuel cycle facility as well. However, in the recycling facilities the cladding is

purposely removed. Instead of matching heat production and removal, the cardinal safety requirement out of the core becomes simply “avoid criticality”.

The safety issues related to containment, shielding, and decay heat removal during fuel transfer and recycle operations are essentially identical for the ADS and FR fuel cycles, but the use of fertile-free fuel in the ADS or FR cycle will affect the functional requirement for avoiding criticality because the fast-spectrum critical mass of pure TRU is small. Pure transuranic fuel, with an η of ~1.8 and a fast-spectrum critical mass of 5 to 15 kg, will have to be handled in small batches. Particular care will have to be taken in accounting for effects of reflection and inventory coupling when designing process equipment and deciding on layout. Similarly, moderating materials will have to be excluded or carefully controlled within the casks and the recycle facility.

Criticality constraints within recycle facilities add still further weight to the preference for use of parasitic absorber material, intimately blended with the fertile-free transuranic fuel itself and chemically similar to a rare earth or actinide so as to follow the transuranics through every stage of recycling and refabrication.

Whether in ADS or FR designed for incineration, shielded remote operations are required because the transuranic elements include strong neutron emitters (e.g. ^{244}Cm), spontaneous fissioning isotopes (e.g. ^{240}Pu), pyrophoric chemical characteristics, and low-temperature volatility (e.g. ^{241}Am). These materials must be handled in remotely operated and remotely maintained shielded facilities under inert atmospheres. Their small critical mass demands that process control and material inventory tracking meet high standards of accuracy and that operations be conducted under strict discipline. Atmosphere control of the hot cells (maintaining inert atmospheres to address pyrophoricity) and discharging aerial effluent only through filtered channels places special requirements on seismic design of structures and equipment. It also makes conflicting demands to maintain effluent filtering during abnormal events (such as a breach of cell containment) while stopping flows in order to smother fires following the access of air to pyrophoric materials. Such issues are peculiar to the presence of TRU or MA fuel types and are common to fuel cycle facilities for FR and ADS alike.

5.6 Conclusions

This chapter analysed the safety-related challenges of a specific class of ADSs employing a fast neutron spectrum and solid, fertile-free fuel with the primary mission of transmuting transuranics or minor actinides.[55] Multiple options for addressing nearly all relevant issues have been developed in the framework of an impact matrix of safety functions required for each distinctive design feature of an ADS, and each distinctive design feature has been tracked back to a specified mission element.

From this analysis, the following conclusions can be drawn:

- The ADS’s dynamic response to changes in reactivity or neutron source strength is the area of greatest difference in safety characteristics between fast reactors and ADSs and an area where no precedents exist in the fast reactor experience base.

- The primary cause for this is (a) the external neutron source which can provoke rapid and, depending on the sub-criticality level, large neutron-kinetic responses, unmitigated by the delayed neutrons, and (b) the fertile-free fuel which features very weak reactivity feedbacks, especially from the Doppler effect.

55. A summary of this chapter has been reported in [158].

- This puts high demands on the control actuators, the fuel behaviour, and the heat removal processes. In particular, the strong dissimilarity of the neutron-kinetic and thermo-structural time constants requires the fuel to be capable of sufficient adiabatic heat storage.
- The weak Doppler effect exhibited by fertile-free fuel affects the energetics of hypothetical core disruptive accidents. If such accidents have to be taken into account in the safety analysis of an ADS, a prompt quenching mechanism relying on a phenomenology other than the traditional Doppler effect will have to be developed.
- The management of the surplus neutrons in sub-critical cores with fertile-free fuel by means of neutron leakage and/or absorption involves delicate trade-offs which affect core design. This applies particularly to TRU burners which feature a high burn-up reactivity loss.
- Regarding passive safety principles, it appears that means for passive decay heat removal are already available, but innovation is needed to achieve passive self-regulation of power.

6. COST ANALYSIS OF P&T

6.1 Introduction

Economics represents an important pillar of sustainable energy development, as was mentioned in Chapter 1. This chapter, therefore, aims at addressing the economics of the range of P&T options examined in this study. This economic assessment is presented only in terms of "top-level" cost trends, and is not supported by the kinds of detail required of a rigorous market survey. These limitations result from the large uncertainties in the technologies to be deployed and the associated cost uncertainties.

As suggested above, the cost analysis of advanced nuclear systems and fuel cycles calls for prudence, since the cost assessment for many elements (e.g. TRU/MA-fuel fabrication and reprocessing) must be based on preliminary conceptual studies where little or no (pre-)industrial experience is yet available. The present study addresses the cost analysis by first defining and evaluating unit costs (e.g. $/kg, $/We, $/kg.y, etc.) for the different interconnected fuel cycle steps (see Section 6.2.2). These costs are then aggregated according a standard levelised costing methodology for the nuclear fuel cycle (NFC) (see Section 6.2.1 and [159,160]) and finally expressed as cost of energy, COE (mill/kWh), for each fuel cycle scheme. Because of the above-noted uncertainties, only energy costs relative to the reference LWR once-through fuel cycle (fuel cycle scheme 1) are presented. The intention of presenting relative energy cost is to emphasise relative differences between the various P&T approaches examined, rather than to present or attempt to make actual "market comparisons and assessments" in the form of absolute costs of energy.

The overarching goal of this cost study, i.e. to identify ways in which specific P&T approaches might be improved to become economically competitive under sustainable conditions, points to a "top-level" (highly aggregated) analysis using a range of unit costs (e.g. low, nominal, high) for the advanced fuel cycle options and operational-performance assumptions. While this methodology has known limitations [160-162], the Expert Group determined that it is the most appropriate for this generic cost analysis. However, it is clear that a definite priority or choice ranking of the schemes cannot be a principal goal of this kind (level) of cost analysis.

A "top-level" mass/energy-balance model[56] for the nuclear fuel cycle schemes is used to generate the material flows and inventories required to estimate annual charges for the cost-of-electricity (COE) evaluation. This model is also "top-level" in that, as in the neutronic analyses of the fuel-cycle schemes described in Chapter 3, an equilibrium steady-state is assumed as a simplifying assumption wherein steady-state mass flows and costs are expressed on a per-TWhe basis. The cost-base systems model performed an independent mass and energy balance and was adjusted to produce mass flows in rough agreement with the detailed neutronic analyses reported in Chapter 3. These mass flows (and inventories) and energy balances were then used in estimating annual charges, AC(M$/y), which were then expressed in COE(mill/kWh) units before final normalisation to the COE estimated for the once-

56. The reader is referred to references [163-166] for more details on the calculational methodology.

through LWR scenario (fuel cycle scheme 1). Specific input items for these calculations are, in addition to the input and discharged fuel compositions as evaluated in the neutronic analyses, the unit-cost database that was developed and reviewed by the Expert Group. While such a combined economics and mass/energy balance model is suited to evaluate the connectivity between economics and most other elements of sustainable nuclear energy, the focus in this study is primarily on COE-versus-environment trade-offs as driven by the combinations of technologies used to define the six principal fuel cycle schemes elaborated in Chapter 3. For the purpose of this combined analysis, the "environmental friendliness" is measured in terms of the TRU losses to the repository, to be denoted "LOSS parameter", which for TRU or its plutonium and MA components assumes the unit of kgTRU/TWhe. Other mass flows, for example the use of natural uranium and fuel fabrication requirements, were also calculated to derive the fuel cycle costs but are not reported in this chapter and are referred to in Chapter 3. Extension of this cost-base systems model has been foreseen to include additional metrics to evaluate nuclear energy systems.[57]

The present chapter first describes briefly the NFC systems model, and then elaborates and justifies the associated unit cost estimating relationships (CERs), that are based on a literature survey and iterative judgement by the Expert Group. The second part of the chapter will discuss the results of the levelised NFC costing model, with the presentation of key indicative cost trends and trade-offs concluding this part of the study. Although many of the choices for unit costs and related financial parameters are subject to uncertainty and compromise, an important goal of this chapter is to lay out in as much detail and clarity as is possible the basis for the unit-cost or CER choices made, to the extent needed to support recommendations for ways in which a given fuel cycle scheme may be developed along desirable economic lines, as well as for setting the course of respective future R&D for all of the P&T schemes that form the study agenda.

6.2 Nuclear fuel cycle model

The systems approach, methodology, and unit-cost database (CDB) leading to the costing results reported herein are described in this section. The basis and level of the NFC costing analysis is shown in Figures 6.1 and 6.2. The six principal fuel cycle schemes,[58] as introduced in Chapter 3, are represented in Figure 6.1, illustrating the essential elements of a generic fuel cycle as considered in this study. Figure 6.2 gives a breakdown and definition of key fuel cycle steps or "centres" where material flows and inventories have been tracked; each of the fuel cycle steps depicted in Figure 6.2 also served as a cost centre for conducting the respective economic assessment. Each of the twelve fuel cycle steps of the NFC described graphically in Figure 6.2 represents the finest resolution of the costing and TRU waste-disposal assessments that lead to the respective sustainability parameters. As mentioned before, these fuel-cycle schemes and, therefore, fuel cycle steps, are considered to be in steady-state conditions.

The next Section 6.2.1 focuses on the methods by which the equilibrium (steady-state) NFCs mass flows are estimated, as well as the procedure used to evaluate the COE (mill/kWh) and the LOSS (kgTRU/TWhe) parameters. Subsequently, Section 6.2.2 will describe the unit cost database.

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57. In a separate, independent study [167] based on this cost-base systems model and using the NEA cost base, a third metric intended to give a measure of proliferation propensity for a given NFC is also included. An approximate Multi-Criteria Analysis (MCA) is performed across all scenarios base on this quartet (cost, loss, resource, and proliferation) of metrics. Such an analyses, however, was not either within the charter or resources of the present study or Expert Group.

58. This generic fuel-cycle model also enabled the fuel-cycle scheme 3c to be modelled as a variant of the 3b scheme.

Figure 6.1. **Generic nuclear fuel cycle model showing the essential elements and options for a nuclear fuel cycle**

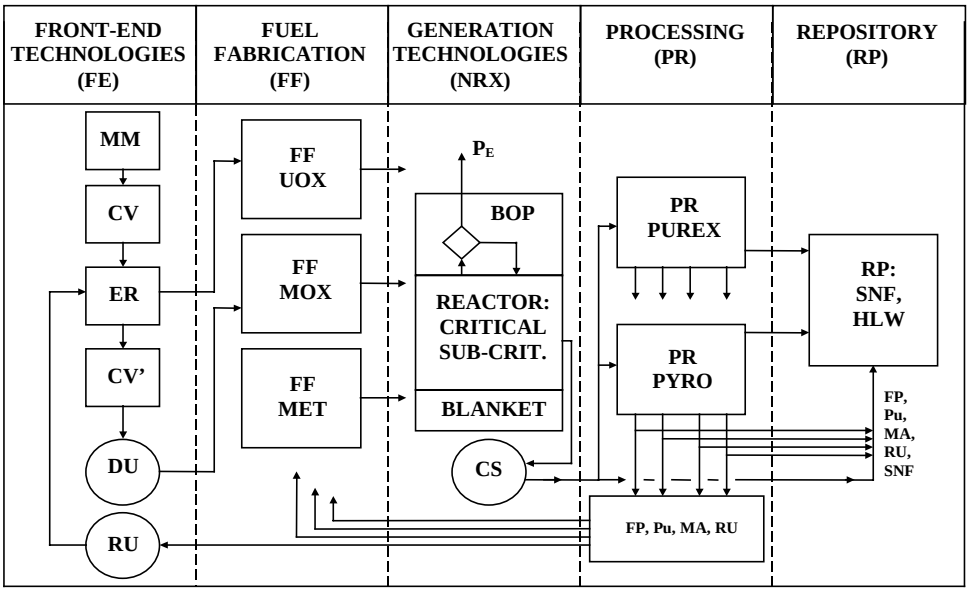
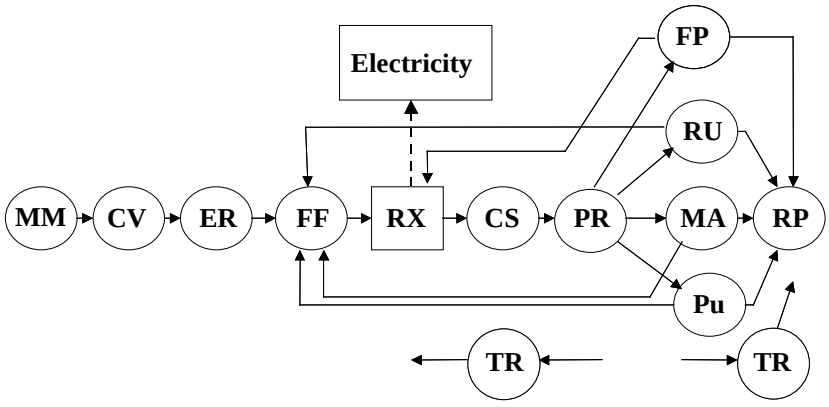


Figure 6.2. **Essential elements of the nuclear fuel cycle (NFC) being considered in evaluating key sustainability metrics for nuclear energy**



MM	**= Minig and Milling**	**RU**	**= Irradiated Uranium**
ER	**= Uranium Enrichment**	**MA**	**= Minor Actinide**
CV	**= Uranium Conversion**	**Pu**	**= Plutonium**
FF	**= Fuel Fabrication**	**TR**	**= Transport**
RX	**= Reactor**	**RP**	**= Repository**
CS	**= Cooling**	**FP**	**= Fission Products**
PR	**= Processing**		

6.2.1 Equilibrium NFC analysis

The simplified, steady-state material-energy balances in the "top-level" model are defined by the assumption of a standard net-electric grid capacity (P_E = 1 000 MWe) for each once-through LWR technology that, with the exception of fuel cycle scheme 5, drives the subsequent technologies in that

fuel cycle scheme. The subsequent technologies that operate in equilibrium with the LWR driver technology are sized to consume the plutonium or TRU generated therefrom. The energy generated by these subsequent reactor technologies in the fuel cycle schemes is then determined by the respective steady-state mass flows from the previous technology in the sequence and added to the total TWhe produced in the overall fuel cycle scheme. Table 6.1 lists the key technology parameters used to make this equilibrium material-energy balance. The parameters are carried over or derived from the neutronic analyses described in Chapter 3, although to maintain parametric independence, a separate material balance was computed instead of taking directly the per-TWhe mass flows from Chapter 3.

The reactor-dependent burn-up parameter BU (MWd_{th}/kg_{HMI}), the per-pass burn-up fraction BU_f, and the LWR "feed rate" are the main determinants of mass flows down-stream from the driving LWR technology. The input and output HM (heavy metal; U, Pu, MA) compositions, y_{HMi}, and y_{HMf} for each reactor type, along with the respective loss (to repository) fractions associated with the processing step that is a part of the particular technology, also drive these equilibrium mass balances.

Once the equilibrium mass flows to and from each of the NFC steps depicted in Figure 6.2 are determined for a given scenario or scheme, along with associated inventories based on assumed hold-up times for each step in the NFC (see Figure 6.1), unit costs, UC_{xx} ($/kg, $/We, or $/kg·y) are used to estimate the annual charges associated with each step xx of the NFC (e.g. $AC_{xx}(\$/y) = UC_{xx} \times MR_{xx}(kg/y)$, were MR_{xx} is the mass flow rate) with these annual charges being summed over the xx fuel cycle steps. The resulting total annual charge, $AC(\$/y) = \sum_{xx} AC_{xx}(\$/y)$, is summed over each of the reactor technologies (nrx) comprising a given scenario, and this total annual charge is divided by the total net annual electricity generation to give the cost of electricity, COE (mill/kWh), as follows:

$$COE(mill/kWhe) = \frac{AC(\$/y)}{8\,760 \sum_{nrx} P_E(nrx) p_f(nrx)} \quad (1)$$

where $P_E(nrx)$ and $p_f(nrx)$ are the net electric power and the plant availability, respectively, of reactor technology nrx (LWR, FR, ADS).

The TRU-loss going to geological disposal is calculated based on the MR_{xx} values and the respective loss factors in the different fuel cycle steps, depending on the fuel cycle scheme. These TRU-losses are then compared with the amount of TRUs going to repository for the LWR-OFC (scheme 1) and result in the RLOSS-parameter, where RLOSS is the normalised TRU loss to repository relative to the once-through LWR base-case scenario. In addition, the added cost of the NFC fuel cycle scheme (again, relative to the once-through LWR base-case scenario) in question divided by the amount of TRU not sent to repository (avoided through the P&T process) leads to an effective "marginal cost", MC($/kgTRU), that is also reported. As stated previously, all results are presented as comparative assessments for the improvement of each concept rather than as a competitive market selection.

Table 6.1. **Summary of technology parameters used to characterise each fuel cycle scheme**

Reactor technology	LWR-UOX	LWR-MOX*	Pu-burner	MA-burner (FR)	Fast reactor	TRU-burner (ADS)	MA-burner (ADS)
Fuel cycle scheme	1 to 4	2, 3c, 4	2, 4	3a	5	3b, 3c	4
Thermal power, P_{tH} (MWth)	4 240	4 240	3 600	1575	1 575	840	377
Net electric power, P_E (MWe)	1 450	1 450	1 450	600	600	275	119
Plant capacity factor	0.85	0.85	0.85	0.85	0.85	0.80	0.85
BU (GWd/tHM)	50	50	185	140	140	140	140
Burn-up fraction, BU_f	0.0514	0.0507	0.1842	0.143	0.143	0.143	0.143
Equivalent full power days (d)	1 325	1 325	1 320	1 550		870	1 241
Blanket breeding ratio, BR	0	0	0	0	1	0	0
Actinide core compositions							
HM core mass, M_C (tonne)	112.24	112.35	25.69	17.55	17.73	2.59	3.15
Initial HM mass, M_{HMi} (kg/TWhe)	2 394.1	2 394.01	559.25	786.34	794.47	508.92	888.599
U initial fraction, y_{RUi}	1	0.919	0.5557	0.6696	0.7644	0.0121	0.0462
Pu initial fraction, y_{Pui}	0	0.081	0.4443	0.2914	0.2287	0.8237	0.4042
MA initial fraction, y_{MAi}	0	0	0	0.03989	0.0069	0.1642	0.5496
Final HM mass, M_{HMf} (kg/TWhe)	2 270.8	2 267.4	456.2	675.2	691.9	382.2	755.5
U final fraction, y_{RUf}	0.9858	0.9361	0.5714	0.6978	0.7856	0.0161	0.0544
Pu final fraction, y_{Puf}	0.0127	0.0582	0.3953	0.2660	0.2068	0.8016	0.4759
MA final fraction, y_{MAf}	0.0016	0.0057	0.0334	0.0363	0.0077	0.1823	0.4697

* For non-homogeneous cores, e.g. MOX, these initial and final mass fraction pertain to the respective MOX core components (f_{MOX}); the remaining core fractions ($1-f_{MOX}$) are described by the LWR-UOX parameters.

6.2.2 Unit cost data-base

Unit costs[59] were allocated to each of the NFC steps described in Figure 6.1. As this analysis aims at charting cost trends that improve both economic and waste-toxicity performance for each of the technologies considered, rather than presenting a "best-estimate" result, a cost range (i.e. lower bound, nominal value, upper bound) was defined for each of these "top-level" (e.g. highly aggregated) fuel cycle steps. The nominal unit cost value generally refers to best available unit costs for the fuel-cycle or operation in question. This nominal value is based mainly on the results reported in the references, as well as on expert judgement, when necessary (e.g. repository costs, or cost of advanced pyro-chemical processing schemes). The upper bound for the unit costs is based on expert judgement by analysing the sensitivity of the nominal values to changes in technology performance, maturity of technology, economy of scale and other considerations. The same approach holds for the lower-bound unit-cost values that may be considered as the lowest envisaged cost for a given NFC step using the projected mature state of technology. Again, economies of scale, learning-curve effects and expert judgements were applied to derive these lower-bound values. As this costing task evolved, it was decided to use only the nominal cost database for the LWR technology, with each element of that data base being bounded by estimates of standard deviations (σ-values), rather than carrying along three databases per se (e.g. low, nominal, high). In this way, statistical analyses could be applied to a set (for each fuel cycle scheme) of relative costs, rather than having to deal with a range of cost databases wherein the basis or normalising cost was also varying. While "this nominal-plus-sigma" approach clarified the normalisation issue, it raises the problem of adopting σ-values for highly skewed unit cost parameters arises. Additionally, the desire to chart future actions on the basis of parametric systems analyses (PSA), while still retaining a normalisation metric, has potential to confuse when that normalising technology or fuel cycle scheme is also part of the PSA. In some cases, therefore, evaluations based on the variations of absolute costs proved to be clearer.

Tables 6.2 and 6.3 summarise the unit costs applied in the P&T cost analysis. These unit costs and related economic performance assumptions are discussed in the following sub-sections. It is emphasised that not all (detailed) fuel-cycle steps have been taken explicitly into account. Some costs related to interim stock between fuel-cycle steps were aggregated and incorporated into previous or subsequent fuel-cycle steps, whereas in other cases the costs were considered to be of less importance in the present scoping analysis (e.g. differences in cost for re-enrichment of irradiated uranium versus natural uranium, disposal of secondary wastes, etc.). In general, over the long term (i.e. beyond 20 years) the evolution of costs of industrial-scale processes is likely to follow the historic trends (i.e. decreasing in constant terms). The magnitude of such decreases, however, will depend on the vitality of the nuclear industry at both national and international levels, and the availability and sharing of operating experience that drives "learning-curve" cost reductions. Also, fuel-cycle cost comparisons need to take into account financial aspects, such as level of funding, R&D efforts, management of provisions and differences in discount rate (e.g. perceived risks associated with the development and application of a given technology). Depending on the special boundary conditions, NFC costs for individual countries and, in addition, for the individual utilities within a country, may deviate significantly from these generic figures. Regional and temporal differences in key technological and financial parameters, however, must be recognised in assessing the results of this normalised economic assessment that has a main goal of identifying technological improvement needed for each technology being studied, rather than providing a market-selection process/mechanism for the range of technologies being examined by this study.

All unit costs have been expressed in US dollars of 2000. An average annual escalation rate of 3%/y was applied to values based otherwise. An exchange rate of one euro per dollar was applied,

59. Or unit cost estimating relationships, CERs, in the form of $/We, $/kgHM, etc.

when needed, on the assumption that the markets would eventually reconcile present differences. Finally, a fixed charge rate[60] FCR for investments was applied, ranging from 8 to 14%/y with a nominal value of 10%/y. To FCR is added an annual charge rate f_{OM}(1/y) for all (fixed and variable, except for NFC-related charges treated separately), which are operations and maintenance (O&M) expenditures. Decontamination and decommissioning (D&D) charges are expressed by increasing FCR by a factor of $(1 + f_{DD})$ to reflect an additional annual charge being delivered to an D&D escrow fund.

Table 6.2. **Unit costs for nuclear fuel cycle (NFC) steps relating to LWRs**

Step	**Description**	**Unit cost** *Nominal value* *(±σ)*	**Unit***
UC_{MM}	Uranium mining and milling	30 (±*10)*	$/kgU
UC_{CV}	Uranium conversion from U_3O_8 to UF_6	5 (±*2)*	$/kgU
UC_{ER}	Uranium enrichment	80 (±*30)*	$/SWU
UC_{CV}'	Uranium conversion from irradiated UO_2 to UF_6	24 (±*5)*	$/kgU
UC_{FF}(UOX)	UOX fuel fabrication	250 (±*50)*	$/kgHM
UC_{Udepl}	Depleted uranium long-term storage	3.6 (±*1)*	$/kgU
UC_{CS}(SF)	Spent UOX-fuel interim storage (standard 2 years)	60 (±*10)*	$/kgHM
UC_{TR}(SF)	Spent fuel transport	50 (±*10)*	$/kgHM
UC_{PR}(UOX)	UOX reprocessing	800 (±*100)*	$/kgHM
UC_{FF}(MOX)	MOX fuel fabrication	1 100 (±*200)*	$/kgHM
UC_{PR}(MOX)	MOX fuel reprocessing	800 (±*100)*	$/kgHM
UC_{RP}(SF)	UOX spent fuel conditioning and disposal	210 000 (± *50 000)*	$/m^3
UC_{RP}(SF)	UOX spent fuel conditioning and disposal	210 000 (±*50 000)*	$/m^3
UC_{RP}(HLW)	Vitrified HLW conditioning and disposal	400 000 (±*50 000)*	$/m^3
CAP_{LWR}	Capital cost of advanced LWR	1 700 (±*100)*	$/kWe
f_{OM}(reactor)	O&M annual charge for reactor operation as fraction of capital cost	4	%/y
f_{OM}(FF)	O&M annual charge for fuel fabrication plants as fraction of (fabrication plant) capital cost	15	%/y
f_{OM}(PR)	O&M annual charge for reprocessing plants as fraction of capital cost	6	%/y

* All costs are expressed in 2000 dollars. Unit costs for other base-years were corrected using an escalation rate of 3%.

6.2.2.1 Fuel cycle steps related to LWRs

Table 6.2 gives an overview of the NFC steps that are supportive of LWRs, as well as those steps that are common to most of the fuel-cycle schemes under consideration in this study. The unit costs shown in Table 6.2 indicate the nominal values based on expert judgement and literature survey where the applicability (i.e. range) of these unit cost values will be detailed in the following paragraphs. The costs may be considered valid for the short-to-medium time frame (i.e. up to the period 2010-2020). As noted previously, the multiple-database approach adopted for this study (e.g. three CDBs designated as low, nominal, and high) for the LWR-based technologies has been replaced by a single

60. The factor that gives equal annual payments for a total investment cost based in turn on interest during construction of a given construction time, debt-to-equity ratios, escalation rate, interest rate, and plant life.

(nominal) cost database and associated standard deviations (σ values), albeit the statistics of the associated cost distributions usually does not warrant the use of such exact statistical terms.

The cost of natural-uranium mining and milling has been relatively low over the past years, and no significant increase in uranium cost is foreseen in the next decade [168]. Significant uranium resources are available, and new primary resources may be found once (if) uranium demand rises again. Conventional uranium resources are estimated at around 4 million tU (Known Conventional Resources, KCR) that are recoverable at costs ≤130$/kgU. Reasonably Assured Resources (RAR), recoverable at costs of ≤130$/kgU[61], account for 2.3 million tU [169]. The conventional resources alone could sustain present-day nuclear power (once-through) capacity for at least the next 100 years, without significant increase in the cost of energy due to fuel charges. Besides conventional resources, large (unproven) resources of uranium exist. Phosphates are known to contain significant amounts of uranium and another practically inexhaustible potential source of uranium is seawater. The availability of these resources and the possible continuous downward pressure on natural uranium prices lead to an expected cost range of 20 $/kgU up to 80 $/kgU. The lower bound may apply if a buyers' market continues, but such low values would not represent a sustainable price, since a significant number of uranium mines would become uneconomical. The upper bound of 80 $/kgU may apply if a resurgence of nuclear energy increases the uranium demand, with the exploration and mining of new primary resources lagging behind.

The potential availability of an essentially unlimited supply of natural uranium (not to mention at least four times the energy resource in the form of thorium) has significant implications for the waste-disposal and P&T options being considered for the back-end of any expanding nuclear fuel cycle that might utilise such enormous resources at moderate costs. In fact, the P&T issue addressed by this study can be divided in economic, administrative, ecologically and social terms into plutonium management (PUM, mainly an economic and proliferation issue) and minor actinide management (MAM, mainly an ecological and biological hazard issue of longer range). While not part of this study or the associated economic analyses, the separation of the overall problem into PUM versus MAM issues cannot be ignored, even if emerging as key conclusions and recommendations for future work from the present study.

Because of the increasing impact of several market mechanisms (i.e. evolution from an inventory-driven market to a production-driven market, over-capacity in uranium enrichment plants, and the possible introduction of a uranium tail re-enrichment market), developments in conversion and enrichment are oriented mainly towards cost-reduction. It is hoped that in the long-term new facilities, based on advanced processes, could achieve enrichment costs of about 50 $/SWU. The nominal cost indicated in Table 6.2, therefore, can be considered valid for the foreseeable future as sufficient capacity for conversion and enrichment is available and new plants will have to operate in a competitive market where the cost of the service is the primary driver [168]. A lower bound of 50 $/SWU and a higher bound of 120 $/SWU are believed to cover the credible span of costs in the future. The range of costs for conversion from U_3O_8 to UF_6 is 3-8 $/kgU.

A cost indication of 18 $/kgU (1987 $) (i.e. 24 $/kgU and ranging from 15 to 30 $/kgU for the re-conversion of UO_2 coming from reprocessing of UO_2 (UOX) to UF_6 for re-enrichment purposes was given in [170]. In most cases, however, these re-conversion costs are avoided by using depleted uranium or natural uranium for MOX-fabrication, and any costs of conversion to oxide are assumed to be included either in the reprocessing or the fabrication costs [171].

61. Note that an increase of in the price of natural uranium to 130 $/kgU, five times the current price, would only increase the forward cost of nuclear energy by 20%.

The (cumulative) cost for storage of depleted uranium depends on whether the depleted uranium stock is managed as UF_6 or as U_3O_8, which require different processes and therefore have different cost structures. Based on [172], an indicative cost range was derived (i.e. 0.7 to 5.4 $/kgU) covering the long-term storage of depleted uranium in U_3O_8 form in vaults, as well as in UF_6 form in cylinders (although eventually, this UF_6 must be converted back to oxide).

The costs for interim storage of spent UOX-fuel were also reported in [159,170,173], ranging from 40 to 80 $/kgU, where an interim storage time of two years is standard. Reference [171] also reports a cost for "away-from-reactor" wet storage of LWR fuel assemblies (in 1987 $) to be a fixed 50 $/kgHM plus $5 \times T$ $/kgHM within a range of plus or minus 50%, where T(y) is the period of storage. Spent-fuel transport costs have been reported in many publications and amount about 50 $/kgHM (40-60 $/kgHM).

Separated plutonium storage and purification costs were reported in [171], where a range from 1 000 to 2 000 $/kgPu/y was judged to be relevant. The cost of purification from americium would be 18 $/g$Pu_{tot}$ (1987 $) ranging from 10 to 28 $/g$Pu_{tot}$, for plants treating about 2 tonnes Pu_{tot} per annum.

Existing over-capacities in a highly competitive market have led to a significant decrease in the UOX-fabrication price, which presently is in a range between 200 and 350 $ /kgU [168]. With respect to the future development of the UOX-fabrication price, the most important factors are technical developments influencing the fuel-assembly demand (e.g. burn-up increases from the nominal 50 MWd/t to values approaching 100 MWd/t), continued efforts to improve further the efficiency of the manufacturing processes, as well as effects resulting from mergers of suppliers (e.g. reduction of excess capacities, formation of cartels). The same considerations apply for MOX fabrication, ranging from 600 to 1 750 $/kgHM, where a traditional factor of four in the cost of MOX versus UOX fabrication remains valid. The situation concerning reprocessing is different than for fuel fabrication, since this market is characterised by only two main commercial entities with a strong reliance on long-term contracts with certain utilities. New contracts making use of existing facilities indicate significant price reductions are possible as a result of the accumulated experience, as well as reflecting the fact that much of the investment costs have been amortised already. In the future, new plants would benefit greatly from the extensive experience gained during the last decades, thereby allowing to simplify the plants, decrease their size and reduce maintenance requirements. Unit costs for LWR-UOX reprocessing (i.e. from 500 to 1 100 $/kgHM) were based on reported contract prices and assumptions made in other studies [168,174]. The cost for MOX reprocessing (i.e. same range and nominal value as for UOX-reprocessing) is valid as long as the MOX fuel can be mixed with UOX-fuel (i.e. fraction of MOX-fuel remains limited to 20 to 30% in a batch of fuel to be reprocessed).

The conditioning and geological disposal of high-level waste (vitrified or direct spent fuel) is not yet based on industrial experience, and most quoted costs rely on estimates and detailed design studies made in the different member countries under differing assumptions and political and legal circumstances. As the geological conditions and amounts of waste differ according to national nuclear energy programmes, the cost ranges are wide, as are the specific long-term properties of repository containment (e.g. relative importance and time-scale of actinide versus LLFP releases to the accessible environment). Important technical factors that affect costs are the size of the system, time schedule of the disposal project, geological medium (particularly the presence and relative location of water and whether the environment is oxidising), and (to a lesser extent) the engineered-barrier system chosen. Next to these technical factors, social and political issues related to the degree to which the future is discounted and equity of future generations is valued also affect these disposition costs. Lastly, the relative importance of fixed and variable repository costs differs from location to location and has a strong impact on the scaling of unit cost (e.g. $/kgHM) with repository size. These factors will affect the siting and licensing process as well as the overall waste management policy [173]. Studies show the variability of normalised (disposal)

costs depending on the size of the system and the waste management policy. Recent studies in Belgium have indicated that previous estimates can be significantly reduced. Disposal costs are estimated to be 0.2 M$/m^3 or less for spent UOX fuel, and about 0.5-0.7 M$/m^3 for HLW [175,176]. It is also important to consider that the volume of HLW conditioned in glass is about four to ten times lower than the equivalent spent fuel in a metallic canister. Where the conditioning of 1 tonne spent fuel in a once-through fuel cycle scheme results in 2 m^3 of conditioned spent fuel for disposal, only 0.115 to 0.465 m^3 of conditioned vitrified HLW waste arises in a reprocessing fuel cycle scheme.[62] A canister of vitrified waste (volume 0.18 m^3, weight 492 kg) typically contains 47.6 kg of fission products and 3.55 kg of actinides (as oxides) [22]. The rest of the container consists of carrier glass (i.e. 94.3 kg) and the container itself (80 kg). The cost has been expressed in $/m^3 units, since this unit has been widely used in most of the repository design studies. The cost includes only the direct cost associated with waste packaging and disposal, and do not include the costs of R&D, site screening and evaluation, and waste transportation outside the repository site. As this study focuses on steady-state equilibrium situations, the exclusion of these latter costs is appropriate, but the question remains as to who pays for the siting, qualification, and construction of the repository. Over the past five to ten years, new cost assessment studies shave been published in several of the Member countries indicating that the proposed unit cost in Table 6.2 remain valid and may range from 100 000 to 500 000 $/m^3 for spent fuel disposal and from 100 000 to 700 000 $/m^3 for vitrified waste disposal. Certain countries, however, have published significantly higher estimates (e.g. spent fuel disposal in Switzerland [167]).

In this cost analysis, it has been assumed that the disposal costs for vitrified waste arising in the fuel cycle schemes 2 to 5 are proportional to the amount of fission products produced. As mentioned above, a glass canister is essentially loaded with fission products, which define the thermal loading and, therefore, the design of the repository according to present practice. As was shown in Chapter 3, an average amount of 118 kg fission products is produced per TWhe. These waste products would result, on average, in 2.5 waste canisters per TWhe, thereby indicating a conditioning and disposal cost of 180 000 $/TWhe (= 0.18 mill/kWh) that is independent of the fuel-cycle scheme. Since these costs are only a small part of the total generation cost for nuclear energy (on average about 0.5% of generating cost of electricity at 5% discount rate), any reduction of disposal costs attributed to P&T may be considered as having limited impact on the overall cost of electricity.

The unit capital costs (i.e. overnight construction costs including owner's cost) for advanced LWRs were reported in [162] and confirmed during the past years in other studies. These unit (total) capital costs may range from UTC = 1 400 up to 2 200 $/kWe depending on the specific licensing situation in Member countries and local variations in materials and labour costs. Fixed operation and maintenance costs amount to about 65 ± 25 $/kWe/y, or about f_{OM} = 4%/y of the original (total) capital cost. Non-discounted decommissioning costs for LWRs account for about f_{DD} = 8% of the initial (total) capital cost; these D&D costs become negligible when escrowed in a discounted account, with the factor f_{DD} being applied here as an effective increase in FCR to account for such funds being set aside. Interests during construction need also to be taken into account in order to arrive at investment costs and to derive costs of electricity. An average construction time of six years may be envisaged with construction costs split in the proportions of 10%, 15%, 25%, 25%, 20% and 5% between successive years. These ground rules, together with assumptions on debt-to-equity ratios, interest rates, and escalation rates, lead to a fixed charge rate in the range FCR = 0.08-0.14 1/y, with FCR = 0.10 1/y

62. The HLW arising in reprocessing amounts to 0.115 m^3 per tonne of spent fuel, where an additional 0.35 m^3 of ILW arises which is mostly conditioned in the HLW glass matrix. Generally, in the HLW and spent-fuel disposal costs listed in Table 6.2, which are given in terms of unit volume of waste package, a "density" conversion factor is taken as 8.70 tHM/m^3 or 0.50 tonneHM/m^3, respectively. Hence, for the corresponding volume-based unit costs listed in Table 7.1, the weight-based costs of SNF and HLW disposal amount to 420 and 46 $/kgHM, respectively.

being adopted for the nominal value. Hence, for the once-through LWR case of UTC = 1 700 $/kWe, the cost of electricity associated with capital, fixed O&M and D&D charges amounts to $[FCR(1. + f_{DD}) + f_{OM}]UTC/(1 - \varepsilon)/ pf/(8\,760) \times 10^6 = 34.5$ mill/kWh, where the re-circulating power fraction is taken as $\varepsilon = 0.02$ and the plant capacity factor is $p_f = 0.85$. Addition of variable O&M costs push this base-case or normalising value up to ~38-40 mill/kWh.

The annual charges for O&M for reactors (2-5%/y), fabrication (10-25%/y) and reprocessing plants (4-10%/y) are in agreement with today's practice, where no significant changes are expected for new plants or technologies. In fact, higher O&M charges may be expected for advanced fabrication and even advanced processing plants as the control on criticality, safety, working with thermally and radioactively "hotter" fuels, and the implementation of non-proliferation measures may become more stringent. It has been reported that different control measures by international organisations in present-day MOX-fabrication plants represent about 15% of the MOX-fabrication cost [177], above and beyond the costs of maintaining such international organisations, per se. Generally, these O&M (and FCR) factors for fuel-fabrication and processing charges are not used directly in the cost estimates, since the unit costs are expressed in $/kgHM units and already incorporate these plant-related financial and operational factors

6.2.2.2 Advanced fuel cycle step unit costs

The assessment of the costs for the advanced NFC steps is based primarily on a literature survey and in some instances on expert judgement. Table 6.3 summarises the unit costs that will be detailed in this section.

Capital cost of FR plant

The basis for the unit cost assessment is the EFR reactor, as designed by Framatome [178], and the S-PRISM design by GE [179]. Information from SuperPhénix [177] and Monju [180] were included as well as results of the studies performed by ORNL in the 1980s and 1990s [159,180,182]. All the FR-plants considered with MOX-fuel were based on sodium-cooled technology.

The Oak Ridge National Laboratory (ORNL) performed in the 1980s and 1990s several studies related to the technology and economics of ALMR Deployment fuel cycle schemes [159,181,182]. In these assessments, a MOX-fuelled ALMR of 1 488-MWe(net) capacity and 86% capacity factor was considered. The economic analysis in [159] reports an initial investment cost (base construction cost) of 2 825 $million (UTC = 1.90 $/We) for a first commercial plant (with an annual O&M cost of 113.3 $million/y, or $f_{OM} = 4.0$%/y) where an n^{th}-of-a-kind plant would have an initial investment of 2 413 $million (UTC = 1.62 $/We) and an annual O&M cost of 89.6 $million/year ($f_{OM} = 3.7$%/y, all 1992-$).

The EFR-studies indicated a capital cost for a EFR plant some 20-30% higher than for LWRs [178] for a first-of-a-kind plant where n^{th}-of-a-kind plant would only be marginally more costly than LWRs. JNC (Japan) indicates a current capital cost for FR-MOX systems of 4 700 $/kWe, where the future target cost should become 1 700 $/kWe [180].

In 1995, RAND Corporation reported a plant capital cost of 2 760 $/kWe (1987 $). In addition, a range of 1 300-1 800 $/kgHM was suggested for the FR-MOX fabrication and a range of 1 440-1 800 $/kgHM was given for the reprocessing of this fuel [183].

Table 6.3. **Unit costs for advanced nuclear fuel cycle (NFC) technologies**

Step	Description	Unit cost			Unit*
		Lower bound ''lo''	*Nominal value ''nm''*	*Upper bound ''hi''*	
FR with MOX-fuels					
$CAP_{FR\text{-}MOX}$	Capital cost for FR-MOX reactor	1 850	2 100	2 600	$/kWe
UC_{FF}(FR-MOX driver)	FR-MOX driver fuel fabrication	650	1 400	2 500	$/kgHM
UC_{FF}(FR-MOXblanket)	FR-MOX blanket fuel fabrication	350	500	700	$/kgHM
UC_{PR}(FR-MOXdriver)	FR-MOX driver fuel reprocessing	1 000	2 000	2 500	$/kgHM
UC_{PR}(FR-MOXblanket)	FR-MOX blanket fuel reprocessing	900	1 500	2 500	$/kgHM
FR TRU burner					
$CAP_{FR\text{-}TRU}$	Capital cost for FR-TRU burner	1 850	2 100	2 600	$/kWe
UC_{FF}(FR-TRU)	FR-TRU fuel fabrication	1 400	2 600	5 000	$/kgHM
UC_{PR}(FR-TRU)	FR-TRU fuel reprocessing	1 000	2 000	2 500	$/kgHM
All FR					
CAP_{FR}	Capital cost for FR	1 850	2 100	2 600	$/kWe
UC_{FF}(FR_driver)	FR driver fuel fabrication	1 400	2 600	5 000	$/kgHM
UC_{FF}(FR_blanket)	FR blanket fuel fabrication	350	500	700	$/kgHM
UC_{PR}(FR_driver)	FR driver fuel reprocessing[63]	1 000	2 000	2 500	$/kgHM
UC_{PR}(FR_blanket)	FR blanket fuel reprocessing	1 000	2 000	2 500	$/kgHM
ADS TRU burner					
$CAP_{ADS\text{-}TRU}$	Capital cost ADS-TRU burner (excl. target and accelerator)	1 850	2 100	2 600	$/kWe
UC_{FF}(ADS-TRU)	ADS-TRU fuel fabrication	5 000	11 000	15 000	$/kgHM
UC_{PR}(ADS-TRU)	ADS-TRU fuel reprocessing	5 000	7 000	18 000	$/kgHM
ADS MA-burner					
$CAP_{ADS\text{-}MA}$	Capital cost for ADS-MA burner (excl. target and accelerator)	1 850	2 100	2 600	$/kWe
UC_{FF}(ADS-MA)	ADS-MA fuel fabrication	5 000	11 000	15 000	$/kgHM
UC_{PR}(ADS-MA)	ADS-MA fuel reprocessing	5 000	7 000	18 000	$/kgHM
Other					
UC_{beam}	Accelerator cost (incl. target)	5	15	20	$/$W_{beam}$

* All costs are expressed in 2000 dollars. Unit costs for other base-years were corrected using an escalation rate of 3%.

S-PRISM (i.e. 1 520 MWe, 93% capacity factor) was estimated to cost 2 200 M$ (1996 $) (i.e. 1 450 $/kWe (2000 $) [179]). Annual O&M costs were calculated as 76.28 M$/y ($f_{OM}$ = 3.5%/y). The same reference for S-PRISM may be used for this type of FR with metal fuel loading. Comparable information is available from the IFR programme (ANL).

The capital cost values listed in Table 6.3 are given in terms of base construction costs using these references, where the lower, nominal and upper bound capital cost values for FR are higher than the nominal capital cost of an ALWR by +10%, +25% and +50% respectively. The decommissioning costs, interest during construction and O&M- costs for FRs may be taken based on the same assumptions as for LWRs (e.g. same construction period and cost schedule). Unlike Table 6.2, the three databases (low, nominal, and high) were retained because of the skewed nature of the uncertainties, with the "high" value further than the "low" from the "nominal".

63. Blanket and driver fuels are considered to be co-reprocessed as a step towards reducing proliferation propensity.

Capital cost of ADS plants

The main reference used to assess the capital cost for ADS-TRU/MA burning systems is the ATW-Roadmap exercise [8]. The ALMR was considered as a reasonable cost basis for the ATW cost assessment. Modifications to the ALMR design are needed and were in some respects detailed in the ATW-Roadmap document.

The capital cost of an ADS has been divided into two parts, respectively for the accelerator and target (ACC) and for reactor and power-conversion (Rest of Plant, ROP). This latter part includes core, vessel, balance of plant, etc. but excludes fuel-fabrication (FF) and processing (PR) plants, since both the capital and O&M annual charges incurred for the latter two items are expressed on a per-kgHM basis, which includes both capital and O&M charges. This separation of ACC and ROP cost accounts implies that no cost benefit has been attributed to the possible elimination of, for instance, control rods. It was perceived within the context of this cost analysis that possible cost reductions may be offset by cost increases related to complications in containment and other systems.

The basic construction cost for an ADS was therefore set equal to that for a FR with an addition to cover capital costs of accelerator and target. Generally, the target accounts for only a few percent of the total accelerator costs, so at the level of the present costing model, it matters little whether it is included with the blanket or the accelerator. At the highly aggregated level of this analysis, the accelerator cost is estimated on the basis of proton beam power, using unit costs in the range UC_{beam} = 5-20 \$/W_{beam}, with the ATW Roadmap Study [8] giving values close to the upper limit. The costing model [163] developed and used for this study also examines the trade-off between accelerator and material-handling (e.g. fuel-fabrication and processing) where, as in TRU-burning ADSs, reactivity swings are large and the accelerator is sized to maintain a constant fission power over the burn cycle: short batch times lead to a reduced reactivity swing and so to reduced accelerator size, but more mass-handling. Lastly, the cost of added power required to drive the accelerator was accounted through the increased ROP needed to supply that re-circulating power rather than charging for external purchases at some exogenous market price.

The cost of beam (and target) has been based on recent technology assessments [167,8,9,183,184] for accelerators in the power range (beam energy and intensity) of the ADS systems considered in this study. It cannot be over-emphasised that the aggregation of all accelerator costs into a parameter like UC_{beam}(\$/W_{beam}), besides offending accelerator physicists, represents something of an oversimplification in ignoring the possible discovery of some options for reducing both capital and operating costs in this large account. More complete and detailed accelerator models must eventually be used [165].

FR and ADS fuel treatment

Metal or nitride fuels have not yet been fabricated or reprocessed on a semi-industrial scale. Only laboratory- or pilot-plant scale experience exists, for instance in treating EBR-II fuel in the Fuel Conditioning Facility (FCF) by the electro-metallurgical process developed by ANL [185]. Information on these fuel treatment processes is therefore limited and mainly based on conceptual technology assessment studies.

The ATW-roadmap [8] studied the fuel-treatment process in detail and reported [186] unit costs for the processes involved. Other references include the MIT report [187], where a comparison with advanced breeder reactor fuel was mentioned, and the recent DOE report on electro-metallurgical treatment of EBR-II spent fuel [185].

Fuel fabrication costs for the ATW project were reported to be around 11 300 \$/kgTRU (1999-\$) for an nth-of-a-kind plant. This unit cost corresponds to about 2 800 \$/kgFM. The other references were based on the same or comparable assumptions and therefore arrived at the same values.

Experience in fabricating LWR-MOX and FR-MOX fuel suggested a cost increase by a factor of about 5 to 10, since the presence of significant quantities of the minor actinides (particularly the high-activity alpha-emitting americium and plutonium isotopes as well as neutron-emitting curium) demands a new design for fuel fabrication plants to allow for remote handling and criticality concerns. Whereas present-day fuel-fabrication plants can use glove-box handling in almost all of the fabrication steps, these new plants for highly active fuel would require shielded cells and disperse substantial levels of decay heating. In addition, added care must be taken to prevent criticality, and the higher neutron source strengths demand additional shielding. The presence of ^{238}Pu and highly active isotopes of americium and curium in fabrication is, therefore, the most important parameter in assessing the cost of fabrication [177] of these fuels.

Experience in handling fuels containing americium lends greater credence to extrapolations of the costs for fabricating them. This is not true of curium-containing fuels. The unit costs listed in Table 6.3 are based on the available information and take account of the shielding requirements in fabrication. It should be mentioned that all assessment studies of advanced fabrication plants do account for a similar or even higher annual O&M-cost (i.e. 20-25%/y, compared to present-day (UOX and MOX) fabrication plants.

The driver and blanket fuel for FRs are considered to be co-processed, although a difference in fabrication cost has been included to reflect the significant difference in fuel composition. The value for fabricating metallic blanket has been assumed comparable with that for FR-MOX blanket fuel, since no specific cost differences are expected between oxide and metallic uranium fuel.

Despite the difference in specific heat load and annual throughput for the TRU- or MA-fuel fabrication plants in schemes 3a and 4, the same unit cost for fabrication was applied. These fuel fabrication costs would essentially be defined by the throughput of curium.

For fuel reprocessing, the increase by a factor of ten in unit costs was judged to be reasonable, since the composition of the fuel, the size of the reprocessing plants (i.e. throughput), and the technology to be applied are all significantly different from present-day practice, with some exception for the (smaller scale) treatment of EBR-II fuel [185]. Unit costs for reprocessing ranging from 6 000 to 20 000 $/kgHM were derived in references [184-187]. The recent report on EBR-II fuel reprocessing suggests an operational cost of about 15 000 $/kgHM (average value for treating driver and essentially depleted uranium blanket fuel) using the existing Fuel Conditioning Facility at ANL-West [185]. Assuming a new plant using the same electro-metallurgical process, and considering that the annual O&M cost fraction remains about 5-6%/y of the initial investment, the full unit cost for fuel reprocessing lies in the range 19 000-24 000 $/kgHM. This value has been chosen as upper bound for the future pyro-process to be applied in the ADS-schemes, without taking account of economies-of-scale, learning effects and technological improvements. Based on this experience, the Expert Group selected an upper bound of 18 000 $/kgHM as appropriate in the long-term. Despite the difference in specific heat loads and the differences in required throughput of the reprocessing plants, it has been assumed that these two factors compensate each other for the TRU-burning and MA-burning cases, and, therefore, the same unit costs are proposed for the fuel cycle schemes 3a and 4.

The HLW arising from electro-metallurgical processes is composed of two forms, ceramic and metal. The bulk of the fission products and transuranic elements are incorporated into the ceramic, which is a glass-bonded sodalite monolith. The metal contains fuel cladding, the remainder of the fission products, and trace amounts of uranium [185]. Based on Reference [185], a unit cost for disposal of these waste forms would amount to about 400 $/kgHM (net present value cost; the cost for disposing a canister of HLW in a geologic repository was estimated to be $475 000). The latter disposal cost was assumed to cover also this kind of HLW waste from the electro-metallurgical process, as would make the value accord with Table 6.2 and the assumption on HLW-cost mentioned above.

6.3 Results

The results of the cost analyses divide into two parts:

- The relative cost and TRU loss to repository according to the data largely summarised in Tables 6.1-6.3 and designated the Point-of-departure (POD) case.
- The impact of single-point parameter variations or departures from that POD case designated as Parametric Systems Analyses (PSA).

The following two sub-sections present these results accordingly.

6.3.1 Point-of-departure case

The aggregated, relative costing and (TRU) loss to repository results, as embodied in the ratios RCOST = COE(nsc)/COE(1) and RLOSS = $LOSS_{TRU}$(nsc)/$LOSS_{TRU}$(1), are summarised in Table 6.4 for the seven fuel cycle schemes considered by this study.[64] Figures 6.3 and 6.4 present parts of Table 6.4 in graphical form to facilitate cost comparison between fuel cycle schemes (Figure 6.3) and the cost-versus-loss trade-offs (Figure 6.4).

The main message from these results is that reduced TRU losses to repository imply an increased overall system cost of electricity, and that certain combinations of technologies cost more than others on the cost data assumed. Furthermore, those fuel-cycle schemes that use more expensive technologies (e.g. ADS-based) show an overall economic benefit in burning as much plutonium as is possible in more conventional systems such as MOX-LWRs and MOX-FRs.

The marginal cost results show that the cost of avoiding disposing of TRUs remains modest for schemes 2, 3a and 4; it amounts to about 30-40% of the conditioning and disposal cost for 1 kg of HM as vitrified HLW.

A comparison of the LOSS values in Table 6.4 with the HLW production values in Table 3.3 shows that the precision of the combined cost and mass balance model is limited. However, this limitation has to be balanced against the simplicity of the model which allows e.g. parametric systems analyses to be carried out easily.

Table 6.4. **Aggregated costing and TRU-loss (to repository) results for the Base or Point-of-departure case**

Fuel cycle scheme (nsc)	**1**	**2**	**3a**	**3b**	**3c**	**4**	**5**
U-loss (kgU/TWhe)	2 299.23	1.77	1.45	1.86	2.09	1.67	4.65
Pu-loss (kgPu/TWhe)	29.53	0.1	0.10	0.16	0.16	0.12	0.26
MA-loss (kgMA/TWhe)	3.66	9.47	0.01	0.03	0.03	0.03	0.005
TRU-loss (kgTRU/TWhe)	33.19	9.57	0.12	0.19	0.18	0.16	0.26
RLOSS	*1*	*0.2883*	*0.0035*	*0.0057*	*0.0055*	*0.0047*	*0.008*
COE (mill/kWh)	38.02	40.70	42.41	53.48	49.44	44.16	56.86
RCOST	*1*	*1.07*	*1.12*	*1.41*	*1.30*	*1.16*	*1.50*
Marginal cost MC (‘000 $/kgTRU)		113	133	468	346	186	572

64. In the remaining of this chapter, “nsc” will indicate the fuel cycle scheme number, e.g. nsc = 2 refers to the fuel cycle scheme 2.

Figure 6.3a. **Comparisons of relative fuel cycle scheme costs to the Base or POD case. (UC_{beam} = 15 \$/W_{beam}; BU(LWR) = 50 GWd/tHM; nominal unit cost values)**[65,66]

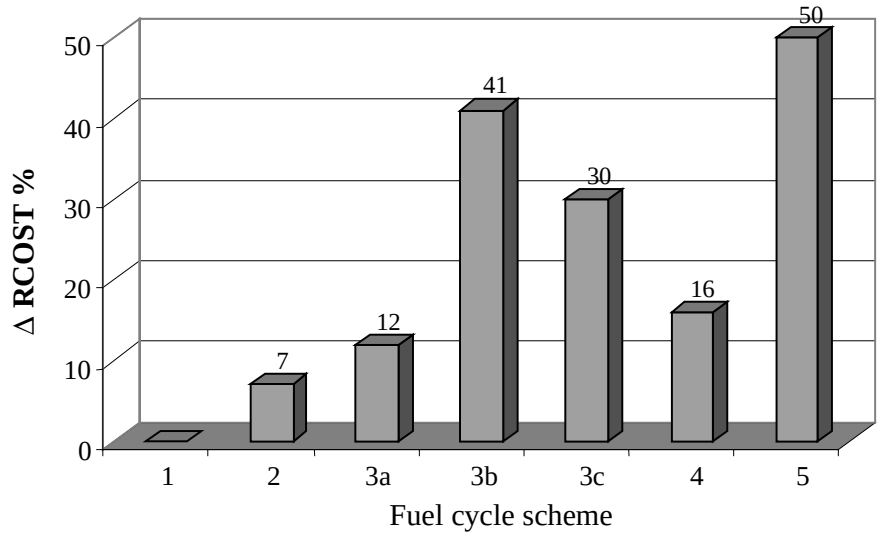
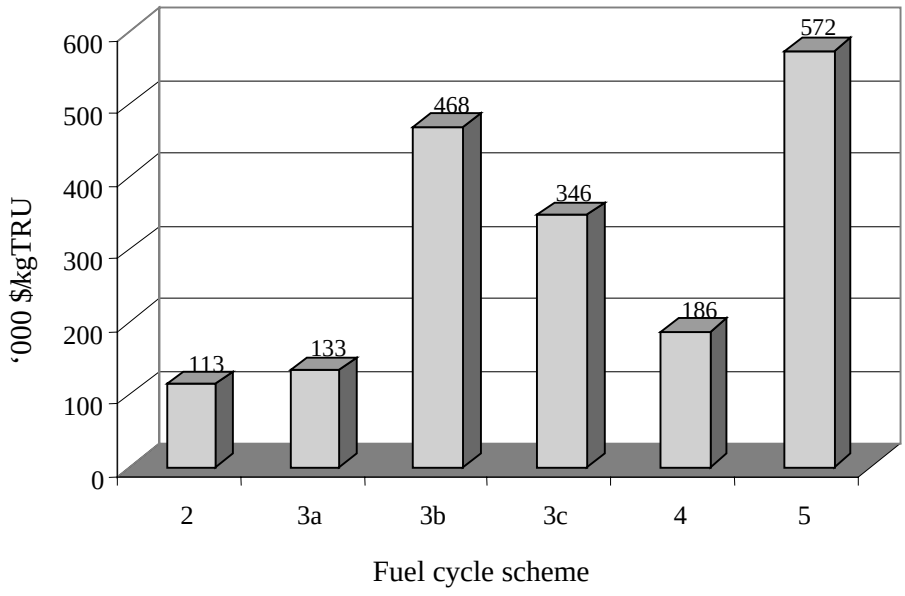
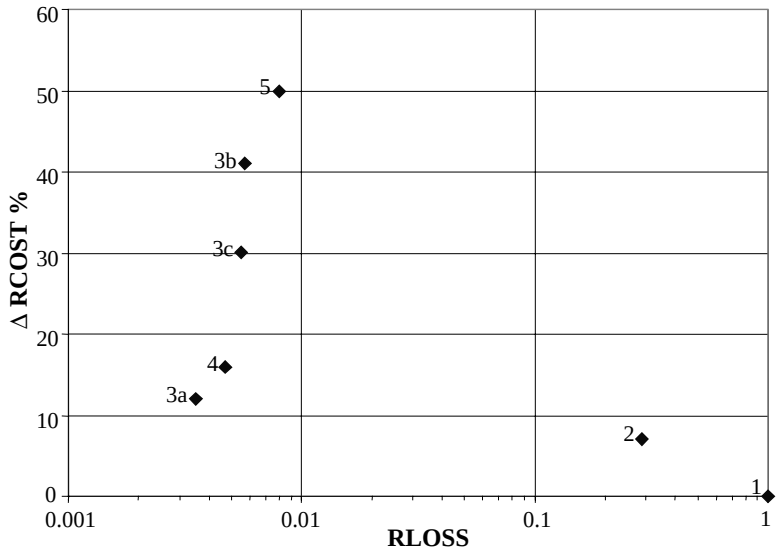


Figure 6.3b. **Marginal cost MC ('000 \$/kgTRU)**[67]



65. ΔRCOST = (RCOST - 1) × 100.
66. The fast reactor model used in this study was optimised for burning (scheme 3a) and had a high surface to volume ratio to promote leakage. Converting it to fissile self-sufficiency required more blanket mass flow than would be the case when the core is originally designed as a low surface to volume breeder; the resultant cost of blanket recycle shown here therefore is an upper bound.
67. MC = [COE(nsc) - COE(1))/(LOSS(nsc) - LOSS(1)].

Figure 6.4. **Comparisons of costs fuel cycle scheme and TRU loss trade-offs with the Base or POD case. (UC_{beam} = 15 \$/W_{beam}; BU(LWR) = 50 GWd/tHM; nominal unit cost values)**



6.3.2 Parametric system analysis

In spite of the simplicity of this highly aggregated, equilibrium nuclear fuel cycle model and the aggregated unit costs used to convert the resulting mass flows and inventories into annual charges, the number of economic and technological parameters is large. Since varying all these parameters is both counterproductive (diffusive) and impracticable, it was decided to vary predominantly those parameters that might improve some of the more costly concepts. Therefore, accelerator cost for ADS-based fuel cycle schemes and fuel burn-up in the driver LWR technology were identified as relevant subjects for these PSAs. Before embarking on these single parametric cost variations, the impacts of the higher-level parameters like unit cost base per se and the burn-up fraction, BU_f, for systems with fully closed fuel cycles are reported.

6.3.2.1 Cost data base variation

The uncertainty in the CDB listed in Tables 6.2 and 6.3 is expressed in two forms: standard deviations (σ-values) for the more developed technologies (Table 6.2); and specific upper ("hi") and lower ("lo") bounds placed on unit costs relative to the "nominal" ("nm") or base-case values (Table 6.3). It can be seen that some of the uncertainties are large. First, emphasis is placed on examining the impact of the cost ranges associated with the advanced technologies (Table 6.3). Figure 6.5 compares the percentage change in relative costs ΔRCOST for each fuel cycle scheme as the cost database (CDB) is changed. According to the structure represented in Tables 6.2 and 6.3, the normalising LWR-OFC cost remains unchanged as the different CDBs are selected. While the cost ordering of fuel cycle schemes remains unaltered as CDB is changed, movement away from the nominal cost base case, CDB(nm), in either "hi" or "lo" directions changes in these relative costs by a factor of about two.

Figure 6.6 shows the correlation of ΔRCOST plotted versus RLOSS for schemes 2, 3a, 3b, 3c and 4. The primary conclusions from these comparisons are that the cost sensitivity over the CDB range embodied in Table 6.3 is large, but the RCOST ordering of the schemes remains the same as for the

POD, and that the economic advantage of pre-burning as much plutonium as possible in less expensive technologies is hence re-confirmed; this ordering is not affected by the choice of CDB.

It must be noted, however, that these results were obtained under the assumption that all low or upper bound values would occur simultaneously for each cost account, and the realism of achieving such an absolute bound is open to question.

Figure 6.5. **Impact of unit cost ranges for advanced-technology costs as reflected in Table 6.3 on the relative costs associated with each fuel cycle scheme**

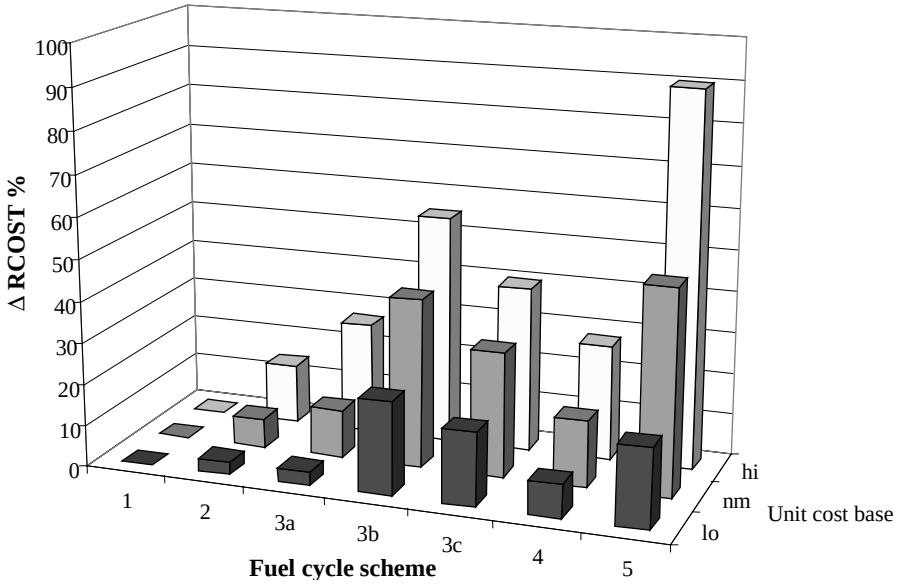
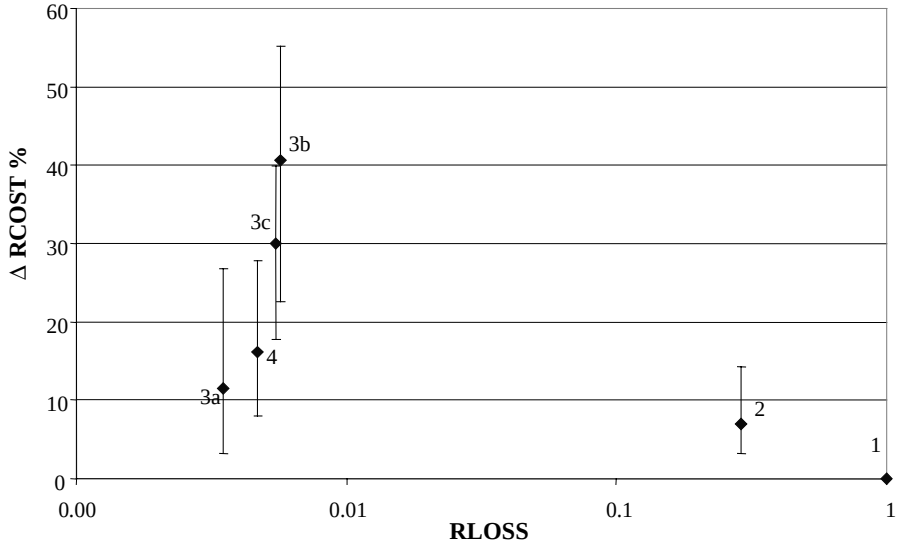


Figure 6.6. **Comparisons of fuel cycle scheme costs and TRU-loss trade-offs (BU(LWR) = 50 GWd/t), with the range for each point (scenario) being determined by the cost-data-base selection CDB (lo, nm, hi)**



6.3.2.2 Burn-up fraction variation for systems with fully closed fuel cycle

The equilibrium mass-balance, aggregated-costing model has been used to perform single-point parametric studies. A crucial parameter in this regard is the per-pass burn-up fraction, BU_f, for the fully closed fuel cycle schemes 3a, 3b, 3c, 4 and 5. Figure 6.7 shows the variation in both RCOST and RLOSS as BU_f for the systems with fully closed fuel cycles is varied. Both cost and loss seem to saturate at or above the POD value of $BU_f = 0.14$, but both increase rapidly as burn-up fractions fall below $BU_f = 0.05$. The lower burn-up fraction results in a steep increase in the number of recycles needed to transmute a certain amount of TRUs, and, therefore, each recycle pass adds to losses in reprocessing and fabrication as well as to the costs for these processes. For high burn-up fractions, the number of recycling iterations becomes smaller and the cumulative effect of losses-per-cycle saturates to a lower value.

As in scheme 4 only the small, second stratum mass flows are affected by the parameter variation, the RCOST and RLOSS values saturate at lower burn-up fractions per pass than in the other schemes. This result confirms again the overall economic benefit from pre-burning plutonium in lower-technology systems to the maximum extent possible.

Figure 6.7. **Relative (TRU to repository) loss, RLOSS, and dependence of relative cost, RCOST, on burn-up fraction, BU_f, for the fully closed fuel cycle schemes (3a, 3b, 3c, 4, 5) for otherwise POD parameters**

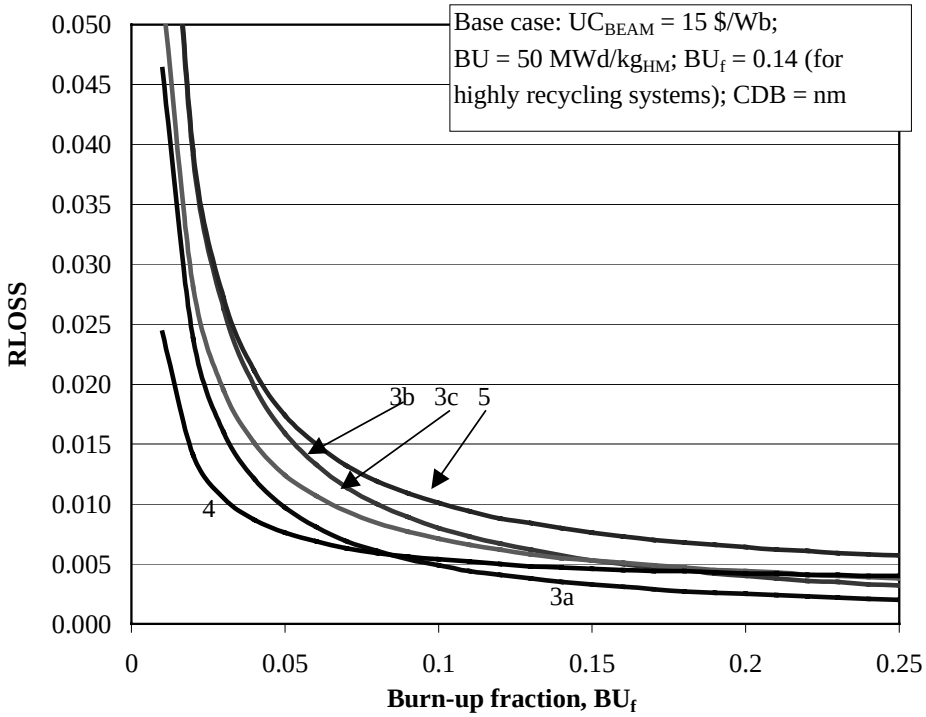
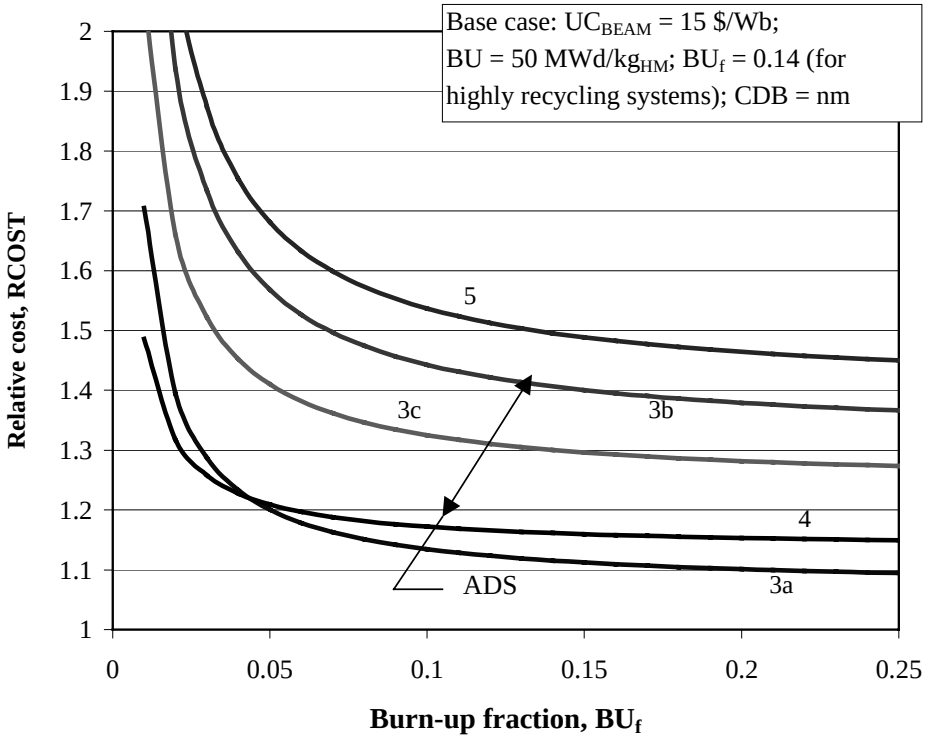


Figure 6.7. **Relative (TRU to repository) loss, RLOSS, and dependence of relative cost, RCOST, on burn-up fraction, BU_f, for the fully closed fuel cycle schemes (3a, 3b, 3c, 4, 5) for otherwise POD parameters (Cont'd)**



6.3.2.3 Accelerator unit cost variation

As elaborated in [165], the equilibrium mass-flow balance model, for each ADS-based technology, cost optimises the cost of accelerator power through the selection of a final blanket neutron multiplication, k_{eff}^{f}, for a given initial value, k_{eff}^{i}. This final multiplication is chosen in a trade-off between accelerator power and fuel cycle operations (i.e. fuel fabrication and reprocessing) so as to minimise the cost of electricity in those ADS systems such as TRU burners that experience large reactivity swings during operation at constant power. Typical accelerator parameters required for such an optimisation are listed in Table 6.5.

The main costs for ADS-driven schemes are the additional Rest of Plant (ROP) costs arising from the power consumed by the accelerator and the capital and non-electricity O&M charges associated directly with the accelerator. In this context, the highly aggregated unit capital cost for the accelerator, UC_{beam} ($/$W_{beam}$), was judged to be the most important parameter on which to perform PSA. The accelerator efficiency was judged to offer little room for improvement, and hence was not subjected to PSA.

Table 6.5. **Typical (generic) accelerator physical and economic parameters required for the ADS accelerator "optimisation"**

Fixed physical parameters	
Target yield for E_P = 1 GeV	22.9
Neutrons per fission, ν	3
Energy yield per fission (MWth.y/kg)	2.7
Fission yield (MeV/fission)	200
Fixed system parameters	
Initial blanket multiplication, $k_{eff,i}$	0.98
Final blanket multiplication (limit), $k_{eff,f}$	0.92
Flux peaking importance function, φ_S	1
Thermal to electric conversion efficiency, ε_{th}	0.37
Accelerator efficiency, ε_A	0.45
Auxiliary (non-accelerator) power fraction, f_{Aux}	0.02
Fixed costing parameters (See Table 6.3)	
Unit cost of transmuter ($/kWe)	2 100
Unit cost of accelerator, UC_{beam} ($/$W_{beam}$)	15

Figure 6.8 illustrates the impact on the relative cost of varying UC_{beam} above and below the POD value of 15 $/$W_{beam}$. Scheme 3b is most affected by accelerator-related charges, since this scheme requires the highest number of ADSs to burn the TRU generated by a given capacity of once-through LWRs (power from an ADS would be 2-3 times as expensive as from LWRs if charges were not shared throughout the system). If the aim is to implement an ADS-transmuter under optimum economic conditions, plutonium discharged from LWRs should preferably be consumed in the less expensive systems, as is done in scheme 3c, and even more so in the double-strata scheme 4. Furthermore, ADSs should be designed to minimise the decrease in k_{eff} resulting from burn-up, and this is especially necessary in TRU burners.

Figure 6.8 also illustrates the increasing economic de-coupling of the costs of electricity from the accelerator cost in the direction of schemes 3b to 3c to 4, as both the magnitude and the gradient of the additional costs are reduced. This behaviour is shown even more clearly in Figure 6.9, which indicates a 12-20% penalty relative to the LWR once-through fuel cycle scheme even for an accelerator with zero capital costs.

Figure 6.8. **Impact of accelerator unit cost on relative cost for all fuel cycle schemes examined; only fuel cycle schemes 3b, 3c, and 4 use accelerator-based technologies**

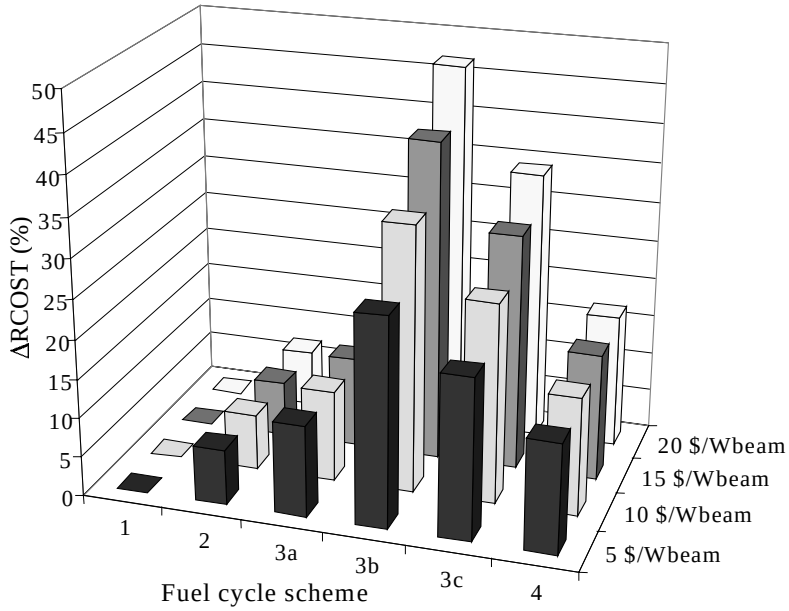
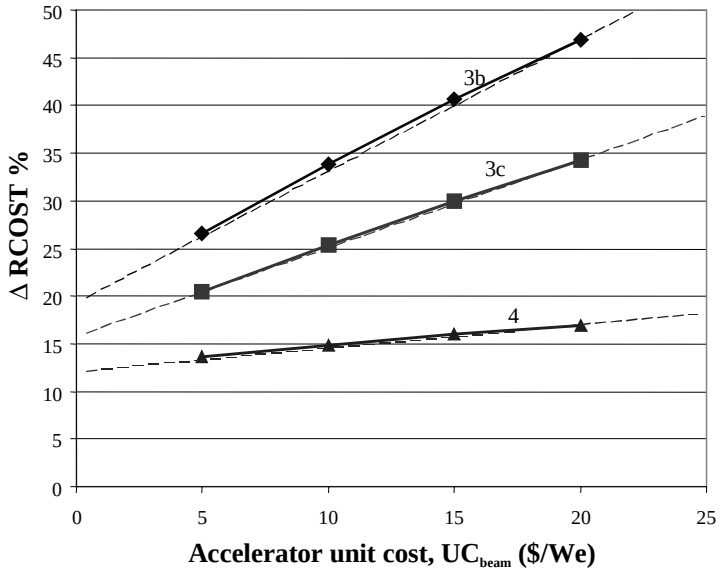


Figure 6.9**. Dependence of relative system generation cost on accelerator unit total capital cost for fuel cycle schemes based on ADS; both the magnitude and the gradient of the accelerator cost impact are reduced as less-expensive technologies are used to consume TRU**

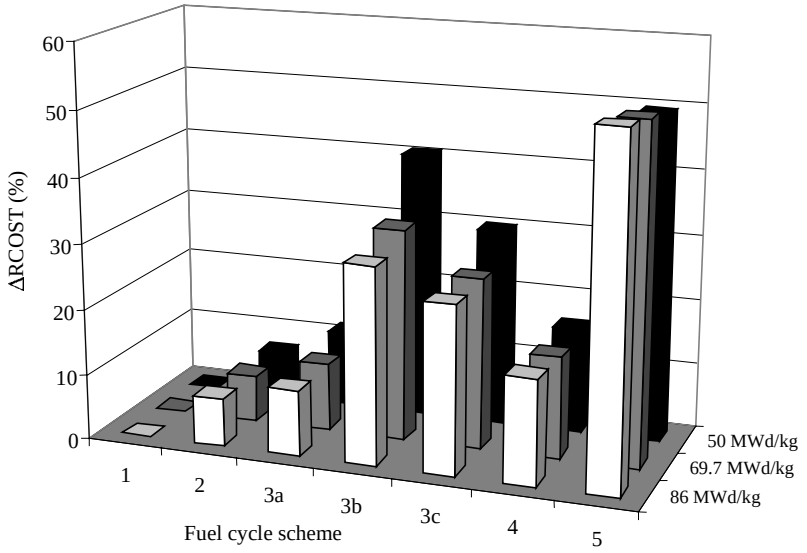


6.3.2.4 LWR burn-up variation

As was shown in the previous section, if ADS-driven transmutation technology is to be realised economically, then either the unit cost or efficiency of using accelerators must be improved, or less-expensive transmutation technologies must assume more of the burden of consuming transuranic elements. Fuel cycle schemes 3c and 4 attempt to achieve this latter goal by first fissioning as much

plutonium as is possible using more standard LWR or FR technologies. A more direct approach to reduce the burden placed on specific transmutation systems would be to achieve greater burn-up in the once-through LWRs. Preliminary results[68] indicating the impact of higher burn-up, BU (MWd/kgHM), in these driver once-through LWRs. Figure 6.10 shows the impact of increasing BU(LWR) on RCOST, and Figure 6.11 presents the shifts in the RCOST versus LOSS relations as BU(LWR) is varied. These cost impacts include the cost of higher ^{235}U enrichments required to achieve the higher burn-up values, but neither any added cost of processing more highly irradiated fuel nor the cost of added cooling storage. It is noted from Figure 6.10 that the relative cost for fuel cycle scheme 5 increases with BU in the once-through LWR technology as the normalising LWR case is becoming somewhat cheaper with increases in its BU value.

Figure 6.10. **Impact of LWR burn-up on relative overall system cost of electricity for all fuel cycle schemes considered**



6.3.2.5 Breakdown of electricity cost

Chapter 3 indicated that the different fuel cycle schemes examined need different combinations of conventional and advanced technology for reactor and fuel cycle facilities to meet the assumed steady-state equilibrium condition. Figure 6.12 shows the breakdown of the cost of electricity for the different fuel cycle schemes according the type of technology involved.[69] In general, the same structure is found as represented in Figure 2.5.

68. These results have been obtained by a different calculational methodology from the one used for the detailed mass-flow calculations in Chapter 3 and therefore need to be seen as an approximation.

69. LWR (UOX and MOX) reactor and fuel cycle facilities as well as FR-MOX reactor (Pu burner) and associated fuel cycle facilities are considered as standard technology, whereas all the rest of the technologies (FR MA/TRU burner and ADS) are considered to be advanced.

Figure 6.11. **Impact of LWR burn-up on the RCOST versus RLOSS trade-off for five of the fuel cycle schemes (the arrow shows the trend for LWR burn-up, BU, varying from 50 to 69.7 and 86 GWdth/tHM)**

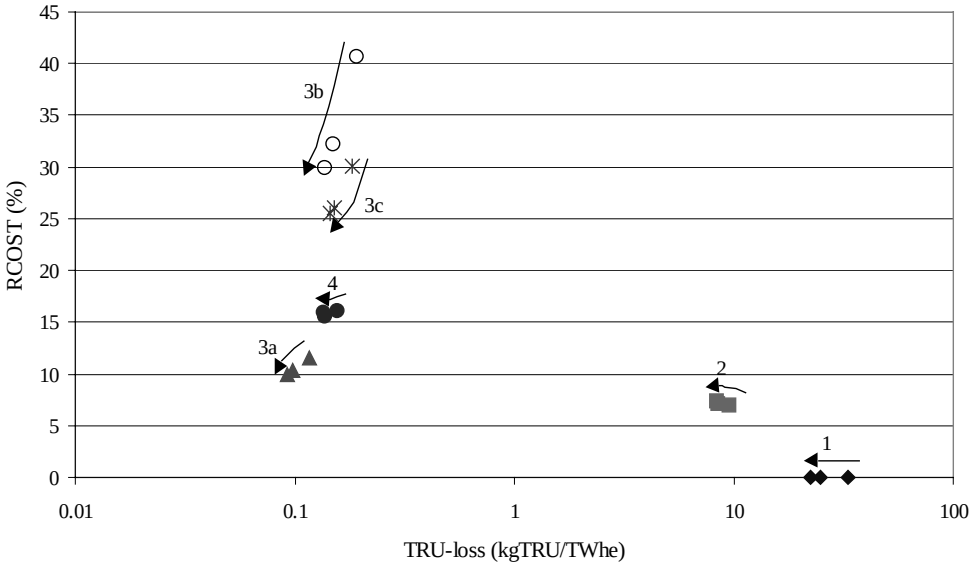
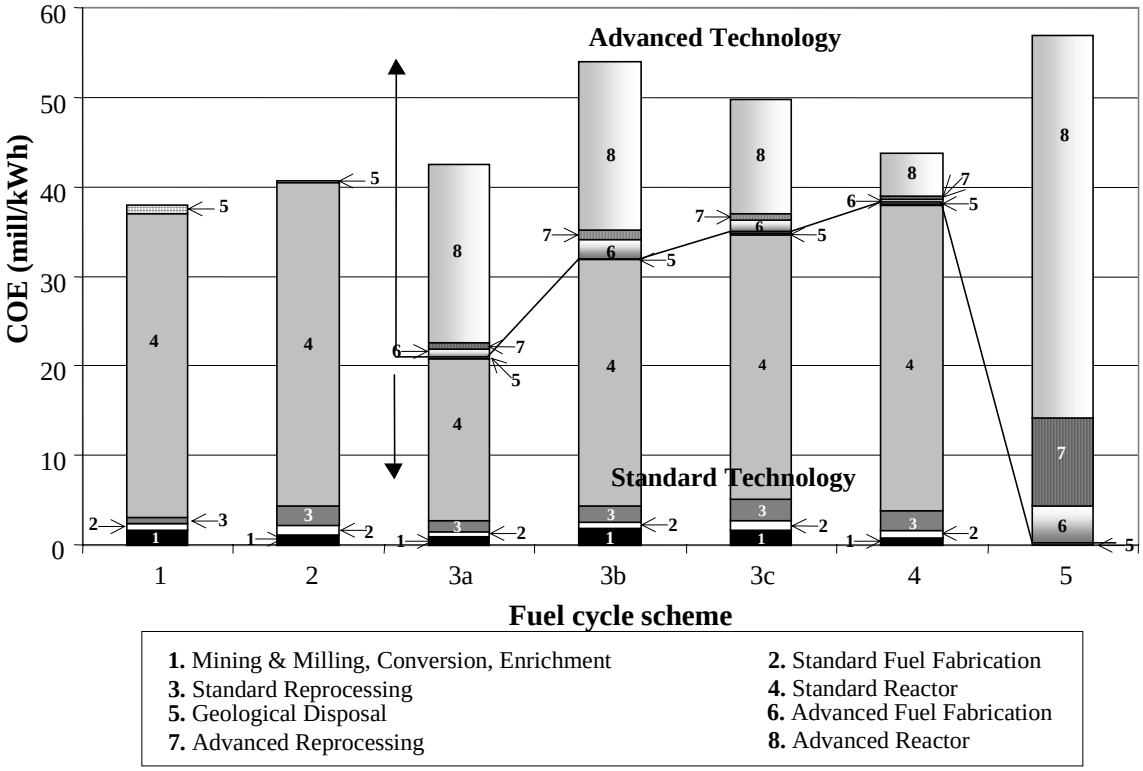


Figure 6.12. **Cost breakdown for fuel cycle schemes**



The dominant capital costs for LWR-technology, including the annual O&M charges, result in the low fraction of COE allocated to the nuclear fuel cycle in scheme 1. Chapter 3 showed that fuel cycle schemes 3c and 4 needed the least amount of advanced reactor technology in the nuclear reactor park. Figure 6.12, in combination with Figure 6.6, shows that fuel cycle schemes 3a and 4 compare rather well in terms of cost-effectiveness. The fuel cycle schemes differ significantly in financial risk, expressed as that part of the system-wide COE related to advanced fuel cycle or reactor technology, with fuel cycle scheme 4 showing both a favourable cost-effectiveness and lower financial risk. The financial risk would, however, become quite comparable if one considers the FR-MOX system to be also advanced technology. Under this assumption, the closed fuel cycles schemes 3a, 3b and 4 would involve a financial risk of about 40 to 50% of the system-wide COE. In these conditions, the fuel cycle scheme 3c would show the lowest financial risk, i.e. about 30% of COE allocated to advanced technology.

6.4 Summary conclusions

This chapter deals with the relative energy costs of the seven fuel cycle schemes examined (1, 2, 3a, 3b, 3c, 4, 5). They are expressed as cost of electricity normalised to that of scheme 1 with the once-through fuel cycle in LWRs. A relative cost of electricity was chosen to focus on the *comparative* and de-emphasise *competitive* market-oriented cost assessments. The level of this assessment is sufficient to indicate the technological improvements that might enhance the future market position of the investigated concepts, but the models used are too broadly aggregated and the extrapolation from the current technology is too large to allow a clear and meaningful rankings of the concepts for the purpose of market-base down selections to be made.

Unsurprisingly, this comparative analysis indicates that a reduction of TRU delivered to repository is accompanied by an increased *system-wide* cost of electricity. This increase is within the range 10-50%, depending on the technologies invoked. It results from increased material handling of highly radioactive fuels (e.g. in fuel fabrication and processing) and the introduction of capital-intensive transmutation technologies needed to deal with the flow of plutonium and MA from the back-end of the once-through LWR fuel cycle. Additionally, certain fuel-cycle schemes involve varying increases in the system-wide cost of electricity, depending both on the efficacy with which the advanced back-end P&T technologies deal with this material flow and the related support ratios needed to achieve the steady-state or equilibrium material balance.

In line with these observations, and in spite of the limitations of the cost-aggregated, equilibrium model used to generate these comparative results, this cost analysis allows the following conclusions to be drawn:

- Fully closed fuel cycles may be achieved with a relatively limited increase in the cost of electricity of about 10 to 20% compared to the LWR once-through fuel cycle. In the case of partially closed fuel cycles, i.e. closed only for plutonium, the cost increase is less and becomes about 7%.

- Among the fully closed fuel cycles schemes, the schemes 3a (TRU-burning in FR) and 4 (Double Strata) result into the lowest increase in system-wide cost of electricity relative to the once-through LWR normalising fuel cycle scheme 1.

- Fuel cycle schemes that involve the use of the more expensive ADS-technology show an overall economic benefit by burning as much of the plutonium as possible in less-expensive, more conventional systems, i.e. MOX-LWRs and MOX-FRs.

- The marginal cost of avoiding sending TRUs to a repository is estimated to be less than 200 000 $/kg TRU. Assuming a nominal 1% TRU content in spent fuel or high-level waste, this translates into an equivalent of less than 2 000 $/kgHM.

- For the closed fuel cycle schemes, the advanced technology contribution to the system-wide cost of electricity accounts for about 10 to 50%. If all non-LWR technology is considered as advanced, this contribution becomes about 30 to 50% except in scheme 5 where it is 100%. Fuel cycle scheme 4 benefits from burning as much as possible of the TRUs in standard technology facilities.

- The costs associated with the TRU-burning in ADS fuel cycle scheme 3b are most influenced by the accelerator-related charges, which are shared comparably between the capital charges for the accelerator and the added generation plant needed to supply it with re-circulating power. If the accelerator costs could be reduced by a factor of three, the increased cost of electricity for this fuel cycle scheme could be reduced to 25% (but still higher than for schemes 3a and 4).

- The economic incentive to increase the burn-up fraction in the TRU- or MA-burners beyond 0.15 becomes marginal. Further reductions in TRU-losses to repository at an acceptable system-wide energy costs are therefore to be obtained preferably by improvements in reprocessing technology (e.g. reduced losses and costs).

- The FBR fuel cycle scheme 5 is more expensive than the other fuel cycle schemes for the cost databases assumed. Large reprocessing charges related primarily to the blanket, needed to breed sufficient fuel for self-sustainability, represent a substantial cost item; this situation can change when more optimistic unit costs for both processing (Purex for blanket and pyro-chemical for driver fuel) and capital plant are used. On the other hand, the fuel cycle scheme 5 is not like any of the others. In addition to dealing with its actinide waste stream, it also utilises the uranium resource approximately two orders of magnitude more effectively than the other fuel cycle schemes, naturally at some cost.

Finally, this cost analysis has once again confirmed that nuclear energy may cope with its waste while limiting the extra cost that this would entail. While outside the scope of this study, it is up to society in the future to decide whether these additional costs are justified and economically acceptable in order to deliver a more sustainable nuclear energy source.

7. R&D NEEDS

7.1 Introduction

Chapter 5 gave an overview of the development and the required figures of merit in the technology for fast reactors and accelerator-driven systems while Chapter 6 addressed the safety constraints on ADS. This chapter focuses on the perceived needs for research and development (R&D) by briefly describing the ongoing and planned R&D-projects, and aims at identifying the possible gaps in R&D activity.

A lot of research was started in the early 1950s on the development of fast reactors and continued until now, however at a slower pace. This initial development was essentially based on the perception that uranium resources could become scarce after a significant deployment of nuclear energy. As history has shown, this initial argument faded over the past decades and today only a few countries remain active in the field of fast reactor development, while the transfer and consequently the build-up of expertise in this field has come to a halt in most of the OECD Member countries. In addition, the experimental facilities, e.g. fast irradiation reactors, have been closed down or have come under strict operating conditions which make further R&D difficult.

As today's society is particularly concerned about the waste management aspects of nuclear energy, a renewed interest in accelerator-driven systems emerged in the 1980s and especially in the 1990s, benefiting from the similarity with fast reactor technology and the existing experience base. While in essence being fast reactors, these accelerator-driven systems aroused interest in re-launching past activities in the fields of cross-section measurements, benchmark and integral experiments, materials research and irradiation programmes, as well as increased cross-disciplinary research with the nuclear physics and accelerator community. Especially since the early 1990s, this revived interest has crystallised in new R&D programmes on ADS and related fields, even in OECD Member countries that took little part in the fast reactor scene.

Several assessments of ADS technology requirements have been undertaken since these early 1990s, the most relevant being the review of the OMEGA project in Japan, the ATW roadmap exercise in the US and the European TWG roadmap. Other countries undertook national evaluations in order to analyse the opportunities for new experiments or even new research infrastructures for ADS development. Some additional funding was granted by Japan, US and the EC to launch several activities in the field but most of these efforts have so far remained less extensive than the funding for FR technology in the 1970s and 1980s.

This chapter aims to give an overview of the different issues to be raised for further development of ADS or FR in the context of the fuel cycle schemes discussed, the ongoing and planned activities as well as the perceived need for increased focus in future R&D-programmes. It will address some elements of the discussion on the need for a new dedicated ADS infrastructure while highlighting the potential interest in increased international collaboration. As in the previous chapters, use will be made of an overview Table 7.1 showing the field of activities and perceived R&D needs.

7.2 Technology goals for P&T, and especially ADS development

This study has focused on the integration of P&T in advanced nuclear fuel cycles and the specific issues relating to the different steps in the fuel cycle and especially to the reactor part, be it ADS or FR. It has become obvious from Chapter 3 that developments in the reactor system are closely connected to the fuel cycle. The significant difference in residual decay heat for comparable fuel types used in different fuel cycle schemes, and the difference in doses and activity during fuel fabrication, are examples of this dependence. Chapters 5 and 6 also raised important issues for further R&D.

In the context of this study, two main blocks of R&D requirements may be identified:

- *R&D related to the fuel cycle*, which is to a large extent the same whether FR or ADS systems are to be used in such fuel cycles. However, some differences appear, such as the type of fuel that may need to be fabricated and reprocessed, the residual decay heats, etc. Main items for consideration in this block relate to:

  – *Adequacy of separation of uranium, TRU, and long-lived fission product* elements from LWR-UOX or MOX spent fuel and FR or ADS transmuter fuel. The separation efficiencies of 99.9% still need to be proven on an industrial scale. As was shown in Chapter 3 and Chapter 7, achieving these high efficiencies is crucial to attaining a significant reduction in mass going to waste. The cost analysis in Chapter 7 indicated that improvements in separation technology have a direct impact on the mass reduction while cost-effectiveness is crucial.

  – *Quantification of total long-lived radioactivity generated* in the FR or ADS transmuter system, including spallation products from the ADS, and the implications for waste streams and waste forms. The quantities actually generated will be functions of the processes used and the separation efficiencies. Design features and scale of operations will determine how much residual waste will still require disposal in a geological repository or other waste facility. Assessing the amounts and compositions of the secondary LLW, ILW and HLW streams may help future assessment studies by indicating where investments for improvement may be most appropriate.

  – *Performance assessment for a geological disposal site using a P&T source term* is necessary to clarify the cost-benefit analysis of a P&T scenario including this geological disposal. Only such a complete assessment can tell whether P&T is really effective. Identifying criteria for future P&T work should be a key driver for such assessment studies. In general, a study looking to new options for repository design, taking into account developments in advanced fuel cycles, would be a welcome endeavour. In fact, the overall P&T issue addressed by this study splits in economic, administrative, ecologically, and social terms into those of plutonium management (mainly an economic and proliferation issue) and minor actinide (MA) management (mainly an ecological and biological hazard issue of longer range).

Table 7.1. **Outline R&D matrix for P&T technology**

Technological objectives		(2) Pu-burning	(4) Double strata	(3b) TRU burning in ADS	(3a) TRU burning in FR (5) All FR
Partitioning	**99% recovery**	*1st Stratum*	*2nd Stratum*	**Dry reprocessing: 99.9% TRU recovery**	
		Wet reprocessing 99.9% U, Pu, (Np) recovery	Dry reprocessing 99.9% TRU recovery		
	R&D needs	Reprocessability of high-Pu-content fuel with high recovery	Separation of U from TRU?		
		Separation of actinides from lanthanides			
	Short cooling times	Needed in order to keep low inventory in cycle			
	R&D needs	FR-MOX reprocessing High residual decay heat requires changes in Purex process: pin-choppers, centrifugal contactors, laser cutting; alternatively, longer cooling times, dilution with LWR-UOX feed	– Material corrosion by salts and high temperature – Batch process: development of transport and loading techniques for material – Pin-choppers or laser cutting techniques in head end		
	Additional or alternative processes	Dry reprocessing of oxide fuel	– Wet reprocessing may remain possible to co-extract Am + Cm – Dilution with LWR-MOX or FR-MOX to reduce mean decay heat	Wet processing: Purex or Urex	
Fuel fabrication	**High burn-up fuel**	Burn-up limitation due to swelling rates and gas pressurisation: modelling, development and irradiation under normal and power transient conditions needed			
	High TRU/MA loading	***Oxide fuels***	***Nitride fuels***	***Metal fuel***	
		– High Pu content (ca. 45%) fuel – Effect of 238Pu on fabrication – MA content reduces margin to melting owing to low thermal conductivity. Limitation to fuels with low residual decay heat	– Reduced stability against decomposition at high temperatures – Fabrication process may be unsuited to “ad hoc” tailoring	– Need to improve thermal properties, adding non-fissile metals with high melting point (e.g. Zr), however low mutual solubility of Np and Zr – Hydriding & de-hydriding behaviour of TRU-Zr fuel as well as its sintering & hot-pressing behaviour to make uniform pellets – Modelling of PCI	
	Fabrication technology	– Vibro-packing or pellet – Maybe need to transform into metal for dry reprocessing	^{15}N enrichment process and recovery needed	– Injection casting – Centrifugal casting	
			Choice of cladding material: its interaction with coolant (LBE) needs especially to be studied		
	Shielding	High neutron source strength demands additional shielding, criticality control, reduced batch sizes			
		High MA loading in fuel; problems in handling Am and especially Cm			
Waste management		High α-contaminated waste	Pyro-reprocessing waste – α-damage to composite glass-ceramic wastes (sodalite) to be studied – Leaching behaviour of pyrochemical ceramic waste in reducing environments to be studied		
		Reduce secondary waste arisings			
		Trade-offs between waste volume and waste form parameters, associated R&D costs, and disposal costs. Various disposal and disposition options for these wastes need to be analysed			
		A complete performance study, using appropriate source terms, should be performed to assess the cost-benefit aspects for the geological disposal site			

Table 7.1. **Outline R&D matrix for P&T technology (cont'd)**

Technological objectives		(2) Pu-burning	(4) Double strata	(3b) TRU burning in ADS	(3a) TRU burning in FR (5) All FR
Reactor (Safety and O&M)		100% LWR-MOX core (design changes needed)			
		Economy in FR needed because of high fraction in nuclear park			
		Exploration of possibility to re-use radioactive sodium in new fast reactors			
	Sub-critical part				
	Thermal hydraulics		Thermo-hydraulics of liquid metals, especially LBE: Assessment of different coolants, make and compare different engineering designs for an ADS, gas- or LBE-cooled		
	Neutronics		Validation of simulation codes and nuclear data for new fuel, new reactor designs and new burnups per batch. MC and deterministic calculation codes to analyse kinetics and core dynamics		
	Target		– Thermal shocks on materials and fuels due to beam variations – Radiation damage to structural materials: (currently limited to 20 μA/cm^2; should be increased for more flexibility in design)		
			– Investigation of the effects of spallation products on the structural materials of an ADS target		
			– Experimental feasibility demonstration of LM-cooled target		
			– Blanket design studies to understand trade-offs between characteristics of beam delivery and blanket – Coolant chemistry: LBE conditioning, cover gas control, impact of spallation products		
			Window: – Dynamics analysis of diffusion and migration mechanisms for chemical species as functions of the temperature – Out of the beam axis, most interactions are with neutrons and generate more dpa than gas release		
	Basic science needs	Basic nuclear data: need for better cross-section values for MAs in thermal and epithermal spectra			
			Better nuclear data for target/ADS – Spallation product yields and distribution in (A,Z) * Intermediate energy data to meet needs of accuracy * For transmutation: cross-section measurements needed in the 1ev-250MeV range (capture & fission for Ac; capture for LLFPs) – For ADS engineering design: cross-section measurements above 100 keV needed (inelastic collisions, (n,xn) for structural, coolant and fuel materials but also (p,x) reactions)		
			Data are needed on the relatively high solubilities of Pu and Zr in Pb and Bi		
	Safety analysis and demonstration		– Safe shut-down – Decay heat removal – Containment of radioactivity		

- The *fabrication of very specialised, dedicated, fuels* needs further research. Several new aspects come into play. While the fuel form must be compatible with the reprocessing scheme, its selection also depends on the constraints imposed by interactions between pellet and cladding, cladding and coolant, etc. While oxide fuel forms may be preferable in the short term, others such as nitride and metal may be more suitable in the longer term but need significant development during the coming 10 to 20 years. In particular the presence of large amounts of curium is a new challenge for fuel fabrication and may need completely new designs of plants. In case of nitride fuel, specific processes are also needed in order to recover the costly enriched ^{15}N. Besides development of the fuel form itself, attention should be paid to the fabrication process, i.e. the need for dedicated fabrication plants, increased requirements for shielding and criticality control, optimisation of processes for small batch sizes, possible co-location of fabrication and reprocessing plants at the FR or ADS site, and so on. These considerations may need, after an initial concept screening exercise, a detailed assessment before further specific R&D can be undertaken.

- *R&D related to the reactor*, whether FR or ADS. The development and demonstration of FR or ADS technology will require several technological challenges to be overcome:
  - *Lifetimes of proposed materials and components in the radiation, thermal, and chemical environments anticipated*. Understanding the behaviour of fuel and structural materials in complex and aggressive environments is a prime R&D activity before any other development can be considered. In addition, R&D in this domain is expensive in time and resources as corrosion tests and irradiation tests should be performed in experimental rigs or facilities that may still need to be constructed. In the case of ADS, such material irradiation tests should be undertaken in existing fast MTRs (i.e. Monju, FFTF, Phénix, BOR-60, … etc). It is therefore essential that these facilities remain available and so irradiation programmes are proposed to keep them in use. At a later stage, especially when more detailed ADS designs are emerging, additional irradiation tests may be needed in dedicated ADS irradiation test facilities and set-ups (e.g. ADTF, XADS, …), for instance, to test window materials in complex geometries. The challenges posed by materials development are particularly harsh for FR or ADS transmuters, where very high neutron fluxes, liquid metals, and high temperatures may co-exist. This will have a significant influence on design evaluations relative to system life, requirements for maintaining or replacing equipment, licensing, and life-cycle costs. On top of this, the primary and secondary proton damage to these fuel and structural materials in an ADS is an additional and essentially new domain that has only limited resemblance to the proton damage experienced in existing spallation target sources for neutron physics research.
  - *Reliability and availability of ADS systems*: ADS systems may be expected to operate with high availability for 60 years. All ADS subsystems would consist of newly designed equipment operating at higher temperature or higher loads than current equivalents. New equipment and components must be designed and tested to assure lifetime reliability and availability. In addition to the reliability and technological feasibility of several parts of an ADS, more important technological trade-offs may need to be made, e.g.:
    - *Core physics*: recent neutronic benchmarks by OECD/NEA [188] indicate that the modelling of such sub-critical systems needs further development as significant differences still exist in static and especially in dynamic responses. Basic science requirements in this area relate to the need for better cross-section libraries extended to minor actinides and higher intermediate energies. Besides these data,

continuous benchmarking of calculational tools is needed in order to reduce the margins of uncertainty in any future design work.

- *Accelerator type and power level*: the technological feasibility of high-power proton accelerators needs further investigation, especially in relation to the desired reliability. Depending on the specific ADS design and depending on its use as MA or TRU burner, a variable beam power may be needed in order to compensate for burn-up,control the power level, and so on. These requirements are specifically relevant in the case of a TRU burner where the decline in k_{eff} could be very significant and where a trade-off is needed between fuel core management and accelerator operational characteristics.

- *Accelerator beam trips*: as was mentioned in Chapters 5 and 6, proton beam trips occur frequently on current linear accelerators. They would cause thermal stresses in fuel and structural materials, and if frequent, cause low cycle fatigue failure of the structures to become a safety and operating issue. R&D is therefore necessary in order to reduce the impact of these beam trips on the core by reducing their number or by designing a "forgiving" core.

- *Cooling and decay heat removal:* the proposed use of relatively new type of coolants (lead, LBE or gas), especially in combination with new fuel types and cladding materials, demands specific attention to the thermal-hydraulics and core mechanics in normal and abnormal conditions. Sodium cooling is another option that might benefit from the existing rather extensive experience base. Again, a trade-off is needed between the level of sub-criticality, accelerator characteristics and the core management and its impact on the thermal-hydraulics. Safety authorities will demand structural integrity of fuel, cladding and core as a whole while the very high residual decay heat of the fuel may add serious constraints to the cooling needed during handling, transport and core refuelling. Next to the cooling considerations for the core, specific attention is needed to the target region and its integration with the reactor core. Basic science requirements relate to developing and benchmarking thermal-hydraulic codes (with coupling to neutronic codes), studying the compatibility of coolant and its operational regime with the fuel and cladding constraints, and to understanding corrosion mechanisms in such coolant and material combinations.

- *Safety analysis of ADS* should identify the possible ways to exclude HCDAs in ADS. If such a HCDA has to be taken into account in the safety analysis, a prompt negative feedback mechanism for quenching such an accident has to be developed. The technology questions related to the integration of a target with the sub-critical core as well as their safety implications need more precise study, for instance of the dynamic response in case of beam-tube flooding.

- *Instrumentation:* New techniques and tools must be developed to control the coolant chemistry, the sub-criticality level monitoring (by beam power or moving control rods), the coupling between power level and accelerator beam power, etc., and especially to increase the reliability of the accelerator.

- *In-service inspection and repair:* Besides instrumentation, providing operating and maintenance tools suited to highly radioactive environments, mostly not visually transparent and in contact with hot fuel, presents a technological challenge already very familiar from the case of sodium-cooling.

7.3 Perceived R&D needs in the short to medium-term

The above description itself shows the need for continuous development of basic science and technology in essentially four domains:

- *Nuclear data, neutronic calculations and kinetic and dynamic core behaviour* for better modelling of core physics, safety, radiation shielding and so on in order to reduce the design uncertainties.
- *Materials research for fuels and structural materials* (cladding, window, etc.) in various coolants and in radiation fields (including protons) where a phased approach may be appropriate.
- *Reprocessing technology*, aqueous as well as pyro-reprocessing technology.
- *Performance assessment* for a geological disposal site using a P&T source term as is necessary in order to clarify the cost-benefit analysis of a P&T scenario including this geological disposal.

While the above domains may need to be approached differently in various countries (for instance, some countries need more technology transfer on pyrochemistry than others), these basic science requirements are recognised universally as necessary steps to be taken in order to perform detailed system studies. Other R&D requirements in more

7.3.1 Nuclear data and neutronic calculations

Neutron cross-section data is available mainly for uranium and plutonium isotopes, reflecting the interest in the U-Pu fuel cycle, and for neutron energies ranging from thermal to fast reflecting the interest in thermal and fast neutron reactors. Although the currently existing nuclear databases are sufficient for a first evaluation of dedicated transmutation ADS and critical reactors, a detailed assessment requires more precise and complete basic nuclear data.

The first point to take into account is the large fraction of the minor actinides and high mass plutonium isotopes in the fuels proposed for the transmutation devices. These isotopes with little relevance for the operation of present reactors will play an important role on the neutronics of the transmuters. Second, the use of innovative coolants like lead will also make to increase the role of the isotopes contained in that material (mainly lead and bismuth). Third, the operation of many transmutation devices in fast spectrum requires to complete the nuclear data (cross-sections, fission yields, isomer production, etc.) for many fuel and structural material isotopes in the region from 1 keV till several MeV. Finally, the eventual transmutation of fission fragments will require a better determination of the associated transmutation (normally capture) cross-sections.

Present knowledge of the spallation reaction mechanism is not yet accurate enough for any technical application at the scale of the anticipated ADS for transmutation. Two main aspects play a major role in designing and constructing the target assembly of the spallation neutron source: the neutron yield (with its energy and spatial distribution) and the residual nuclei produced in the reaction.

The work on basic nuclear data is largely driven by a few national laboratories that have started extensive programmes on various accelerator-driven projects. However, there is also a more widespread effort to determine data related to transmutation. One may distinguish between conventional nuclear data, below 20 MeV, and intermediate energy nuclear data, above 20 MeV. The

first set is necessary for fast reactor cores, i.e. fuel and structural materials, while the intermediate energy data are essentially needed for the structural materials in ADS applications. In general, the initial focus is on assessing priorities, including sensitivity and uncertainty analysis, and on evaluating key data which may not have received sufficient attention in the past. The actual provision of intermediate energy data to the users is progressing rapidly. Although present uncertainties may allow reasonable pre-conceptual design assessments, future detailed studies will require more accurate data, with drastically reduced uncertainties. The relevant sensitivity studies have started, but they have not yet tackled satisfactorily the problem of accuracy in the intermediate (i.e. 20 MeV ≤ E ≤ 200 MeV) energy range. However, sensitivity studies for the different parts of the whole device are indispensable as a parallel area of research in the field of intermediate energy nuclear data. They may provide a valuable guideline on isotopes and reactions to be measured and evaluated.

Several high-energy transport codes exist containing an intra-nuclear cascade model that is mainly successful for the primary stages of the reaction, including the production of several types of hadron. For energies below about 150 MeV, when the predictive power of the high-energy transport codes becomes suspect, nuclear data libraries are required.

The general recommendation is that for more materials, the available data libraries should be extended from thermal and intermediate energies to 150 MeV. This will require a significant effort from both the experimental and the theoretical nuclear physics communities. It is therefore necessary to revisit the evaluation process over the whole energy range, which should ensure a smooth transition from low to high energies. Some new experiments are proposed or under way in order to fulfil this task.

Japan

JNC has organised, together with Japanese universities, several projects on nuclear data measurements for LLFPs and MAs over recent years. These researches include for instance fast neutron induced fission cross-sections of americium isotopes, and capture cross-sections of ^{237}Np, ^{99}Tc among others [189]. JNC is now planning to extend the nuclear data measurements to capture and fission cross-sections and decay data for important LLFPs and MAs from the thermal energy region up to a few MeV. In particular, more precise determinations of the capture cross-section in nuclides such as ^{99}Tc and ^{129}I are intended.

EC[70]

A large experimental programme was initiated in Europe a few years ago in order to improve our knowledge of the spallation reactions. These experiments are set-up in order to provide accurate data to benchmark more reliable model calculations. Neutron multiplicities were investigated using liquid-scintillator-based detectors with a large angular acceptance in the Berlin Neutron Ball [190] and ORION [191] used by the NESSI collaboration (Berlin – Ganil – Jülich). This collaboration has conducted a large experimental programme to determine the neutron yields produced in thin and thick targets by a large range of primary projectiles and energies. In addition, the unique and essential GSI experiments derived the (A,Z) distributions of spallation products by using the inverse kinematic technique. Spatial and energy distributions of spallation neutrons were measured at Saturne [192].

70. A more complete overview of the EC's programme of work is given in [193].

New experiments have been proposed and are to be conducted within the EC 5th Framework Programme within the n-TOF and HINDAS projects.

n-TOF

The main goal of the n-TOF project is to produce, evaluate and disseminate high precision cross-sections for the majority of the isotopes relevant to waste incineration and design of the ADS, i.e. capture and fission cross-sections for the MAs, capture cross-sections for the main FPs and (n,xn) reactions for structural and coolant materials. Most of the measurements will be performed using the CERN (Geneva) accelerator complex. The experiment will cover low as well as intermediate energies, 1 eV – 250 MeV.

HINDAS

In the HINDAS project (sponsored by EC), nuclear data in the 20-2 000 MeV region will be provided by a combination of nuclear models and appropriate intermediate- and high-energy experiments. A whole panoply of European accelerators will be utilised to provide complete sets of experimental data for key elements and energies. Nuclear model codes will be improved and validated by these new experimental data and then used to generate enhanced ENDF-formatted data libraries below 200 MeV, and cross-sections for high-energy transport codes above 200 MeV. The impact of new data libraries and high-energy models will be directly tested on some important parameters of an accelerator-driven system.

USA

Neutron total cross-sections have been measured at LANSCE from 5 to 560 MeV on 31 elements and isotopes covering the range from hydrogen to uranium. These measurements were supported by the APT project as part of a programme to improve the physics in the modelling codes for neutron transport up to several hundred MeV. For nearly all of the target materials, the data are accurate to better than 1% (both statistical and systematic) in 1% neutron energy bins. These data are essential for neutron transport codes and for nuclear modelling.

7.3.1.1 Spallation product analysis

Besides developing better cross-section libraries for fuel and structural materials, other required nuclear data relate to spallation products in the target and coolant material. These data are very important in material selection to improve, for instance, the target window lifetime, the arisings of secondary wastes in the target area, etc. During the irradiation of an ADS target with protons a large number of spallation products are formed. Most have short lifetimes and no significant impact on the behaviour of the system. However, a considerable number of spallation products or their successors have longer lifetimes and must be taken into account with respect especially to consequences for waste arisings and thus for the back-end of the system. Recently a Russian study has been published with a comparison of long-lived residual activity characteristics of liquid metal coolants for advanced nuclear energy systems [153]. This study emphasises the importance of ^{205}Pb and of ^{210m}Bi with lead and LBE coolants respectively.

The significance of spallation products in a fast spectrum ADS has been investigated in some detail. It has been shown that in a fast spectrum, neutron absorption in the spallation products competes only weakly with nuclear decay as long as the half-life is less than about 30 days. The most important spallation products have been listed in IABAT and show that quite a large number of isotopes are missing from the evaluated data files, e.g. ^{205}Pb. Some of the most outstanding experiments to measure residue production are performed by the German-Spanish-French collaboration at GSI. The technique used in these experiments takes advantage of inverse kinematics and the full identification in mass and atomic number of the reaction residues by using a magnetic spectrometer [194].

7.3.1.2 Neutronics

The further development of calculational tools is as important as the previous action on basic nuclear data and the experimental benchmarking of these codes is especially necessary. Calculational benchmarks have been undertaken at laboratory and international levels, co-ordinated by OECD/NEA or IAEA. Experimental benchmarks are ongoing where one of the most important experiments is performed in CEA Cadarache.

The MUSE experiments

The MUSE experiments, launched in 1995, simulate the neutronics of a source-driven sub-critical system, using the physics characteristics of the separation of the effects due to the presence of an external neutron source from the effects of the neutron multiplication. In fact, for a wide range of sub-criticality values (e.g. k_{eff} = 0.9 - 0.99), the space dependence of the energy distribution of the source neutrons is quickly (in approximately one mean free path) replaced by the fission-dominated neutron energy distribution.

In practice, external known neutron sources have been introduced at the centre of a sub-critical configuration in the MASURCA reactor. The more recent of these experiments is made of a deuton accelerator and a target (deuterium or tritium) at the centre of a configuration where actual target materials (like lead) are loaded in a buffer close to the target, to provide the neutron diffusion representative of an actual spallation source. The neutrons issued from (d,d) and (d,t) reactions, after crossing the lead buffer, provide a reasonable simulation of the spallation neutrons, in terms of energy distribution. Static (e.g. flux distributions, spectrum indexes, importance of source neutrons) and kinetic parameters (e.g. time dependence of neutron population, effective delayed neutron fraction, with appropriate weighting, etc.) have been or will be measured. Sub-criticality itself is measured by static and dynamic techniques. Continuous monitoring of sub-critical reactivity in future accelerator-driven systems will become important. Several core monitoring techniques, including noise related techniques (Feynman-α), would also be tested in this MUSE-experiment.

Finally, the proposed experiment MUSE-4 start-up procedure with:

- A critical configuration with accelerator hole but no beam.
- A sub-critical configuration with accelerator hole but no beam.
- The same, but with beam on.

allows a precise reactivity scale to be established in Step 1, and then used both to calibrate control rods if needed and to measure in a standard way (e.g. with the modified source multiplication, MSM, method) the level of sub-criticality in Steps 2 and 3.

The TRADE experiment

The present basic experiments do not provide the validation of the concept at low but significant (~1 MW) power and with the coupling of an accelerator with a sub-critical core. On the contrary, to demonstrate the feasibility of stable operation and dynamic behaviour as well as to investigate the safety issues of an ADS, it is of primary importance to perform a first global experiment to demonstrate the coupling a proton accelerator with a spallation target and a sub-critical system of sufficient size to produce a sizable power. Moreover, operational experience in this domain, along with the definition of licensing procedures for such a system, could be extremely beneficial to the realisation of a future fast neutron demonstrator.

A joint ENEA-CEA working group has recently launched the idea to carry out this pilot experiment, first example of ADS component coupling "at real size", in the TRIGA reactor at the ENEA-Casaccia Centre. This reactor is an existing swimming pool reactor of 1 MW thermal power, cooled by natural convection of water in the reactor pool. The TRIGA reactor, made sub-critical by removing the innermost ring of the fuel core, will be coupled with an upgraded commercial proton cyclotron (proton energy of 110 MeV and current in the range 0.5-2 mA) through a tungsten solid target

This experiment – actually called TRADE, i.e. TRIGA Accelerator-driven Experiment, could be performed at levels of several hundred kW sub-critical core power and few tens of kW in the target, thus providing, among other, valuable insight into the dynamic behaviour of an ADS in presence of reactivity feedback effects.

The experiments of relevance to ADS development to be carried out in TRIGA could concern:

- The dynamic regime: the possibility to operate at some hundred kW of power and at different sub-criticality levels (0.95÷0.99) will allow to validate experimentally the dynamic system behaviour versus the external source effectiveness and to obtain important information on the optimal sub-criticality level both for a demonstrator and, by extrapolation, a transmuter.
- Sub-criticality measurements at significant power.
- Correlation between reactor power and proton current. This correlation can be studied at different sub-criticality and power levels.
- Reactivity control by different means and possibly by neutron source importance variation, keeping the proton current constant. In principle, this can be obtained changing the neutron diffusion proprieties of the buffer medium around the spallation source (e.g. using different materials in the empty innermost fuel ring close to the target).
- Start-up and shut-down procedures, including suitable techniques and instrumentation.

Moreover, an important feature of the TRIGA layout is the possibility to carry out, before the coupling with the accelerator, a propaedeutic experimental campaign by inserting into the current central thimble a known standard fixed neutron source, and performing static and dynamic measurements for different sub-criticality levels. For example, such a configuration of the TRIGA reactor allows the application of a wide variety of techniques devoted to the determination of the sub-criticality level, like source jerk, pulsed neutron source analyses, rod drop, Modified Source Multiplication. Such set of experiments will provide a link with the MUSE experiments and characterise the sub-critical core from a safety point of view.

The preliminary studies have been completed and a first feasibility report has been issued on June 2001; analysis have been performed on neutronics, power and temperature distributions, structures damage, thermal-hydraulic of the target, safety parameters, general lay-out of the facility and some licensing issues.

The preliminary analysis and results show no major obstacles, even if some more detailed studies should be performed. The feasibility report shows that all relevant experiments (at different power levels in a wide range of sub-criticalities) can be performed, with only relatively limited modifications to the existing TRIGA reactor.

7.3.2 Materials research

Materials research may be subdivided into three domains, i.e. fuel, structural materials including cladding and window materials, and the coolant. It should be remarked that these are

mutually interacting and a cross-disciplinary approach is needed, to cover also the reprocessing technologies for fuel and cladding research.

7.3.2.1 Fuel research

For both critical and sub-critical dedicated cores, the major issue in the path towards feasibility demonstration is the development of fuel. Many candidates have been considered (see for example Table 7.2), but limited experimental work has been done, in order to characterise the basic properties of these potential fuels, their fabrication processes and their behaviour under irradiation. See also Chapter 3, Section 3.4.1. for a more complete discussion of fuel and target fabrication and behaviour and the interaction with their reprocessability.

Practically all the major transmutation programmes lack well-structured coverage of fuel development. A significant exception is the JAERI programme, focused on nitride fuels.

Table 7.2. **Dedicated Pu + MA fuels**

Metal fuels	– Need to improve thermal properties ⇒ add non-fissile metal with high melting point (e.g. Zr) ⇒ Pu-MA-Zr alloy – However: mutual solubility of Np and Zr may be troublesome
Oxide fuels	– Mixed transmutation oxides as a logical extension of MOX – However: smaller margin to melting (low thermal conductivity)
Nitride fuels	– Good thermal behaviour – However: need enrichment in ^{15}N – Lower stability against decomposition at high temperatures
Composite fuels: the role of Zr	Ad-hoc "tailoring": – MgO + (Zr, An)O_{2-X} (CERAMIC-CERAMIC) – Zr + (Zr, An)O_{2-X} (CERAMIC-METALLIC) – Zr + (An, Zr) alloy (METAL-METAL) However, fabrication can be difficult (also: size and distribution of the dispersed actinide phase)
Coated particle fuels	Special form of composite fuels. However in the case of fast spectra, little is known on potential candidates (TiN?) ⇒ A generic problem: the high production of helium under irradiation.

Irradiation performance issues

The primary performance criteria for a fuel rod in general are that for all anticipated conditions the fuel should retain its position in the core, contain fission products and maintain a coolable geometry. Experience with various fuel forms has proved the following characteristics to be important to fuel performance and lifetime:

- *Dimensional stability*: the dimensions of the fuel can change dramatically if the material swells or grows significantly on irradiation. Such effects have implications for the neutronic performance of the core and can introduce stresses into the cladding that lead to a breach.
- *Fission gas pressurisation*: some fuel types, especially metallic, release large amounts of fission gas into the fuel rod plenum. Therefore, if the plenum is not adequately sized to

accommodate this fission gas content, then pressure-induced stresses can lead to cladding failure, particularly during transient-induced temperature increases.

- *Phase stability or micro structural evolution*: the high-temperature, high-flux environment of the reactor core (which also induces temperature gradients) typically alters the microstructure or local composition of a fuel material from its initial state. Such changes include development of porosity that can vary in morphology across the radius of a fuel rod, establishment of different phases and redistribution of fuel constituents through the fuel material. Accumulation of fission products during irradiation can also contribute to these effects. Ultimately, they manifest themselves by degrading thermal conductivity, by introducing local high-power zones in the fuel, by enhancing chemical interaction between fuel and cladding, or possibly by affecting gas release or swelling phenomena.

For many fuel forms, contact of the fuel or fission products with the interior cladding surface leads to interactions that embrittle the cladding or otherwise compromise its ability to withstand stresses. This is particularly true for some metal alloys, in which fuel constituents and fission products interdiffuse with cladding constituents, resulting ineffective thinning of the cladding and incorporation of low-melting phases or compositions in the fuel, cladding or the interaction zone at the fuel-cladding interface; formation of such zones often has implications for fuel reliability during high-burn-up, steady-state operation and during certain transients. Experience shows that fuel performance almost always degrades as the plutonium content of the fuel is increased. The proposed fuels employ an actinide alloy that is primarily plutonium. Furthermore, the incorporation of such a large quantity of minor actinides into the fuel alloy has never before been tested.

A major complication to the irradiation test programme is the lack of an appropriate test reactor in which to conduct the irradiations. Furthermore, no LBE test loop exists at any irradiation facility in the world. However, it should be remarked that more intensive research should be conducted towards simulating materials behaviour under irradiation conditions in order to shorten the necessary irradiation campaigns. Such a modelling of materials behaviour, under proton and neutron irradiations, would also permit a better focusof the experiments in the scarce and expensive irradiation devices.

Compatibility between the fuel and stainless steel cladding must be confirmed. Considerable data exist in this area for metallic, plutonium-containing EBR-II or IFR fuels and a variety of stainless steel claddings. Of particular interest will be the class of stainless steel alloys in use by the Russians in LBE applications, and the effect of the MAs on compatibility. For the dispersion fuel form, compatibility between the fuel alloy and the zirconium matrix must be demonstrated. Additionally, compatibility between the fuel and the LBE must be characterised. Although little data currently exists in this area, the relatively high solubilities of plutonium and zirconium in lead and bismuth indicate the need to consider dissolution of fuel material into coolant after a breach of cladding. The major issues in this area must be resolved before beginning the irradiation test programme.

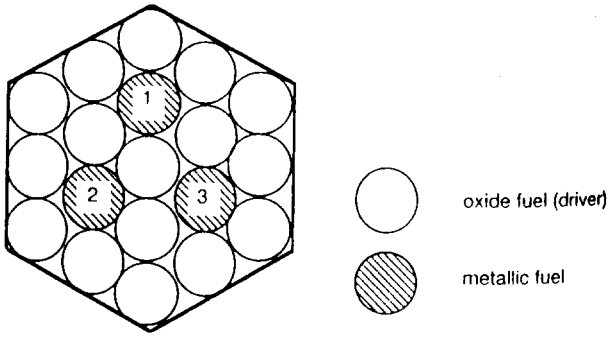
A number of issues related to the fuel-cladding gap must be resolved before fabricating the initial fuel for irradiation testing. For the primary dispersion fuel form, it must be determined whether fabrication techniques allow for co-extrusion of the fuel and cladding, thus eliminating the need for an open gap and a thermal bond material. Should these be required or desired, a thermal bond material must be selected that is compatible with the fuel, cladding and coolant; this material will probably be a liquid metal, such as LBE or sodium.

While many material properties may be conservatively estimated for the purposes of obtaining approval for the initial irradiation experiments in test reactors, this will not be enough to prepare a sound technical safety case for an ADS core. This is because the material properties may be very

conservatively estimated for the purposes of irradiation testing, relying on the fact that only a small amount of the test fuel is being introduced into the reactor core, thus limiting the consequences of any failure to an acceptable level. Such consequences are generally not acceptable when failure is extended to a significant fraction of the core. Thus, a fairly comprehensive experimental programme to measure the important fuel properties directly must reduce the large uncertainties associated with the conservative estimates of material properties.

Besides the developments in the USA that could be re-started in the context of the AAA-programme, other countries and international laboratories have undertaken comparable experiments. Activities started at CRIEPI (Japan) and have now extended to TUI-Karlsruhe. These new activities also cover fuel reprocessing. In particular, an experiment (METAPHIX) is planned, in order to irradiate metal fuel pins, loaded with MA and rare earths (RE).

Figure 7.1. **Arrangement of fuel pins in a rig for the METAPHIX experiment (CRIEPI-TUI)**



Nine metallic fuel pins have been prepared for the METAPHIX irradiation study: three pins of UPuZr, three pins of UPuZr-MA2%-RE2%, three pins of UPuZr-MA5%, and UPuZr-MA5%-RE5%. They are planned to be inserted in the positions 1, 2 and 3, respectively, in the rig. Three rigs consisting of three sample metallic fuel pins and sixteen driver oxide pins will be prepared, one for each of the burn-up values 1.5, 5 and >10%.

On homogeneous recycling in standard oxide fuels, some experimental knowledge has been obtained with the SUPERFACT experiment. More will come from experimental programmes conceived at JNC to take place in JOYO beyond 2003. However, no experience exists on MA-loaded oxide fuels in standard light water reactors.

In the context of a heterogeneous recycling mode, apart from conceptual studies at JNC and CEA, experimental activities have been launched in Europe (e.g. the EFTTRA collaboration) and some useful indications have been gathered. The EFTTRA-T4 and T4-bis experiments concern ^{241}Am, at a 12% volume fraction, in a matrix of $MgAl_2O_4$, for a maximum fission rate of 28%. Swelling due to the decay of the ^{242}Cm produced by neutron capture has been significant, and triggered further research on the form of inert matrix-actinide fabrication (e.g. micro-dispersion versus macro-dispersion). It is worth noting that experiments performed up to now did not cover the presence of ^{243}Am and curium. Further experiments are planned in France, and in particular the ECRIX experiment, which should take place in PHENIX, and the CAMIX and COCHIX experiments also planned in PHENIX. The CAMIX experiment will provide information on "micro-dispersion" of a $(Am, Zr, Y)O_{2-x}$ compound in MgO, and COCHIX information on the same compound "macro-dispersed" in MgO or $(Zr_{0.6}\ Y_{0.4})O_{1.8}$. All three experiments are planned to reach a fission rate equivalent to 30 at%.

A significant global experiment is presently planned in the framework of the collaboration between MINATOM (Russia) and CEA (France) with FZK and TUI-Karlsruhe, as partners of CEA. In this experiment (AMBOINE), americium targets of $AmO_2 + UO_2$ and $AmO_2 + MgO$ will be fabricated at RIAR by the VIPAC process. These targets should be irradiated in BOR-60 and reprocessed by pyro-processing after further irradiation at RIAR, so providing a full validation of the whole fabrication – irradiation – reprocessing cycle for CER-CER targets.

The CONFIRM project, sponsored by the EU 5th FWP, is devoted to nitride fuels ((Pu,Zr)N and (Am,Zr)N). The project aims to fabricate, characterise and irradiate these fuels, and addresses also the issue of ^{15}N enrichment.

7.3.2.2 Structural materials research

Besides issues related to selecting an appropriate cladding material, materials research is focused on the choice of window material for the target of an ADS. Regardless of the technology chosen, the major research and development activities that are required for the successful development and demonstration of a target include:

- Establishing requirements for design data (material properties such as strength, ductility and fracture toughness as a function of displacement damage, in-beam and out-of-beam corrosion resistance, swelling tendencies, etc.).
- Performing irradiation and corrosion tests to provide the design data.
- Conducting irradiation tests on near-prototypical structures and components designed, fabricated and irradiated to simulate anticipated service situations.
- Post irradiation examination and analysis of test samples, structures and components.
- Acquiring spallation physics data and developing methods to predict yields of spallation neutrons and products.

Perhaps the most significant issue with regard to the target design, and certain the most costly to address, is the change in materials properties due to irradiation (proton and neutron) and exposure to LBE coolant. Iron, chromium and nickel, components of many structural steels, are soluble in liquid LBE and experience corrosion or erosion when exposed to streams of it. However, compatibility with the liquid LBE can be significantly improved by adding silicon to steel and controlling the oxygen content in the LBE alloy. Using silicon modified steels and oxygen control measures apparently provide the basis for successful application of LBE coolant technologies to several Russian nuclear systems, including Alpha-class nuclear submarines. There is little or no data on the effects of high-energy proton beams and spallation neutrons on materials properties at the temperatures of interest to ADS. Therefore, an irradiation test programme is necessary to quantify the effects of displacement damage, hydrogen and helium build-up and the accumulation of spallation products on material properties. Recent studies of candidate materials for the APT, irradiated and tested at or near 130°C, have shown that fission reactor data do not accurately represent the effects of irradiation by high-energy particle beams. These studies also confirmed the accumulation of high levels of hydrogen and helium in the irradiated materials. The temperatures anticipated for the LBE coolant in ADS are in the range where helium embrittlement of steels is well documented. These observations illustrate the necessity for irradiation testing under anticipated ADS temperatures and proton or neutron spectra.

Target window lifetime[71]

The window of the spallation target is a critical part of this device. It is directly exposed to the proton beam, it must be transparent to protons so it cannot be thick, and being an interface between accelerator and reactor environment, it will most probably be an interface between low and normal or high pressure. Moreover, it will also be exposed to back-scattered spallation neutrons and neutrons coming from the surrounding sub-critical core.

The following Table 7.3 presents the main parameters applied to assessing the lifetime of the window for each of the target types, using lead, mercury or tungsten plates for a 1 GeV beam. This table shows that lead and mercury targets suffer similar damage to the window; however, the mercury and tungsten targets have been designed for a higher power. Interstitial atoms of phosphorus and sulphur migrate towards grain boundaries at high temperatures and might cause local embrittlement. These phenomena still require more investigation from the materials standpoint and should include dynamic analysis of diffusion and migration mechanisms for chemical species as function of the temperature. The lifetime of the window is essentially determined by its mechanical properties. Here, two arbitrary limits in the deterioration of the material are used: maximum helium production (2 000 appm He corresponds to the dose accumulated in the Inconel window tested at LANL) and maximum dpa (100 dpa is half the objective for FBRs in order to be conservative). The second Table 7.4 indicates the lifetime of the window in a beam current of 33 mA for the different targets.

Table 7.3. **Factors affecting the lifetime of windows**

	Pb	**Hg**	**W plates**	**Units**
Neutron yield	28.2	24.6	15.5	n/p
Gas release				
H,D,T	740	790	680	appmH/mA.y
^{3}He, ^{4}He	40	43	39	appmHe/mA.y
Interstitials				
Phosphorus	6	5	5	appmP/mA.y
Sulphur	4	3	4	appmS/mA.y
dpa	0.92	0.94	0.68	dpa/mA.y

Table 7.4. **Limiting lifetime of windows with various target materials**

Limiting factor	**Liquid Pb**	**Hg**	**W**	**Units**
^{3}He, ^{4}He	1.5	1.4	1.6	Years
dpa	3.3	3.2	4.5	Years

The previous tables do not include “reactor range” neutrons and as these account for more than 50% of the overall damage, they should clearly be taken into account.

Radiation damage to the window ofthe spallation target and enclosure walls

Table 7.5 gives an evaluation of the damage by high-energy particles in the main system components as well as of gas production by spallation particles in the main target components. We

71. See also the description of spallation target technology in Chapter 4, Section 4.3.3.

allow for 4.6 nuclear interactions per incident proton, 0.5% of which take place in the window, 50% in the liquid target, 37% in the core and 12.5% in axial and radial shielding. An upper neutron shield is twice as much exposed (4%) as a lower shield.

Table 7.5. **Effects of high-energy particles on system components**

Component	**H, D, T**			**He**		
	appm/mA.day	appm/mA.y	appm/ dpa	appm/mA.day	appm/mA.y	appm/ dpa
Beam tube	1.165	425.52	644.73	0.032	11.7	17.27
Core	0.017	6.21	34.5	0.003	1.1	6.11
Dummy belt	0.56	204.54	401.06	0.022	8	15.69
Radiation damages to ADS components induced by high-energy particles						
dpa/mA.y	Core	Beam tube	Window #2	Dummy test belt		
	0.18	0.66	20.93	0.51		

The analysis shows that off the beam axis, most interactions come from neutrons. These interactions generate more dpa than gas release, as indicated in the previous table.

In Europe, the Fifth Framework programme has included a specific programme, entitled SPIRE, to investigate irradiation damage on such structural materials. This programme addresses the effects of:

- Spallation elements on microstructure and mechanical properties before irradiation.
- Spallation elements on microstructure and mechanical properties via ion implantation.
- Fast neutron irradiation on mechanical properties to complement existing data.
- Irradiation in a mixed neutron-proton prototypical spectrum on mechanical properties and microstructure. In addition, modelling is dedicated to characterising irradiation damage, predicting hardening and computing segregated boundary cohesion energy.

The main outputs are a validated data-base on tensile, Charpy, fracture toughness and the selection of a reference steel, for design studies.

Experiments

TC-1

The TC-1 loop is being designed, fabricated and pre-tested by IPPE, funded by ISTC. This loop is designed to the physical constraints and beam specifications of the LANSCE Area A. The TC-1 will be an in-beam test of LBE as a spallation neutron source target. It will be used to demonstrate operation and control of the LBE target, both in and out of beam; to investigate spallation product accumulation via cover gas sampling and post-irradiation examination of LBE; and to investigatedegradation of component performance with accumulation of spallation products, corrosion and radiation effects. This loop was planned to be installed in-beam in the second half of 2001.

LiSOR experiment

One of the major unknowns in liquid metal target development is related to whether liquid metal – solid metal reactions are enhanced under irradiation in the presence of static or cyclic stress. Since this is a problem that must be solved before a liquid metal target can be irradiated in a proton beam for an extended period of time, an experiment has been initiated to use PSI's 72 MeV cyclotron to irradiate stressed steel specimens in contact with flowing liquid metal. Currently the rig is being designed by SUBATECH with support from CNRS and CEA. LiSoR was originally planned as a stand-alone investigation. Owing to its immediate relevance for MEGAPIE, it is intended to be incorporated into the initiative, but for the time being it is still pursued on an independent basis. This is mainly due to the temporal restrictions which result from PSI's intention to discontinue operation of 72 MeV cyclotron in 2001 and from the time when results are needed to affect the MEGAPIE design. Support for LiSoR is being sought under the first phase of the EU 5th Framework Programme.

MEGAPIE

MEGAPIE is an international experiment undertaken by CEA, PSI, FZK, CNRS France, ENEA, SCK·CEN, US-DOE and JAERI. It is to be carried out in the SINQ target location at the Paul Scherrer Institute in Switzerland and aims at demonstrating the safe operation of a liquid metal target at a beam power in the region of 1 MW. The minimum design service life will be 1 year (6 000 mAh).

The target material will be the LBE mixture. Existing facilities and equipment at PSI will be used to the largest possible extent. In fact, the MEGAPIE target will be used in the existing target block of SINQ.

The target will be designed for 1 MW of beam power at a proton energy of 575 MeV, i.e. a total beam current of i_p = 1.74 mA.

The major objectives of the MEGAPIE initiative are:

- Full feasibility demonstration of a spallation target system.
- Evaluating radiation and damage effects on structures and beam window in a realistic spallation spectrum.
- Testing effectiveness of the window cooling under realistic conditions.
- Investigating interactions between liquid and solid metal under radiation and stress.
- Post irradiation examinations (PIE).
- Demonstration of decommissioning.

Two EU contracts established in the framework of the 5th FWP, SPIRE (material irradiation) and TECLA (physico-chemical properties of lead alloys, corrosion etc.), provide a relevant R&D back-up to the MEGAPIE project. Moreover, experimental laboratories have been launched in support of these activities (like the KALLA laboratory in FZK-Karlsruhe) or re-oriented (like the ENEA laboratory in Brasimone, the CHEOPE loop). In addition, a specific experiment is under way to study the possibility of early embrittlement under irradiation and inthe presence of LBE (the LISOR experiment).

7.3.2.3 Research related to liquid metals, particularly as target materials

The advantages and drawbacks of different coolants have been described in Chapter 4. Some experiments are currently under construction or already performed in order to test basic aspects of coolant behaviour, besides some technological development tests in relation to specific system designs. Moreover, at present most ADS target designs are based on LBE and this is the driving force for most of the R&D programmes indicated below.

Several groups have proposed R&D-programmes in this field and some test loops have been constructed:

Japan

As part of the R&D for the Accelerator Material Irradiation Facility, a liquid LBE loop for material testing was installed in JAERI/Tokai at the end of January 2000. The loop was successfully operated at 450°C with 50°C of temperature difference for more than 1 200 hours. For the safety analysis of the ADS physics experimental facility, a preliminary evaluation of a hypothetical accident showed that the dose rate around the facility can be controlled at a low level by the emergency shutdown mechanism. The groups for the experimental facility design and for research on the transmutation system were merged in April 2000. This new group, named as "Nuclear Transmutation Group", will undertake broad research and development for P&T technology as well as the development of an ADS Experimental Facility.

USA

In 1997-98, a first test loop to develop LBE technology was built at LANL. The objectives were to acquire experience in constructing and operating an LBE system, to collaborate with Russian experts on technology transfer, to implement and demonstrate reliable instrumentation for thermal-hydraulic parameters and to implement control systems for safe operation of the loop. The loop allowed temperatures in the range of 250-400°C and flow velocities of about 5 m/s (36 m^3/h capacity). A new test loop, integrating more Russian technology and experience, was constructed in 2000. This new loop is designed to study material compatibility and thermo-hydraulics. A higher temperature range (350-500°C), higher temperature changes (delta T = 100°C) and the possibility of natural convection makes this new test loop appropriate for experiments on corrosion control and coolant quality maintenance, material compatibility, thermo-hydraulics and heat transfer, equipment performance and natural convection.

EC

KALLA Lab (FzK, Germany)

In Germany, the HGF Strategy Fund Project aims at developing new methods and technologies to design and manufacture thin-walled thermally highly-loaded surfaces (such as the beam window) which are cooled by a corrosive heavy liquid metal (LBE). The results of this project will be the basic scientific-technical tool for the conception and the design of an ADS spallation target and later on a European Demonstrator of an ADS systems. Three fields are covered by this project (see following Table 7.6):

- *Thermal-hydraulic investigations*: if necessary for experimental thermal-hydraulic physical models of conductive and convective heat transfer along thermally highly-loaded surfaces such as a beam window in turbulent LBE flow. In parallel, a thermal-hydraulic computer programme isbeing validated for fluid LBE at low Prandtl numbers. Finally, a complete spallation target isto be numerically designed.
- *Material specific investigations*: using physical methods to define corrosion mechanisms in flowing LBE and ways to overcome their effects on potential structure and window materials, with and without surface treatment.
- *Oxygen control*: in the field of reaction kinetics, a physico-chemical method to measure and control the oxygen potential in a LBE loop is being developed in order to prevent corrosion of the materials used.

Table 7.6. **Experimental investigations performed in the KArlsruhe Lead LAboratory (KALLA)**

Technology loop	Thermal-hydraulic loop	Corrosion loop
Oxygen measurement	Single-effect investigations	Corrosion mechanisms
Oxygen control	Solid beam window	
Measurement techniques	Windowless design	Protective layers
Heat transfer and turbulence	Closed target module	Mechanical tests
High-performance heaters	Fuel element	
	Steam generator	
	Heat exchanger	
	Integral investigations: Core heat removal Decay heat removal	
Fluid volume: 0.1 m^3	Fluid volume: 0.5-4.0 m^3	Fluid volume: 0.03 m^3
Temperature: max. 550°C	Temperature: max. 550°C	Temperature: max. 550°C
Flow rate: max. 5 m^3/h	Power: 0.3-4.0 MW	
	Flow rate: max. 100 m^3/h	Flow rate: max. 3.5 m^3/h

TERM experiments

In order to study some of the unresolved problems related to the design of liquid metal targets in the context of the ESS project and in preparation of a data base for thermal hydraulic studies for a possible later SINQ liquid metal target, a Test Experiment at the Riga Mercury Loop (TERM) was set up. The main goal was to study experimentally questions of heat transfer between the window and the fluid and related flow distributions in various geometrical configurations. The first phase, which used the geometry of the SING target, has been finished. Methods developed and used include Ultrasonic Velocity Probes (UVP), based on a through the wall measurement of the Doppler effect in the fluid, Heat Emitting Temperature Sensitive Surfaces (HETSS) and Surface Thermography. Data from this phase of the experiment are still being evaluated. Ongoing experimental work now concentrates on the geometry of the ESS target and the effect of gas in the fluid on the cooling of the beam window. The full-scale SINQ target model is also still available for further investigations.

PSI LBE loop

In order to be able to carry out experiments even more realistic for SINQ than were possible at the Riga Mercury Loop, an LBE loop has been constructed and is being commissioned at PSI. Without a test section attached, the loop contains 0.12 m^3 of LBE and has a total height of 5.1 m. Operating temperatures are rated at up to 250°C. It is equipped with an EM pump (32-58 kVA) with a head of 1.5 m LBE and a capacity up to 200 l/min. The pressure rating of the loop is 1 to 2 bar. Test sections can be added to the loop depending on the problem under investigations. The loop is intended for testing of individual components as well as studies of flow configuration and heat transfer problems.

CIRCE

ENEA, though its ADS Project Team and in collaboration with Ansaldo, has decided to build CIRCE – a Pool Test Facility based on LBE – carrying out R&D in support to the ongoing ADS design activity. In particular, CIRCE will allow testing the key operating principles of the LBE Experimental Accelerator-Driven System (XADS) currently being designed in Italy. The size, the LBE load of 100 tonnes, the layout, and the basic features of the CIRCE facility have been set to meet the aforementioned R&D needs and utilise the former PEC building structures and components

in Brasimone. The facility has been conceived to feature basically natural and enhanced circulation of LBE in a fairly large pool with a controlled and instrumented environment, electrical heating of the test volume and removal of 1 MW. The wide access to the test pool from the top with dedicated test sections supported by the cover plug makes the facility suitable for different tests: thermal-hydraulic, LBE purification and material compatibility, special instrumentation and remote blind operation in a LBE environment, integral component prototype, and benchmarks for scaled system analysis. Table 7.7 provides the main parameters of the facility.

Table 7.7. **CIRCE Facility Main Parameters**

Parameters	**Value**
Main vessel	
Outside diameter, mm	1 200
Wall thk, mm	15
Height, mm (from bottom head to top flange)	8 500
Material	AISI 316L
LBE inventory, kg (max)	~90 000
Electr. Heat tracing, kW	47
Cooling air flowrate, N-m^3/s	3
Temperature range, °C	200 to 550
Main vessel cover gas pressure	
Operating, kPa (gauge)	15
Design, kPa (gauge)	450
Argon gas	
Flowrate, N-liter/s	15
Injection pressure, kPa (gauge)	600
Electr. heaters (prospective) for core power simulation, MW	1.1

Test results from CIRCE can intersect a wider interest on HLM, the basic features of the CIRCE facility offer flexibility to conceive test on a more general frame of interest related to the HLM use. The tests with the CIRCE facility could give confirmation on: material corrosion in oxygen-controlled eutectic in “pool” configuration, effectiveness of different filtering elements for the Pb-Bi purification, Pb-Bi natural circulation, Pb-Bi enhanced circulation by gas injection system, performance of a secondary loop with low-vapour pressure organic diathermic fluid, overall plant performance and systems interaction during operational and accident conditions, hydraulics of a windowless target eutectic Pb-Bi, kinematic links of the fuel handling machine in cover gas and in the melt, ISI technology, instrumentation operating in Pb-Bi.

The basic configuration of CIRCE, including the first test section, has been completed and commissioned at the site of Brasimone (Italy) in October 2001.

TECLA

This EC 5th FWP-funded programme aims at carrying out several investigations in order to demonstrate the applicability of LBE technology or develop new systems. Moreover, thermal-hydraulic experiments on fundamental topics typical of any ADS system have to be performed in order to define analytical correlations for lead alloys and to validate codes for design. The final goal of this activity is to demonstrate the feasible use of lead or LBE as spallation target and coolant.

Several additional and smaller Pb and Pb-Bi loops had been build in some of the institutions participating in this TECLA program.

VICE

VICE (Vacuum Interface Compatibility Experiment) is intended to answer questions on the direct coupling of an accelerator to a liquid LBE target in a windowless design for the MYRRHA-project. The objectives of VICE are to:

- Clarify the possible interaction of the accelerator, demanding a high vacuum, with material emanating from the LM in the windowless design.
- Qualify and test corrosion protection methods for the loop wall (protective coating or oxygen control).
- Assess initial out-gassing rates of the LM and vessel as a function of temperature and other parameters affecting diffusion and cleanliness of the LM.
- Assess the migration of material towards the accelerator, whether gases or metal vapours under quasi-operational conditions.

7.3.3 Reprocessing research

Chapter 3 introduced the description, and especially the applicability, of the two main categories of process that might be applied to the separation of long-lived radionuclides: hydrochemical and pyrochemical processes. It was indicated that, to avoid radiolysis effects, pyrochemical processes are the better suited to the TRU and MA recycling schemes. This section will therefore briefly review the two processes where some emphasis will be given to the R&D requirements for the pyrochemical processes. Additional information with extensive descriptions of these processes and the R&D programmes is given in the literature [8,9,2,195].

Numerous concepts have been consolidated or newly developed during the last few years, both in hydrochemical and pyrochemical processing of HLWs or spent fuels and targets for advanced nuclear systems. Tests on “real objects” were carried out successfully in several countries, including the EBR II demonstration test at Argonne-West (USA) on pyro-processing of spent FR fuels. In the domain of hydrochemical processes, development is flourishing. Multi-step processes look promising but most of the systems developed so far appear complex and probably need to be simplified. In the domain of pyrochemical processes, interest has strongly revived in “old concepts” including fluoride volatilisation.

Hydrochemical processes

Extending the current industrial Purex processes towards improved separation of neptunium, and further to the other minor actinides, is a research topic in many countries. The future R&D tasks to develop such advanced aqueous reprocessing technology may be summarised as:

- Reducing the size of head-end equipment.
- Enhancing and optimising equipment with respect to corrosion resistance, extraction performance, etc.
- Improving the efficiency of MA recovery processes to reduce waste.

It also seems important to simplify the hydrochemical MA and LLFP separation processes, and reduce their space requirements. Some routes for improvement can be proposed:

- Single-cycle operation.
- Considering High Active Concentrates instead of High Active Raffinates as source material (large volume reduction factor).
- Integrating MA and LLFP separation processes.
- Considering less familiar LLFPs for possible partitioning.
- Maintaining the "CHON principle" to minimise secondary solid wastes.

Table 7.8 gives a brief overview of the status of R&D on the various aqueous partitioning techniques as was reported in the first-phase P&T systems study "Status and Assessment Report of Actinide and Fission Product Partitioning and Transmutation" [2].

Three phases were distinguished:

- *Phase 1* corresponds to research on the principles of the process. In many cases, it overlaps the basic research conducted in the laboratory (for example, research on new extractant compounds). Its completion demonstrates the scientific feasibility of the process.
- *Phase 2* is the process development step. It includes all research designed to develop the complete flow chart, describe its application, and guarantee its performance. The conclusion of this step demonstrates the technical feasibility of the process.
- *Phase 3* relates to the industrialisation of the process. It is aimed to ensure overall active operations in industrial conditions. In practice, these studies are essentially conducted by engineering design. It terminates in the industrial feasibility of the process and its potential application in an industrial installation.

For all these processes the development of new extractant molecules and the improvement of existing ones is carried out world-wide. In particular the most challenging separation, that of actinides from lanthanides, has inspired work on diphosphines in Russia, research on dithiophosphinic acid derivatives in China and Germany, the improvement of TPTZ and BTP derivatives in France and in India the examination of sulfoxide-type extractants.

For the sake of process industrialisation, the economics and the radiation resistance of the organic molecules are important R&D issues. The aim is to develop sustainable, environmentally friendly processes. A direct selective extraction of actinides from the Purex raffinate would reduce the number of process steps, and pre-concentration of the raffinate by a factor of ~10 the volumes of liquid to be handled. Cost-effective, robust and simple processesare needed with either well-established technologies (pulsed columns, mixer settlers and centrifugal contactors) or new technologies such as hollow fibre modules. In all cases it is important to keep a good balance between fundamental chemical research, process development and qualification using genuine high active wastes originated from real spent nuclear fuels.

Table 7.8. **Status of R&D on aqueous separation techniques**

	Phase 1	**Phase 2**	**Phase 3**	**Remarks**
U and Pu separation (PUREX)	–	–	–	Achieved industrially
Np separation (PUREX)			X	95% separation
(PUREX)		X		>95% separation

(DIDPA)		X		
(HDEHP)		X		
(TRUEX)		X		
Am + Cm separation:				
– based on An/Ln co-extraction				
(TALSPEAK)			X	
(DIDPA)		X		
(TRUEX)		X		
(TRPO)		X		
(DIAMEX)		X		
– based on An selective extraction				
(TPTZ)	X			
(Picolinamides)	X			
(CYANEX 301)	X			SF=5900
(BTP)				
– based on precipitation	X			
(Ferricyanide)				
Am separation in the oxidised state				
(SESAME)		X		Am/Cm separation
Tc separation				
(PUREX)			X	Soluble Tc
(PUREX)	X			Insoluble Tc
Tc-PGM separation				
(Denitration precipitation)		X		
(Active carbon adsorption)		X		
I separation(PUREX)			X	95% separation
Zr separation (PUREX)		X		
Cs separation (Calixarenes)		X		
(Zeolite)		X		
Sr separation (Titanic acid)		X		
Cs and Sr separation (Dicarbollides)			X	
Pd (PGM), Se, Ru separation				
(Electrolytic extraction)	X			Soluble Pd, Se, etc.

Pyrochemical processes

With the completion of the demonstration review by the NRC and a positive non-proliferation assessment, the Department of Energy (DOE) decided to use this technology to process the remaining EBR-II fuel (approximately 25 tonnes) and some sodium-bonded metal fuel from the Fast Flux Test Facility (FFTF). After completion of an environmental impact statement, these production operations started in September 2000. The work performed to date on the treatment of nitride and oxide fuels has been on either the laboratory or engineering scale. The feasibility of the processes has been demonstrated, but large-scale tests have not been performed with irradiated spent fuel.

The Spent Fuel Treatment Program at ANL demonstrated many parts of the pyroprocess fuel cycle, but there are still key aspects that have yet to be demonstrated on a large scale with radioactive materials. The main outstanding issue is the recovery of transuranics. Large-scale equipment has been fabricated for transuranic recovery, but with the termination of the IFR program, the equipment and process was never tested beyond the laboratory scale.

The remote fabrication of IFR fuel was not part of the Spent Fuel Treatment Program, but the same technology was used to fabricate cold fuel for EBR-II and a demonstration of another pyroprocess (melt refining) for recycling EBR-II in the 1960s employed remote fabrication for 34 500 fuel elements.

One challenge for a pyroprocessing system is selecting the appropriate materials of construction for the high temperature processes. Material improvements are needed in order to lessen the formation of dross streams and increase material recovery and throughput.

The quantity of waste generated that requires geological disposal from pyroprocessing appears to be comparable at present to modern commercial aqueous processes. Advancements are being pursued to further reduce the disposal volumes through zeolite ion exchange processes. This technology has not been demonstrated beyond the laboratory scale.

Most of the radioactive work performed to date has been on the pyroprocessing cycle for metal fuel. Laboratory work has been performed on the head-end operations for oxide reduction and on the nitride fuel cycle. Demonstrations of these technologies with actual spent fuel are still needed. Additionally for nitride fuels, demonstrating the recycle of nitrogen is critical since ^{15}N is specifically required for the fuel in order to eliminate the formation of radioactive ^{14}C.

Further work is required in a number of areas related to process design and equipment. Actual irradiated fuel should be used to verify the behaviour of the TRUs and fission products. In addition, much work needs to be done to understand the fundamentals of the salt-recovery step to provide a basis for construction of more efficient cells and to understand the behaviour of fission products in this step. Alternative, lower-cost oxygen-evolving electrodes must be developed for the salt-recovery step.

Directions for improving the pyrochemical processes appear to be:

- Minimisation of TRU losses in wastes and increase of the purity of the separated fractions.
- Actinides that can be obtained through the combined use of several separation techniques and multi-stage techniques.
- The waste problem, which is mostly corrosion related owing to the aggressive process media.
- Character of the media and the high process temperatures, needs to be precisely estimated.
- Consideration of the possible separation of LLFPs.

Table 7.9 shows the current status of dry methods in analogy to the Table 7.8 for the aqueous reprocessing techniques.

Table 7.9. **Status of R&D on dry separation techniques [196]**

Process/Fuel type	Phase 1	Phase 2	Phase 3	Remarks
Pyroprocessing (LWR oxide fuel)				
Fluoride volatility		X		Process operated in Czech Republic in 1980s; experimentation in US. in 1960-1970s; supplemental experience via enrichment process
Electrorefining (U-Zr metal alloy fuel)				
U recovery		X		From EBR-II fuel treatment application
TRU recovery	X			Laboratory-scale work with liquid cathode (Pu only)
TRU separation	X			Cadmium distillation process (Pu only)
Noble metal fission product extraction		X		From EBR-II fuel treatment application
Ln, Cs, Sr fission product extraction	X			Laboratory-scale zeolite process
Tc, I recovery	X			Not considered

Electrorefining (U-Pu-Zr metal alloy fuel)				
U recovery	X			Not tested with irradiated fuel
TRU recovery	X			Laboratory-scale work with liquid cathode (Pu only)
TRU separation	X			Cadmium distillation process (Pu only)
Noble metal fission product extraction	X			Not tested with irradiated fuel
Ln, Cs, Sr fission product extraction	X			Laboratory-scale zeolite process
Tc, I recovery	X			Not considered
Oxide electrowinning (RIAR Process)				
U recovery		X		LWR fuel processed for BOR-60 MOX fuel production
Pu recovery		X		LWR fuel processed for BOR-60 MOX fuel production
MA recovery	X			At early development stage
Noble metal fission product extraction	X			At early development stage
Ln, Cs, Sr fission product extraction	X			At early development stage
Tc, I recovery	X			Not considered
Pyroprocessing (Non-fertile metal alloy fuel)				
TRU recovery	X			Laboratory-scale work with Pu only
FP extraction	X			Laboratory-scale work with lanthanides only
Tc, I recovery	X			At early development stage
Pyroprocessing (Non-fertile oxide fuel, inert matrix)				
Direct electrochemical reduction/electrorefining	X			Laboratory-scale work (U, lanthanides only)
Pyroprocessing (Non-fertile nitride fuel, inert matrix)				
Electrorefining process	X			At early development stage
Pyroprocessing (Non-fertile graphite particulate fuel)				
Fluoride volatility	X			Some experience with aqueous processing applies
Pyroprocessing (Molten salt fuel)				
Fluoride volatility		X		Work at NRI-Rez (Czech Republic), and Oak Ridge National Laboratory

Collaborations

It seems a pressing necessity to maintain, or best to increase, the collaborations in this complex field of reprocessing research at:

- National levels: maintain or create network(s) between academic and applied research bodies. As an example, in France, two networks exist working under the auspices of the December 1991 Nuclear Waste Act: the so-called PRACTIS and NOMADE Groupes de Recherches.
- Bi-national levels: numerous collaborations exist, e.g. CRIEPI-ANL, CEA-JNC, CEA-JAERI etc.
- Regional level. As an example, at the European level common works exist which are partly financed by the EU, e.g. the PARTNEW, CALIXPART and PYROREP programs within the EC 5th Framework Programme (2000-2003). The role of ITU at Karlsruhe is also very important for European and wider collaborations,
- At the International level, the roles of OECD/NEA for Workshops and Working Parties and also of IAEA appear essential.

7.3.4 Technology development

Where the previous paragraphs discussed the needs on basic R&D, the path towards testing, demonstration and finally deployment of such MA or TRU transmuters will be a long one where technological development will increasingly be needed. In particular, heat removal and ancillary systems meeting stringent licensing conditions will need further exploration once a basic concept has been accepted.

Several systems will need to be developed where, today at least, only limited experience and limited infrastructure are available to test the components. Therefore, technological development will need to be considered in the fields of:

- Primary and secondary pumps.
- Intermediate heat exchangers and steam generators.
- Vessel features, such as head, support structures and fuel handling equipment.
- Ancillary systems such as the coolant cover gas system, cold trap and filter.
- Penetration of the proton beam line through the reactor building and vessel.

As the development of such advanced nuclear fuel cycles with TRU/MA-transmuters is a long undertaking, it is to be envisaged that technology development, e.g. the building of pilot or demonstration plants, will most probably have to be initiated by R&D-organisations within an international context and, most probably, is a phased programme.

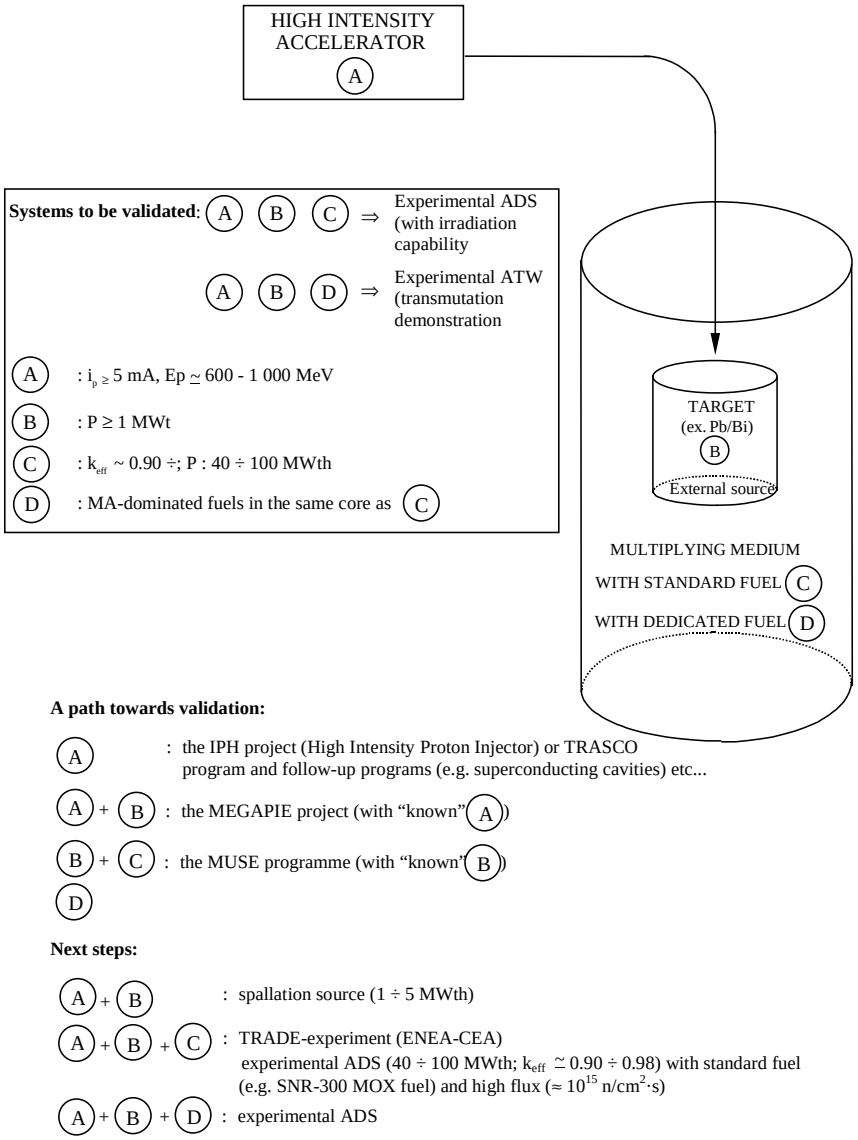
7.3.4.1 Pilot plants and demonstration facilities

The previous sections have already indicated the different activities needed to develop ADS facilities. Some of these projects are integrated in an R&D programme oriented towards developing a prototype, pilot or demonstration plant. Some examples of these initiatives are ADTF (USA), ADS Experimental Facility (Japan), XADS (Europe), Gas-cooled ADS (France), Lead-bismuth cooled (Italy), MYRRHA (Belgium), HYPER (Korea) and others.

As was indicated above, some very specific R&D issues, associated with experimental requirements, are to be handled first as a means to finalise detailed designs or, at least, to respond to the stringent licensing demands for such pilot plants. The continuous availability of irradiation devices in existing MTRs will therefore be very important. The neutron fluxes and/or the irradiation conditions in such critical (fast) MTRS, if available at all, may not always be representative enough for the mixed irradiation fields in the ADS field. For those countries embarking on the technology development of ADS, a demonstration ADS-type irradiation facility in an international programme may be needed.

A phased approach to the development of ADS may therefore be highly recommended while maximising the use of existing research facilities world-wide. Such a phased approach is schematised in Figure 7.2. Some of the ongoing programmes, for instance the KEK/JAERI programme in Japan, work through the intermediate and multi-purpose phase of a multi-megawatt spallation neutron source.

Figure 7.2. **A step-by-step approach to validating and demonstrating the ADS concept**



7.3.4.2 Multi-purpose facilities?

The "transmutation" community has recently become involved in discussing multipurpose facilities, based on a high power proton accelerator which provides neutrons by spallation on one (or several) target(s) for different applications. Some synergy may be sought between the development of ADS and FR but also between ADS and other fields of nuclear science and technology, e.g. spallation neutron sources, medical applications, radioactive beam facilities, and so on. Some OECD Member countries have envisaged such synergy, i.e. the joint KEK/JAERI programme in Japan, while other countries are studying proposals, e.g. the AAA programme in the USA.

There may be some good reasons to search for such synergy between the ADS-development for transmutation purposes and other uses, e.g.:

- More effective use of scarce financial and human resources.
- Phasing the development while better managing the inherent risks involved.

- Sharing technological development among different disciplines.
- Optimising the learning-curve process in developing ADS.

However, other considerations from a scientific and technical perspective counterbalance these advantages for synergy and may even jeopardise the prospects of a dedicated ADS development for transmutation purposes, i.e.:

- *Sharing of the available beam*. In some cases, technical measures can be used to change the time structure of beams to optimise them for particular applications.[72] Total available intensity must be considered. Clearly, there are limits to the ability to share a finite proton flux, however large, among users who require maximum intensity for their experiments. In addition, acceleration of negative ions and of protons may have to be supported in the same accelerator.
- *The needs and characteristics of the user communities*. The size and operating modes of user communities are important factors in deciding whether a shared large facility is warranted. In general, dedicated, longer-term projects cannot be easily combined with those where scientists from many disciplines come to the facility to perform short experiments (as is the case for many users of neutron sources). In addition, ADS-transmuter applications may have special safety and regulatory constraints that would pose particular organisational problems at a shared facility. In some cases, especially for those machines that are at or beyond the state of the art, accelerator physicists would need frequent access to the machine, with unscheduled down-times and frequent modifications. In addition, most of the other users do not require the high reliability demanded by ADS and may therefore not accept the over-design, and thus costs, for such a shared accelerator.
- *The time-scales of the various scientific programmes*. Long-range plans for the applications are in various stages of maturity at national and regional levels. Besides the difficulties of adding yet another level of co-ordination, there is the simple observation that each field should proceed according to its own optimum pace, which may not be compatible with the concept of multipurpose facilities.

In addition, the successful implementation of any of the facilities, whether for single or especially for multiple purposes, will depend on the ability of scientists and engineers to design and operate High Power Proton Accelerators (HPPA) with beam energies in the Giga electron-volts (GeV) region, and power levels ranging from 1-5 megawatts for some applications to 50 megawatts for others. In several critical areas, the necessary levels of performance greatly exceed the state of current knowledge and technological capability (see Chapter 4).

The notion of combining several HPPA applications at one facility deserves serious attention, with a realistic appraisal of the needs, benefits, and difficulties. Opportunities for joint R&D should be exploited whenever possible, even when separate facilities are in order. While ADS for transmutation purposes would definitely benefit from sharing the technological development of HPPA with other users, other users are not always receptive to such sharing of facilities for the reasons given.

It may therefore become accepted that ADS may need to develop its own accelerator, including all aspects related to licensing, over-design, high reliability etc adding to the costs. This may remain true until such high power and high reliable accelerators become readily available in the market-place.

72. Intermediate options are worth considering, viz., a single site with more than one accelerator and shared infrastructure such as power distribution and cooling systems, or office space.

As a conclusion, these developments are grounds for the nuclear community to look for further increased international collaboration if developing such ADS is considered a priority.

7.4 Conclusions

Based on the above considerations and supported by the conclusions in the previous chapters, we may conclude that:

- *Basic R&D* is needed for the new FR and ADS in the fields of nuclear data and neutronic calculations, fuel technologies, structural materials, liquid metals, reprocessing technologies, target materials and high power accelerators (the last two only for ADS).
- *Experimentation on fuels is a priority*. No concept can be considered seriously unless the appropriate fuels are defined, which means characterised, fabricated, irradiated and reprocessed.
- Since fuels play a central role in all scenarios of waste minimisation and nuclear power development, an international sharing of efforts on nitrides, oxides and metals should be organised in order to ensure an optimum use of resources in the few existing laboratories to handle very active fuels.
- In that connection, *the availability of irradiation facilities, in particular able to provide fast spectra and high damage rates, is a key point and a major concern*. Again, an international initiative could be envisaged to harmonise programmes and to allow the best use of existing resources. Identifying the experimental irradiation needs in such a shared international facility would be a worthwhile undertaking.
- *Demonstrating at the appropriate scale the performance of pyrochemical processes* (level of losses, secondary waste, etc.) is needed in order to assess the technico-economic viability of certain fuel cycle scheme options.
- In the field of basic R&D supporting FRs as well as ADS, the discussion of *coolants for FR or ADS* would benefit from a better international agreement on the advantages and drawbacks of the different options.
- *Improved modelling tools to simulate the materials behaviour* under (mixed) irradiation conditions, and possibly high temperatures, may prove to be a very valuable approach and a sharing of expertise and benchmarking within an international context may be advocated.
- *Safety analysis of ADS* should identify the possible paths to exclude HCDAs in ADS. If such a HCDA has to be taken into account in the safety analysis of an ADS, a prompt negative feedback mechanism for quenching such an accident has to be developed.
- And last but not least, *performance assessment studies for a geological disposal site using a P&T source term* are necessary in order to clarify the cost-benefit analysis of such advanced fuel cycles, including this geological disposal.

8. FISSION PRODUCT TRANSMUTATION

8.1 Introduction

Transmutation of actinides and fission products using thermal reactors has been extensively discussed in the "Status and Assessment Report on Actinide and Fission Product Partitioning and Transmutation" [2].

As regards the transmutation of long-lived fission products to shorter lived or stable nuclides, the report concluded that this is theoretically possible in some cases, but higher fuel enrichments are necessary in LWRs. The transmutation of ^{99}Tc and ^{129}I would take a long time, characterised by a half-time by transmutation of about 44-70 years. Intensive development of target and fuel assembly materials would be required, e.g. in the case of iodine, as well as refined isotopic separation in the case of elements such as Se , Zr, Cs and Sn.

This chapter will again touch upon these aspects and will briefly describe new considerations in relation to the possibility of transmuting fission products and the respective role of the ADS.

8.2 Fission product transmutation

The first phase P&T systems report [2] already discussed the possibilities and limits to transmute fission products. In essence, this study concluded that from a view of reducing radiotoxicity, transmuting fission products is of very little interest. The majority of the fission products has decayed after about 250 years, and their contribution to the radiotoxicity of the spent fuel, which was very high during the first 100 years of storage, has become small. However, some fission products are very mobile in certain geological environments and can thus contribute significantly to the radiological effects of disposal in underground repositories. In addition, the treatment of spent fuel results in releases through gaseous and liquid effluents which also contribute to the long-term radiological effects of nuclear power generation. The fission products that deserve most attention in this respect are ^{129}I, ^{135}Cs, ^{79}Se, ^{99}Tc and ^{126}Sn (see Chapter 2, Section 2.2).

Unlike transuranics, fission products in a transmutation process produce no supplementary neutrons but are purely consumers. As was seen previously in Chapter 2, neutron consumption is the most important parameter, if one wants to assess the potential of transmutation in a given nuclear system.

The rate of transmutation of a nuclide J can be characterised by the time T_J^{transm} needed to incinerate half of an initial mass, which is function of the cross-section $\sigma_{n,\gamma}^J$ (barns) and of the neutron flux ϕ (n/cm^{2}s):

$$T_J^{transm} = \frac{\ln 2}{\sigma_{n,\gamma}^J \phi \times 3.15 \times 10^7} \text{ years}$$

Transmutation of the toxic fission products in nuclear reactors and sub-critical systems may be sensible if rates of nuclear interactions with neutrons are much higher than rates of natural decay, which are defined by decay half-life $T_{1/2}$. That is, transmutation in a neutron flux can be reasonable if $T_{1/2} >> T_J^{transm}$. Table 8.1 gives some properties of long-lived fission products (LLFP) and Table 8.2 compares natural decays and transmutation rates.

Table 8.1. **Physical parameters of the major LLFP**

Isotope	**Period (y)**	**Decay mode**	**Thermal power (W/Bq)**	**Dose (ingestion) (Sv/Bq)**	**Fraction in an irradiated fuel (g/t)[(a)]**
^{14}C	5.7×10^{3}	β	1.6×10^{-14}	5.7×10^{-10}	1.3×10^{-1}
^{36}Cl	3.0×10^{5}	β^-, β^+	4.4×10^{-14}	8.2×10^{-10}	1.6×10^{0}
^{79}Se	6.5×10^{4}	β	6.5×10^{-15}	2.3×10^{-9}	4.7×10^{0}
^{90}Sr	2.9×10^{1}	β	2.8×10^{-14}	3.9×10^{-8}	5.0×10^{2}
^{90}Y	7.3×10^{-3}	β	1.5×10^{-13}		1.3×10^{1}
^{93}Zr	1.5×10^{6}	β	2.6×10^{-15}	4.2×10^{-10}	9.8×10^{1}
^{99}Tc	2.1×10^{5}	β	1.4×10^{-14}	3.4×10^{-10}	8.2×10^{2}
^{107}Pd	6.5×10^{6}	β	1.4×10^{-15}	3.7×10^{-11}	2×10^{2}
^{126}Sn	1×10^{5}	β	4.2×10^{-14}	5.1×10^{-9}	2.0×10^{1}
^{126}Sb	3.4×10^{-2}	β	5.0×10^{-13}		6.9×10^{-6}
^{129}I	1.6×10^{7}	β	1.3×10^{-14}	7.4×10^{-8}	1.7×10^{2}
^{135}Cs	2.3×10^{6}	β	9×10^{-15}	1.9×10^{-9}	1.3×10^{3}
^{137}Cs	3.0×10^{1}	β	3.2×10^{-14}	1.4×10^{-8}	1.1×10^{3}
^{137m}Ba	4.9×10^{-6}	β	1.1×10^{-13}		1.7×10^{-4}
^{151}Sm	9.0×10^{1}	β	3.2×10^{-15}	9.1×10^{-11}	1.6×10^{1}

(a) UOX from a PWR (3.5%, 33 GWd/t).

For the "transmutable" fission products in Table 8.2, the capture cross-sections are high enough for transmutation to be much faster than natural decay. For most of the transmutable fission products, the thermal spectrum is as good as or better than the fast spectrum (exception ^{107}Pd). For three isotopes, ^{90}Sr, ^{137}Cs and ^{151}Sm, natural decay is or as fast as or much faster than transmutation and it is more reasonable to put them into interim storage to decay. As for "questionable" isotopes (^{79}Se, ^{126}Sn and ^{94}Nb), they are rather long-lived, however, owing to small cross-sections, their transmutation will be slow. However, the yield of these isotopes is limited indicating that total toxicity is rather modest.

In Section 2.4.1 of Chapter 2, we introduced the overall neutron excess parameter -D which is defined as the total number of neutrons which have to be spent to incinerate a radionuclide including its daughter-products. Table 8.3 gives values of D for separated isotopes and Table 8.4 for elements.

Table 8.2. **Parameters of LLFP to be eventually transmuted in a fast (E_n (neutron energy) = 0.2 MeV) and thermal (E_n = 1 eV) spectra with standard flux levels: $\Phi = 10^{15}$ (n/cm^2·s) and $\Phi = 10^{14}$ (n/cm^2·s) respectively**

Isotopes, J	$\sigma^J_{n,\gamma}$ (barn)		$T_{1/2}$ (year)	T^{transm} (year)		Recommendation from neutronic viewpoint
	Fast spectrum	Thermal spectrum		Fast spectrum	Thermal spectrum	
^{79}Se	0.03	0.1	6.5×10^4	7.3×10^2	2.2×10^3	*Questionable*
^{90}Sr	0.01	0.14	29	2.2×10^3	1.6×10^3	*Non-transmutable*
^{93}Zr	0.03	0.28	1.5×10^6	730	790	*Transmutable*
^{94}Nb	0.04	2.2	2.0×10^4	5.5×10^2	1×10^2	*Questionable or transmutable*
^{99}Tc	0.2	4.3	2.1×10^5	110	51	*Transmutable*
^{107}Pd	0.5	0.3	6.5×10^6	44	730	*Transmutable*
^{126}Sn	0.005	0.05	1×10^5	4.4×10^3	4.4×10^3	*Questionable*
^{129}I	0.14	4.3	1.6×10^7	160	51	*Transmutable*
^{135}Cs	0.07	1.3	2.3×10^6	310	170	*Transmutable*
^{137}Cs	0.01	0.02	30	2.2×10^3	1.1×10^4	*Non-transmutable*
^{151}Sm	0.7	700	89	31	0.3	*Non-transmutable or questionable*

Table 8.3. **Overall neutron excess parameter (-D) of "transmutable" and "questionable" isotopes together with their yields (Y_J) per fission in LWR (UOX) after 5 years of cooling time. Time interval between reprocessing steps: 3 years (removable nuclides: all fission products except all isotopes of Zr, Tc, Pd, I, Cs, Sn, Nb, Se)**

Transmutable and questionable isotopes (J)	-D (neutron/transmutation)	Y_J nuclei/fission in NP
^{93}Zr	-2.01	0.050
^{99}Tc	-1.01	0.055
^{107}Pd	-2.04	0.015
^{129}I	-1.008	0.009
^{135}Cs	-1.002	0.017
^{126}Sn	~-2	0.0012
^{94}Nb	-0.985	6.3×10^{-7}
^{79}Se	~-2	0.0004

Table 8.4. **Overall neutron excess parameter (-D) of "transmutable" nuclides (including all isotopes) together with its yields (Y_J) per fission in LWR (UOX) after 5 years of cooling time. Time interval between fission products reprocessing: 3 years (removable nuclides: all fission products excluding all isotopes of Zr, Tc, Pd, I, Cs)**

Transmutable nuclides, J	-D (neutron/transmutation)	Y_J (nuclei/fission in NP)
all Zr	-2.03	0.26
all Tc	-1.01	0.055
all Pd	-3.22	0.095
all I	-1.01	0.011
all Cs	-0.58	0.13

In fact, it is helpful to use two types of neutron consumption definitions, depending on the choice of units, -D* (neutron/transmutation) and -D (neutron/fission). D can be obtained as the product of D* and of the yield of a nuclide per fission, Y. If a LLFP transmuter is fed constantly with a group of nuclides, then the D value of this group is the sum of $Y \times D^*$ of the group components.

For example, taking into account LLFP yield, one can calculate the total neutron consumption needed to incinerate all "transmutable" and "questionable" long lived isotopes of fission products:

$$D(^{93}Zr,^{99}Tc,^{107}Pd,^{129}I,^{135}Cs,^{126}Sn,^{94}Nb,^{79}Se) = \sum_J D_J^* Y_J \cong 0.22 \,(\text{neutron/fission in NP})$$

This value defines the total neutron consumption for incineration of all LLFP presented above, if preliminary isotope separation of fission product in LWR discharge has been realised.

To incinerate all Tc, I, Cs without isotopic separation, one needs about 0.15 (neutron/fission in nuclear park). Isotopic separation of ^{99}Tc, ^{129}I and ^{135}Cs allows to reduce this neutron consumption to 0.08 (neutron/fission in NP) where 0.009 and 0.056 neutrons per fission are needed, respectively, for ^{129}I and ^{99}Tc.

To transmute elements such as Tc, I, Cs in a fast spectrum transmuter one needs to know the neutron surplus G available and to the fraction (f) of these transmuters in a Nuclear Power Park (NPP).

Taking into account a "standard" value of the neutron parasitic capture (CM) and the neutron leakage (L) as CM + L = 0.3 neutron/fission (which is valid for a fast reactor of an intermediate size and traditional composition), one gets for the neutron surplus (for example in a sub-critical system):

$$G = -\sum_J \varepsilon_J \times D_J - (CM + L) + \mu ,$$

where ε_J is a fraction of J-nucleus in fuel, μ is a neutron spallation source ($\mu \approx 0.15$ neutron/fission if $k_{eff} = 0.95$).

It is obvious that the neutronic potential of a fuel cycle scheme for the transmutation of fission products depends on the fraction of fission product transmuters in the scheme. Table 8.5 quantifies this potential for the three principal transmutation schemes, assuming that the fission products are transmuted in the TRU or MA burners.

Table 8.5. **Performance of transmutation schemes for fission product transmutation**

	TRU burning in FR (Scheme 3a)	**TRU burning in ADS (Scheme 3b)**	**Double Strata (Scheme 4)**
Contribution of TRU or MA burner to total thermal power	0.37	0.222	0.0581
k_{eff}, averaged over reactor cycle		0.946	0.945
Neutron excess (-D from Table 2.1)	1.48	1.29	0.79
Overall neutron balance corrections:			
– for capture and leakage	-0.3	-0.3	-0.3
– for sub-criticality	0	0.15	0.15
Neutrons available for transmutation:			
– per fission in ADS	1.18	1.14	0.64
– per fission in fuel cycle scheme	0.44	0.253	0.037

These results show that the ADS-TRU burning scheme could, in principle, transmute all "transmutable" and "questionable" LLFPs whereas the ADS-MA burning scheme would be able to handle the transmutation of ^{129}I and, in addition, 50% of the ^{99}Tc. In contrast, the FR-TRU scheme would be able to transmute all "transmutable" and "questionable" LLFPs.

Finally, the maximum rate of transmutation (RT, measured in number of transmutations per fission) of any isotope in a given transmuter can be easily evaluated if the neutron surplus G available for LLFP transmutation is known:

$$RT_J = \frac{G\,(\text{neutron / fission})}{D_J^*\,(\text{neutron / transmutation})}$$

One can then calculate the maximum rate R_J^{max} of transmutation of a given isotope J per GWth $\times$ year of a transmuter:

$$R_J^{max}\,(\text{kg / GWth} \times \text{year}) \approx 1.6 \times A_J \times RT_J$$

where A_J is the atomic number of isotope J.

For example, D_J^* is equal to 1.01 for ^{99}Tc and, in a fast spectrum sub-critical transmuter based on TRU from LWR-UOX, G = 0.96. Then, $R_J^{max} \simeq 160$ kg/GWth $\times$ year if all the neutron surplus is devoted to ^{99}Tc transmutation.

In practice, for the sake of transmuting LLFP, one can imagine coupling the most favourable characteristics (i.e. high flux and high cross-sections) in a neutron field. This has been achieved for example in the so-called LSD (leakage with slowing down) concept, illustrated in Figure 8.1 and demonstrated in the TARC-experiment [94].

Figure 8.1a. **Leakage slowing down concept for LLFP transmutation**

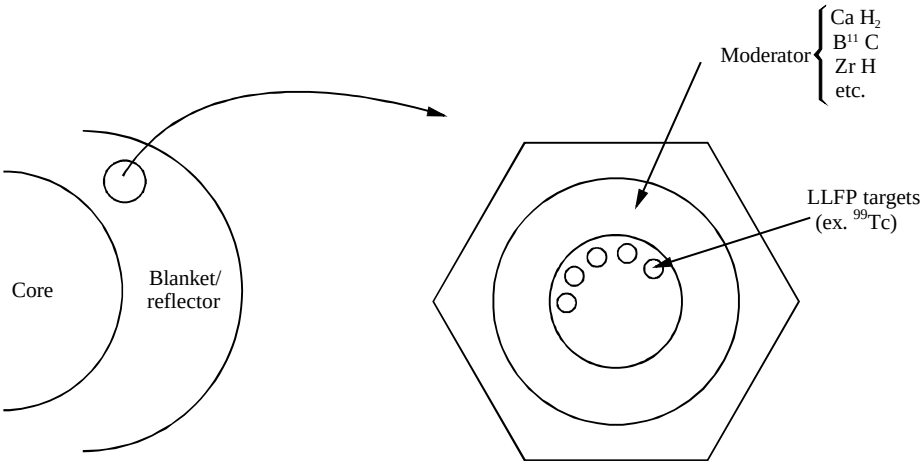


Figure 8.1b. **Example of LSD for the reaction rate of ^{99}Tc**

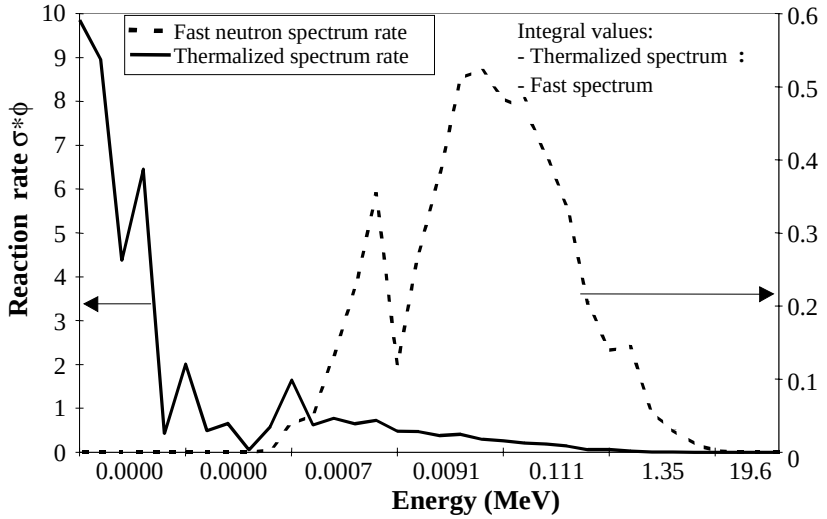


Table 8.6 gives some quantitative values for the transmutation of ^{99}Tc and ^{129}I in both a pure fast and moderated spectra.

Table 8.6. **Transmutation of ^{99}Tc or ^{129}I**

	Masses (kg/TWhe)			σ_c	Φ	$T_{1/2}$ **(transm.)**
	Loaded	Transmuted	%	Barns	10^{15} n/cm^2/s	Years
In fast spectrum						
^{99}Tc	60.5	6.13	10.1	0.32	0.758	91
^{129}I	(26.0)	(3.96)	15.2	0.31	1.21	59
In moderated spectrum.						
1% ^{99}Tc	1.17	1.08	92	11.9	0.493	3.7
5% ^{99}Tc	5.86	3.62	62	5.85	0.372	10
10% ^{99}Tc	11.73	4.88	42	3.46	0.353	18
20% ^{99}Tc	23.46	6.19	26	2.19	0.317	32
To be compared to productions:			≈ 3 kg/TWhe (^{99}Tc)			
			≈ 0.7 kg/TWhe (^{129}I)			

8.3 Conclusions

- Excess neutrons produced by critical and sub-critical burners can, in principle, be utilised to transmute fission products. With the neutron fluxes available in these systems, it is theoretically possible to transmute the long-lived fission products; the transmutation of the more abundant short-lived fission products, however, is impracticable due to insufficient transmutation rates. In practice, the necessity of isotopic separations and difficulties in the preparation of targets present difficult obstacles for the fission product transmutation which currently reduce the number of candidate nuclides to only one or two, i.e. ^{99}Tc, and, possibly, ^{129}I.

- Minimising the fraction of specialised transmuters in the reactor park can result in an insufficient neutronic potential for transmuting the long-lived fission products of the entire park. The present study shows that critical and sub-critical TRU burners perform similarly in this respect. If the transmutation would be limited to ^{99}Tc and ^{129}I, all TRU burning strategies could, in principle, accomplish the task; however, a neutron shortage would not allow these fission products to be completely transmuted in the minor actinide burners of a double strata scheme.

9. ALTERNATIVE ACTINIDE TRANSMUTATION APPROACHES

9.1 Introduction

This study is essentially focused on partially or fully closed fuel cycles using solid fuel in fast spectrum reactors. There are, however, other transmutation concepts that are proposed using other fuel and reactor types, including thermal-spectrum systems. The Expert Group therefore recognised that an overview of alternative approaches to the options described in the previous chapters would be appropriate in order to complement the study.

9.2 Transmutation systems using thermal neutrons

As was mentioned in the introduction, this report aims to give a comparison of FR and ADS based on solid fuel fast spectrum devices. Despite this initial focus, this section will address those systems using other approaches, e.g. thermal spectra and liquid fuels. In particular, molten salt systems were not assessed in depth in the first phase report [2] and are not in this report, but in recent years there has been a renewed interest in those systems both conventionally critical and accelerator driven.

Accelerator-driven systems are very difficult to operate with thermal neutrons and solid fuels (as in conventional LWRs) owing to prohibitive power peaking problems. Therefore, ADS concepts based on liquid or quasi-liquid fuels offer a way round these serious constraints. Two categories of reactors and ADS capable of transmutation are under serious considerations nowadays:

- Molten salt systems.
- Particulate-fuelled gas cooled reactors or ADS.

9.2.1 ADS and reactors with liquid or quasi-liquid fuel

9.2.1.1 Molten salt reactors and ADS

One of the alternative concepts for a nuclear reactor or accelerator-driven system having a significant transmutation potential is a molten-salt system, derived from an idea of a fluid-fuel reactor extensively investigated at the dawn of the nuclear era. Indeed, it is very appealing with its potential for a very effective consumption or transmutation of nuclear fuel (most of the constraints which limit burn-up in solid fuel are relaxed), inherent safety features regarding super-criticality accidents, etc. The molten salt fuel at operating fissile concentrations provides inherent protection against criticality accidents during handling. In thermal neutron designs, the graphite moderator is required for criticality criticality which can therefore occur only in the core. In other concepts, the design would have to exclude vessels that are not safe from criticality with credible fuel mixtures.

Molten salt reactors (MSR) can be designed with or without on-line processing, or with only partial processing. With reprocessing, some of the common nuclear reactor terminology does not apply. Except for some start-up periods, molten salt reactors operate at an equilibrium steady state. The fuel concentration and content do not vary with time. Fissioned or consumed fuel is replenished by feeding or by breeding. The term “fuel burn-up”, commonly used for solid-fuel reactors, thus has not the same meaning, as there is no specific amount of energy generation related to a particular identifiable original amount of fissile material. For the same reason, there is no excess reactivity to compensate for burn-up or for the progressive poisoning caused by fission product accumulation as in solid fuel reactors. Also decay heat problems for the MSR are not as severe in comparison with traditional solid fuel because the concentration of UF_4 in the fuel salt is small (l-2 mole%).

Molten salts can operate at high temperatures and low pressures, and have favourable heat transfer properties. These properties result in high thermal efficiency for the reactor and freedom from hazards associated with high pressures. The salts are chemically stable and non-flammable, averting fire hazards, and there are no energetic chemical interactions between the salts and water.

One of the major advantages of the fluoride-based MSRs is the potential for an integrated fuel recovery capability. The processing is based on the high volatility of UF_6. By sparging the salt with fluorine, uranium can be removed as UF_6, which can then be converted back to UF_4 and recycled into a fresh batch of fuel salt. The residual salt, free from uranium, could be subjected to any of a number of processes to remove fission products and concentrate them. The carrier salt components (lithium, beryllium, fluorine) could also be isolated and recycled if that were economically desirable. All of these steps could be made independent of the reactor operation [197].

One of the first molten salt reactors was an experimental type for aircraft [198], which operated successfully in 1954 in a “proof-of principle” short-term test at a power level of 2.5 MWth and at temperatures up to 860°C. The fuel was a solution of UF_4 in other fluorides with Inconel-clad beryllium as the moderator. At a very early stage of this project it was realised that the molten-salt system was an attractive option for a power reactor. In 1957 Oak Ridge National Laboratory began work on a commercial application. A reactor experiment (MSRE) was set-up and operated successfully at a power level of 7.3 MWth until the end of 1969 [199,200], delivering data for over 17 000 hours of critical operation and confirming the feasibility of eventual power production.

Building on MSRE experience ORNL has developed further concepts, particularly of a Molten Salt Breeder Reactor (MSBR) [201] using ^{7}LiF-BeF_2-ThF_4-UF_4 as the fuel salt in the Th-^{233}U fuel cycle. It reached only a very low breeding ratio of 1.07 but with a very advantageously low nuclear fuel concentration in the salt and consequently a very low inventory of fissile materials in the core.

In the mid-1970s, MSR development in the USA had progressed far enough to justify a greatly expanded effort leading towards commercial deployment, but competed with the LMFBR programme. To avoid diverting substantial funding from the latter, the MSR programme was instead stopped [202].

Other countries such as France, Japan and Russia put some effort into developing MSR concepts in the 1970s and 1980s, recognising their potential for more effective use of nuclear fuel through a gradual introduction of Th into the cycle.

Reducing the accumulation of Pu in LWR spent fuel was initially a main driver in the development of another concept of MSR. The fluoride salts used in thermal neutron systems were not suitable for fast neutron cores because of their neutron moderating ability, so interest turned towards the use of molten chlorides. 60NaC1-37$UC1_3$-3$PuCl_3$ was suggested for the fuel and 60NaCl-40UCl_3 (% by moles) for the blanket.

There are very serious material challenges in chloride systems, particularly at high temperature. Experimental data for individual metals show very poor corrosion resistance in a molten chloride environment, several orders of magnitude worse than in fluorides. Also the chloride mixtures are less stable chemically than fluorides.

9.2.1.2 Molten salt systems for an improved nuclear fuel cycle

A few reactor and accelerator-driven systems based on molten salt fuel have been proposed since the late 1980s, addressing different objectives.

Molten salts are mostly presented as "nuclear system solutions" not only for transmutation purposes but also for a long term, synergetic development of nuclear power based on introducing Th into the fuel cycle. A few systems are focused on the specific objectives of improving the existing U-Pu fuel cycle by transmuting existing nuclear waste in dedicated accelerator-driven systems.

Furukawa *et al.* proposed a concept of THORIMS-NES ("Thorium Molten-Salt Nuclear Energy Synergetics") [203], composed of:

- A molten-salt reactor [204], without continuous chemical processing or core-graphite exchange.
- Fissile-fuel producers utilising spallation/fission reactions of 1 GeV-proton – Accelerator Molten Salt Breeder (AMSB): ASO-series [205].
- Pyro-processing plants.

CEA has put some efforts into the concept of a thorium fuelled accelerator-driven sub-critical system for both energy production and TRU- incineration, called "TASSE" (**T**horium based **A**ccelerator-driven **S**ystem with **S**implified fuel cycle for long term **E**nergy production) [206], designed for nuclear energy production with reduced radiotoxicity in the waste. Tasse takes advantage of the Th fuel as a feed (breed) material and a good neutron economy in the so-called super-thermal spectrum which is possible in molten salt cores or designs like a high temperature reactor.

Since the early 1990s, Ch. Bowman has been proposing different molten salt systems for Accelerator-driven Transmutation of Wastes (ATW) with a special emphasis on non-proliferation aspects and incinerating weapon Pu [207-214]. Bowman's 2-stage molten salt system, so-called Tier-1 and Tier-2, is aimed to simplify and improve nuclear fuel cycle with an eventual goal of sustained nuclear energy without serious proliferation concerns and without advanced reprocessing of spent fuel.

AMSTER – Actinides Molten Salt TransmutER concept – a continually reprocessed molten salt critical reactor, moderated with graphite and burning TRUs on a support of uranium or Th with various levels of enrichment, has been proposed by Vergnes *et al.* [23]. The AMSTER reactor design has benefited from a detailed analysis of transmutation performance in different systems and especially in a conventional type of molten salt reactor.

Finally, the Kurchatov Institute in Moscow developed a concept of the cascade sub-critical molten salt reactor (CSMSR) to optimise the nuclear fuel cycle [215]. The cascade, divided into fast and thermal neutron zones, allows the accelerator power to be ten times less than in other ADS-concepts.

9.2.1.3 Thorium molten-salt nuclear energy synergetics – THORIMS-NES

Furukawa *et al.* have worked for over 20 years and published a number of papers [203-205] on a molten salt reactor concept and its capabilities for sustainable nuclear energy generation with a significant potential for Pu-incineration. The principles of THORIMS-NES concept were:

- Using thorium as a breeding nuclear fuel.
- Application of molten fluoride fuel technology.
- Separation of breeder reactor plants (MSB, based on advanced technologies for external neutron sources like spallation or even fusion) from power generating reactors (MSR).

The first stage of the THORIMS-NES concept has produced pre-conceptual designs of Small Molten Salt Reactors, as shown in Table 9.1.

Fuji Molten Salt Reactor concepts are based on the MSRE design, having molten salt circulating through a graphite moderator enclosed in a Hastelloy reactor vessel. The design was focused on achieving graphite lifetime of 30 years, and on-line chemical reprocessing has been limited to removal of volatile fission products.

An Accelerator Molten Salt Breeder AMSB concept presented in 1981 was a rather naive design with an integrated target-fuel molten-salt pool. The main objective for this system was breeding ^{233}U (up to 800 kg/year), incurring a high cost in electric power for the rather unrealistic 1 GeV, 300 mA accelerator.

1. The preliminary assessments of the transmutation potential of Molten Salt Systems indicate that fission products like ^{129}I, ^{135}Cs, ^{151}Sm and possibly ^{90}Sr, ^{93}Zr, ^{126}Sn and ^{137}Cs could be transmuted in the form of molten fluorides. However, there are no really reliable data on effective transmutation rates and requirements for isotopic separations.

2. The potential for TRU-transmutation in the modified FUJI-reactor, called FUJI-IV, in which 1/3 of the ^{233}U inventory is replaced by TRU (up to 200 kg in the core) is claimed to be about 50 kg/yr for a reactor or molten salt ADS of about 300 MWe. A so called Fuji-Pu small molten salt reactor of 250 MWth, specially designed to incinerate Pu, would incinerate about 100 kg Pu per year.

All the data quoted above are based mainly on relatively simple calculations and should be considered rather as an assessment of the potential of a Th-based nuclear fuel cycle phased into the existing LWR fuel cycle. The chemistry of TRU-fluorides is not really well known and all the neutronic calculations must be thoroughly verified with experimental results on solubility limits for Pu and MA-fluorides and their chemical stability.

Table 9.1. **Some characteristics of small molten salt reactors in comparison to MSRE-ORNL**

	Fuji-II	**Mini Fuji-II**	**MSRE-ORNL**
Thermal power (MWth)	350	16.7	7.3
Net electric generation (MWe)	155	7	–
Thermal efficiency (%)	44.3	42.	–
Reactor vessel inner diameter × height (m)	5.5 × 4.1	1.8 × 2.1	1.45 × 2.2
Maximum core diameter (m)	1.4, 3.4 (II zone)	0.6	1.14
Core graphite fraction (vol%)	93, 90 (II zone)	90	77.5
Core-Blanket power density – average/peak (kWth/l)	9.5/17.5	16.4/24.9	2.9/6.6
Neutron flux (10^{14} n/cm^2·s) Max. thermal Max. over 50 keV (damage to graphite)	 8.3 0.8	 0.58 0.75	 0.5 0.3

9.2.1.4 Tier-1 – Tier-2 Molten Salt Transmutation Systems

C. Bowman's concepts depart partially from the Li-Be based fluoride salts in favour of NaF-ZrF_4. A molten salt system called Tier-1 is a once-through accelerator-driven transmuter (see Figure 9.1), while a system called Tier-2 becomes a final burner of actinide remnants from Tier 1 operation. Unlike the Tier-1 system, Tier-2 is based on "conventional" Li-Be fluoride salts.

The goal of Tier-1 is to reach a single-pass remnant near the 10 % level without recycling or back-end chemical reprocessing (see Figure 9.2). In Tier-1 (see Figure 9.3), spent fuel assemblies from an LWR are first converted to fluorides to remove the uranium as UF_6, and facilitate the removal of fission products, and to prepare the rest of the waste for insertion into the transmuter. The primary constituent of the input to the transmuter from the spent fuel is the cladding as ZrF_4. NaF is added to the mixture and the NaF-ZrF_4 becomes a carrier for the actinides and the fission products. The waste flows through the transmuter continuously spending about five years on average in an effective flux of 2-4 × 10^{14} n/cm^2·s. Most of the actinides are burned away and the remnant isotopic composition is uninteresting as weapons material and incapable of supporting a thermal spectrum chain reaction. Without fission product removal at the input, the burn-up factor is 0.33; with fission product removal the burn-up factor improves to 0.217. If this once-through remnant from Tier-1 is geologically stored, there is greatly reduced concern for the repository as a plutonium mine for weapons material or for unused and concentrated nuclear fuel. The risk of spontaneous criticality is much reduced or eliminated and indefinite supervision becomes unnecessary.

Figure 9.1. **Tier-1 molten salt transmuter [214]**

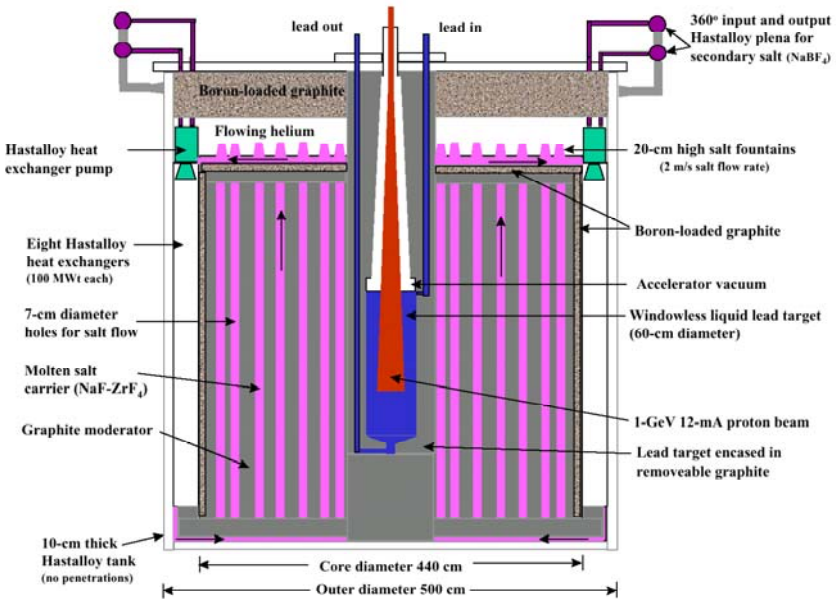
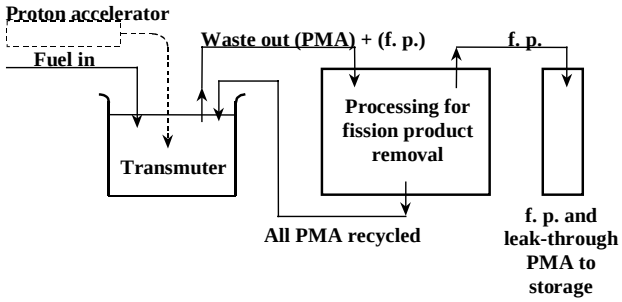


Figure 9.2. **Key feature of the Tier-1 approach [214]**

The conventional transmuter shown in the upper part of the figure requires chemical processing on the back-end to remove fission products and return the TRU for further burning. In the Tier-1 approach, the neutron economy enhancement by the accelerator and the burn-out of the fission products in the thermal spectrum allows a high burn-up without back-end chemical separations.

Conventional approach



New approach

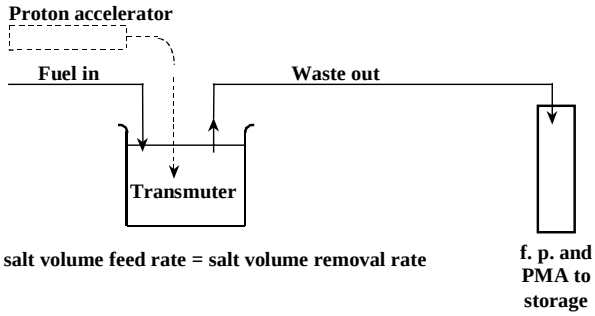


Figure 9.3. **Implementation of the once-through transmuter**

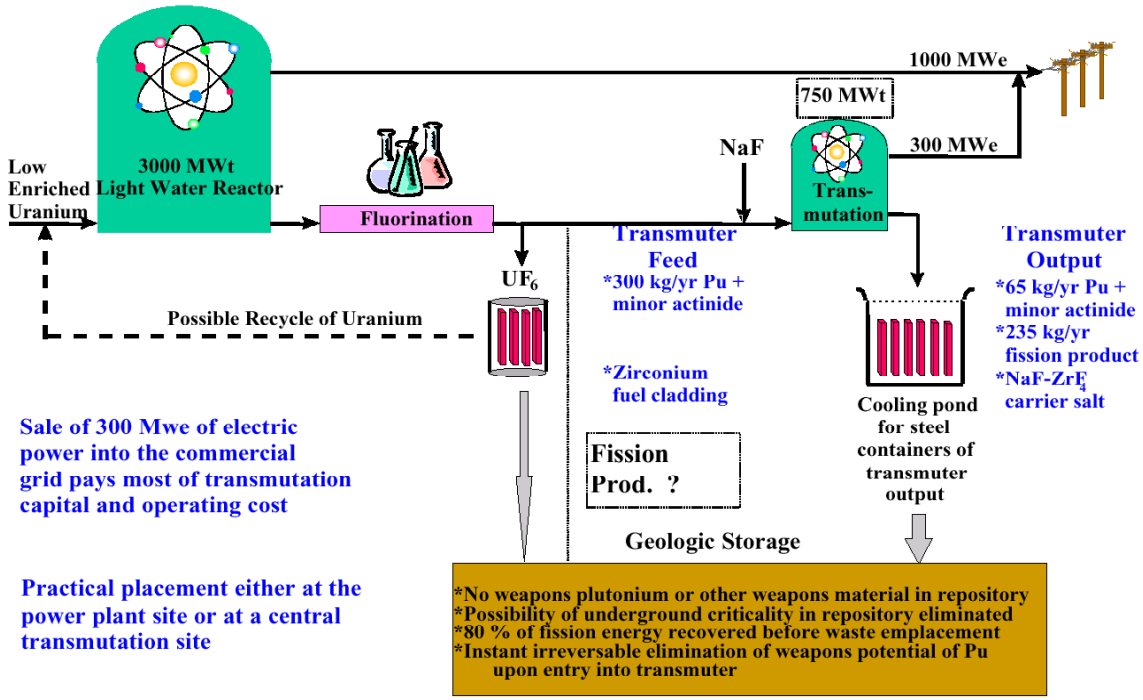
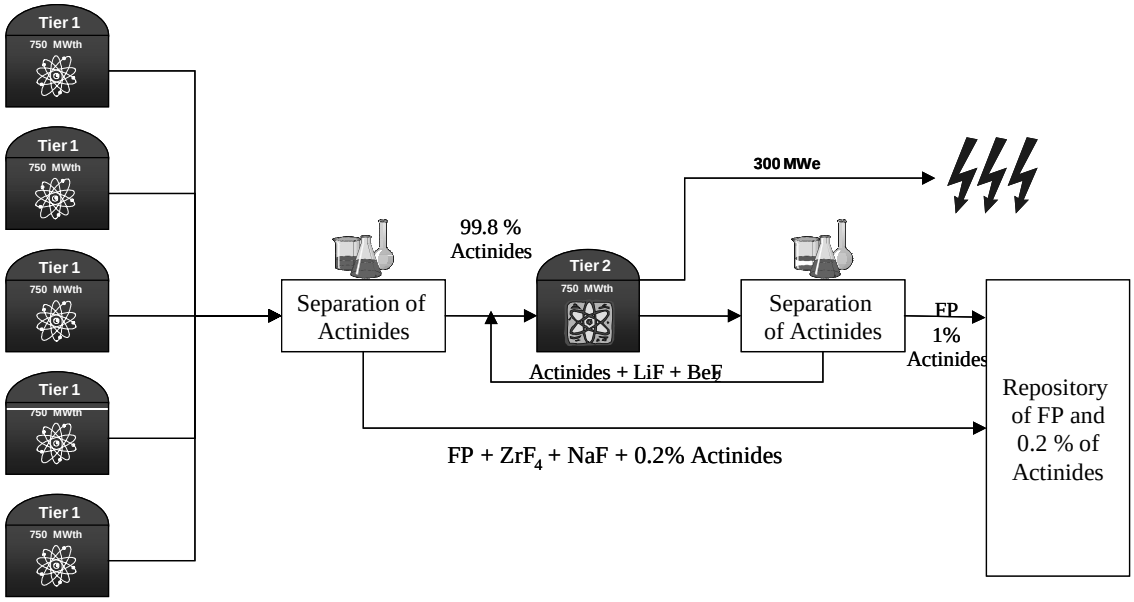


Figure 9.4. **The concept of Tier-2 systems [216]**
For a nuclear park corresponding to French nuclear power, 60 Tier-1 and 12 Tier-2 should be deployed, i.e. Tier-2 can support 5 Tier-1 facilities.



If there is further concern about the remnant actinides from Tier-1, they may be fed into Tier-2 to reduce these isotopes and their radioactivity perhaps by a factor of 1 000 or more. However the Tier 2 (see Figure 9.4) system follows the conventional approach shown at the top of Figure 9.2 and the performance depends on the efficiency of recovery in the chemical separation, which might be 99.9%.

This efficiency is degraded in the recycle process since each time the separation is performed some of the actinide is lost to the fission product waste stream. If the burn-up remnant is 90%, the proportion lost is increased by a factor of nine from 0.001 to 0.009. Bowman claims that the build up of curium and higher elements in his thermal spectrum system is not evident. Results presented in [216] show that Tier-1 may well be an effective burner of ^{239}Pu in a single pass. Although there is a some build up of the higher isotopes of ^{242}Pu and ^{244}Cm, ^{242}Pu is the most stable of the common plutonium isotopes and ^{244}Cm decays with a half-life of 18 years to ^{240}Pu.

The Tier-2 system requires a back-end separations facility and the elimination of fission products from the Tier-1 waste stream before feeding to Tier-2.

The total actinide inventory for the Tier-1 system operating at 750 MW thermal fission power and at a flux of 4×10^{14} n/cm^2·s is 193 kg. The actinide inventory for Tier-2 is 1 126 kg for a unit of 750 MWth power level. Since the fuel dwell time is 3.2 years, the back-end separations rate should be 350 kg/year.

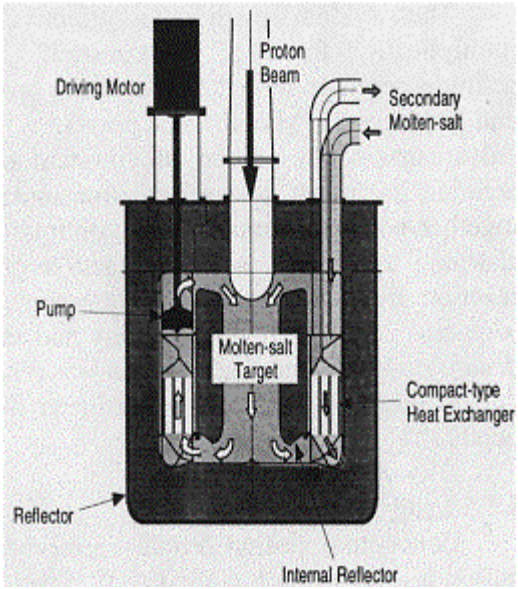
For Tier-2, the remnant waste is mainly ^{244Cm} and ^{246}Cm, but the actinide content of the Tier 2 waste is smaller by a factor of about 400 than in the Tier-1 waste stream. After 1000 years the ^{244}Cm has decayed to ^{240}Pu; most of the ^{246}Cm remains and decays with approximately the same half-life as ^{240}Pu. The alpha decay rate from Tier 2 at 1000 years is 1/1400 of that for unprocessed waste stored for the same period.

9.2.1.5 JAERI molten-salt and molten-alloy ADS concepts

JAERI has conducted conceptual design studies on an 800-MWth molten-salt target/blanket system for a dedicated accelerator-driven nuclear waste transmutation system. Figure 9.5 shows it schematically. A mixture of 64NaCl-5$PuCl_3$-31$MACl_3$ (where MA represents Np, Am, and Cm) has been chosen for the molten-salt system based mainly on the consideration of actinide solubility. Molten chloride is an attractive option since it has high Pu solubility and the mass number of Cl is about twice of that of F. The NaCl-$PuCl_3$ system has an eutectic temperature of 726 K for the composition 64NaCl-36$PuCl_3$. The solubilities of MAs in the salt are not known, but Pu in the chloride salt may be replaced by any minor actinide.

The molten-salt acts at once as fuel-target material and coolant. This significantly simplifies the target/blanket system configuration eliminating the physical and functional separation of target and core. One of the disadvantages of the fast neutron molten salt system is a large actinide fuel inventory. To reduce the primary molten-salt inventory, main pumps and heat exchangers are contained within the primary vessel. In the 800 MW molten salt system with an effective multiplication factor of 0.92 and a 1.5 GeV, 25 mA proton beam, the transmutation rate is approximately 250 kg/y, or 4.6% of the inventory per year assuming a load factor of 80%.

Figure 9.5. **Concept of JAERI's molten-chloride ADS [217]**



Another candidate for a fast-spectrum molten salt system is $PbCl_2$-$AnCl_3$ (where An refers to an actinide). Table 9.2 compares the parameters of the both systems. The difference seems not to be large, but the results are not conclusive owing to the lack of reliable property data for these salts.

The major issue of the molten-salt concept is the compatibility of structural materials exposed to a high temperature flowing chloride salt.

Table 9.2. **Comparison of sodium-based salt and lead based salt systems**

Target/Coolant	**Molten-chloride salt**	
Salt	$64NaCl$-$36AnCl_3$	$70PbCl_2$-$30AnCl_3$
k_{eff}	0.93	0.88
Proton Beam (GeV)	1.5	1.5
Neutrons per proton	37	40
Average neutron energy (keV)	800	768
Power density (keV/cm^3/p) max/ave	66/27	54/16
Power peaking	2.5	3.5
Primary system volume (m^3)	2.7	3.2
Molten-salt/Actinide inventory (kg)	10 000/5 400	17 000/4 100

* Averaged over target/core region, excluding IHX region.

Molten alloy ADS concept

Molten actinide alloy could be a possible alternative to molten salt as a liquid target/fuel for a fast neutron transmutation system. An evolutionary concept was studied by Katsuta *et al.* [218] in order to achieve a minimum inventory of fissile material by this means. Figure 9.6 shows a concept of an ADS

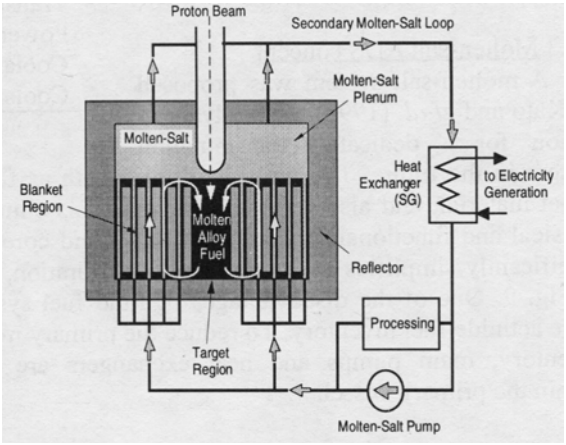
with a molten-alloy target/fuel, a graphite blanket with vertical coolant channels, and an upper plenum of molten fluoride salt. The preliminary study was performed on the alloy with the composition (11-32.5)Np-(4-12.5)Pu-24Co-(60-30)Ce-Tc.

The secondary molten fluoride salt (Li_2BeF_4) directly contacts with the molten-alloy through the vertical channels in the blanket. Efficient heat, mass and momentum transfer is expected at the contact interface between two co-current fluids. This eliminates the need of primary molten-alloy pumps and heat exchanger hardware.

The system with the effective neutron multiplication factor of 0.9 transmutes 145 kg of actinide per year with 1.5 GeV-16 mA proton beam and produces 455 MW thermal power.

Advantages of the molten-alloy system are a small actinide inventory and a high transmutation rate, together with the possibility of continuous on-line charging of minor actinides and removal of reaction products. The system, however, poses problems of material compatibility and safety. The design study of fluid-fuel systems was halted owing to safety concerns of about reduced defence-in-depth.

Figure 9.6. **JAERI's concept of molten salt ADS [217]**



9.2.1.6 TASSE-concept

The TASSE concept, a sub-critical nuclear system with simplified front and back end fuel cycles based on Th and with mobile fuel, pursues the following goals [206]:

- Simplifying the fuel cycle through:
  - Elimination of fuel enrichment.
  - Elimination or significant reduction of recycling through "on-line" technology without separation of TRU and fission products.
- Significant reduction of long-lived nuclear wastes due to both the negligible TRU-production and the high fuel burn-up.
- Burning out TRU from the current Nuclear Power park (PWR's) without toxic long-lived wastes on the way to the complete replacement of PWRs by TASSEs.

- Practically inexhaustible fuel resources for future NP:
  - Significant potential enhancement of deterministic safety due to stable reactivity and sub-criticality.

TASSE is not yet specified in technical details and following engineering solutions are under consideration:

- For the fast neutron spectrum option:
  - Molten salt compositions 30ThF4 + 25NaF + 45PbF2 with fuel density 7 t/m3 at 600°C.
  - Fuel burn-up: ~25-30% h.a. in the equilibrium once-through cycle.
  - A constant level of sub-criticality during core life due to the continuous feed-discharge regime of the fuel.
- In the super-thermal spectrum option:
  - Molten salt (32ThF4 + 14NaF + 54LiF) or HTR-type fuels (with a graphite to fuel proportion 4000/1 as optimal for HTR fuel).
  - Once-through fuel cycle.
  - Constant level of sub-criticality taking advantage of quasi-continuous feed-reloading regime for HTR-type fuel.

Figure 9.7 [219] presents the conceptual scheme for transition from present nuclear system to a TASSE system, using TRU to start the new TASSE system.

Two fuel cycles are possible for the TASSE concept, once-through with no processing of irradiated fuel and a version with on-line separation of fission products (see Figure 9.8).

The essential feature of the TASSE concept is the choice of the appropriate burn-up. The main idea is to choose the burn-up for the once-through cycle, tailored to an “optimal” value of the core reactivity, kept approximately constant over all the cycle. This can be achieved through an optimised choice of the fuel salt avoiding too large epithermal component or through an appropriate choice of the flux level.

In a sense the sub-criticality level in TASSE-system is adjustable to fuel enrichment. TASSE gives significant gains in terms of radiotoxicity reduction compared to PWR fuel cycle, up to an order of magnitude for the long term [219].

9.2.1.7 AMSTER

AMSTER is a new design of a molten salt critical reactor intended to burn TRUs with a uranium or Th support. AMSTER is based on the same salt composition as MSBR, i.e. 61LiF-21BeF_2+18AcF_4 (Ac stands for actinides). The concept is now under continuous development and can be considered as revisiting the principles of ORNL molten salt reactors but focused on incineration of TRU instead of breeding.

Figure 9.9 shows the conceptual layout of the AMSTER reactor with a working temperature range of 550°C-800°C and pumps placed within the graphite core.

Figure 9.7. **TASSE system in transition from LWRs and U-fuel cycle**

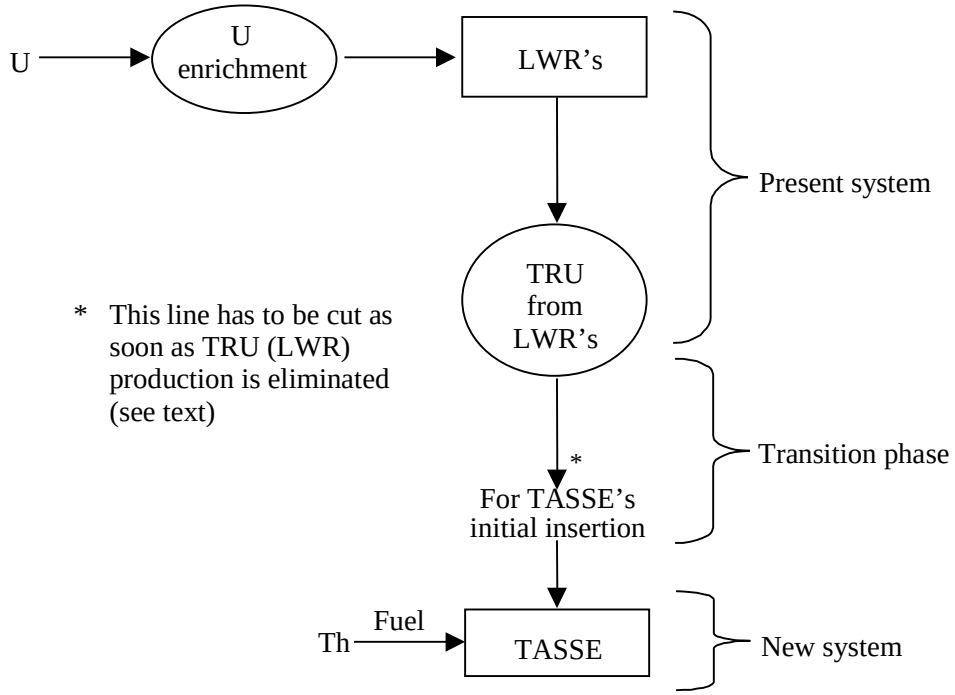


Figure 9.8. **Fuel cycle options for TASSE at equilibrium stage**

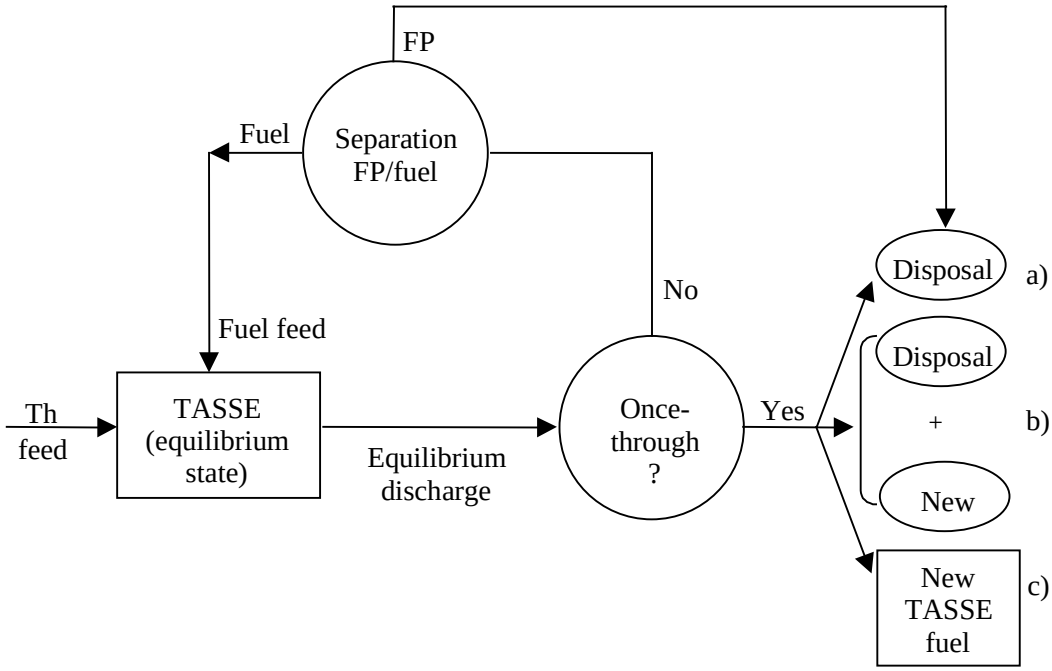
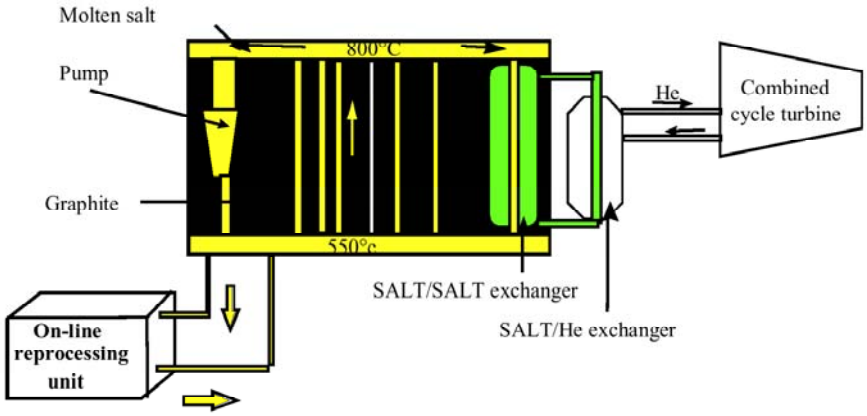


Figure 9.9. **A concept of the AMSTER reactor [220]**



Several fuelling options are envisaged for AMSTER:

- Core at equilibrium: based on spent PWR fuel with continuous addition of $^{235/238}$U or Th.
- TRU incinerator: core in which the TRU inventory is higher than that of the LWR spent fuel and kept constant by continually adding ^{235}U/Th and TRUs.

Table 9.3 presents the main characteristics of AMSTER with enriched U fuelling. Table 9.4 shows characteristics of the more advanced AMSTER reactor with two zones and Th feed.

There are two efficiency criteria for a TRU incineration strategy, namely:

- Minimising TRU losses per TWhe.
- Minimising the residual inventory in the cycle after e.g. 60 year decay.

AMSTER shows a very good performance compared to other transmutation systems [23]. It may be concluded that AMSTER promises a low TRU inventory with relatively small losses, owing to less degradation of the isotopic quality of the TRUs and above all to the contribution of highly fissile ^{235}U (in a system with U-feed) allowing a deep burnup of the molten salt fuel.

AMSTER may be considered as an effective critical transmutation system.

Table 9.3. **Main characteristics of AMSTER with U fuelling**

Load	**Core at equilibrium**	**TRU-incinerator**
^{235}U enrichment (%)	2.45	3.3
TRU content	2.6	4.7
TRU inventory in active core per GWe(t)	1.19	2.1
TRU inventory in reactor per GWe(t)	1.9	3.3
TRU consumption per TWhe (kg)	0	12
^{235}U consumption per TWhe (kg)	65	60

Table 9.4. **Characteristic of the 2-zone AMSTER with a Th-feed**

Ratio of volumes salt fertile/salt fissile	**1.5**	**2**	**2.5**
Inventory in the core (kg/GWe)			
^{232}Th	138 800	153 580	167 360
Uranium	3 354	3 650	3 900
Transuranium	53	54	54
Consumption of ^{233}U (kg/TWhe)	2.11	-0.12	+1.91
Core radius (m)	4.95	5.04	5,11
Volume of the salt in the reactor (m^3)	82	91	99
Power of the fissile zone (MWe)	1 895	1 800	1 714

9.2.1.8 Cascade Sub-critical Molten Salt Reactors (CSMSR)

The CSMSR concept is based on four main ideas [221]:

- Molten-salt fuel.
- Accelerator-driven sub-critical reactor.
- Cascade scheme in the sub-critical reactor.
- Non-aqueous methods of reprocessing the nuclear fuel.

CSMSR is envisaged to close the fuel cycle of LWRs.

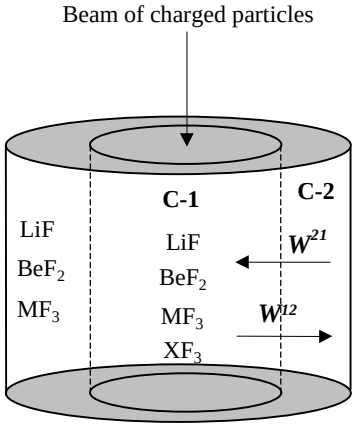
For the operation of a standard sub-critical reactor with $k_{eff} = 0.95$ and thermal power 1 GW, an external neutron source is required with an intensity of $\sim 10^{17}$ n/s.

The advantages of the sub-critical systems in combination with MSR are stated for CSMSR as:

- Reactivity accidents are excluded.
- Absence of instabilities, typical for critical MSR.
- Effective control of the reactor.
- Surplus neutrons that can be used for transmuting fission products and actinides.

Figure 9.10 shows a very schematic layout of the cascade sub-critical reactor with molten salt fuel. A beam of charged particles from the accelerator-driver spalls primary neutrons in C-1. These neutrons are multiplied by a factor $n \sim 1/\Delta k_{eff} \sim 20$ at sub-criticality $\Delta k_{eff} = 0.05$. In C-2 the number of such neutrons is amplified to $N \sim n^2 W_{12}$, where $W_{12} \sim 0.4$ is the probability to initiate fission in C-2 by neutrons from C-1. The number of fissions in C-2 per primary neutron in C-1 is $N_f \sim (n^2/\nu) W_{12} \sim 100$ at $k_{eff} = 0.95$. Besides the salt composition (LiF, BeF_2, MF_3), where M are minor actinides (Cm, Np, Am, etc., plus U, Pu and Th) C-1 contains also salts XF_3 of thermal neutron absorbers X (Gd, Sm) which depress the probability W21 of neutron penetration from C-2 to C-1.

Figure 9.10. **A very schematic layout of the CSMSR. C-1: a fast core, $k_{1\infty}$>1 – first cascade; C-2: a thermal core, transmutation zone $k_{2\infty}$ <1 – second cascade; M-actinide; X-thermal neutron's absorber.**



To provide the condition W21~0 in the considered scheme of CSMSR it is enough to introduce Gd at a proportion of 10^{-3} into the molten salt of C-1. Such admixture provides the necessary value W_{21}~10^{-2} - 10^{-3} and decreases the number of neutrons by only ~0.5%. In the traditional scheme of the sub-critical reactor with k_{eff} = 0.95 and thermal power 1 GW the accelerator is required to have a beam power of 50 MW. The cascade scheme allows the accelerator power to be reduced by a factor of 10 without loss in safety, and electron accelerators to be used, instead of proton accelerators. In the first case the accelerator consumes ~1%, and in the second ~10% of the electric power generated by CSMSR.

Table 9.5 presents data for different TRU in the equilibrium state of Nuclear Power Parks (NPP) of various structures with recycling of all actinides. It is assumed that these systems are fed with 1 000 kg/year of heavy atoms (^{238}U, ^{235}U) to give a power of 1 GWe. In the ideal case only fission products would be discharged and buried – about 1 000 kg/year.

Table 9.5. **Amount of TRU in equilibrium closed fuel cycles for thermal (LWR) and molten salt reactors (MSR), tons/GWe**

	LWR	**75% LWR + 25% MSR**
Neutron flux – average over cycle, n/cm^2·s	10^{14}	10^{14} (LWR), $5\cdot10^{15}$ (MSR)
^{237}Np	0.72	0.12
Pu (total)	5.8	3.1
^{241}Am + ^{243}Am	0.76	0.08
Cm (total)	1.38	0.09
TRU (total)	8.7	3.4
TRU without Pu	2.88	0.30
Heavy nuclides (total)	265	283

Table 9.6. **Amount of TRU in equilibrium closed fuel cycles for thermal (LWR), fast (FR) and molten salt reactors (MSR), t/GWe**

	FR	**89% FR + 11% MSR**	**51% LWR + 38% FR + 11% MSR**
Neutron flux – average over cycle, n cm^{-2} s^{-1}	10^{15}	10^{15} (FR), 5×10^{15} (MSR)	10^{14} (LWR), 10^{15} (FR), $5 \cdot 10^{15}$ (MSR)
^{237}Np	0.11	0.02	0.02
Pu (total)	21.1	18.0	10.4
^{241}Am + ^{243}Am	0.77	0.10	0.19
Cm (total)	0.19	0.04	0.11
TRU (total)	22.2	18.2	10.7
TRU without Pu	1.10	0.17	0.3
Heavy nuclides (total)	121	117	221

Having considered an overall neutron economy for the fuel cycle one can conclude that for a NPP of LWR's (with recycling of all actinides), the feed consists of 250 kg/year of ^{235}U and 750 kg/year of ^{238}U. For this system the equilibrium amount of MA reaches 2.88 t/GWe. All MA should be recycled in MSR. In this case the equilibrium amount of MA is reduced to 0.3 t/GW(e) and the ^{235}U feed to 200 kg/year, but the total amount of heavy nuclides in the fuel cycle is increased.

For 3-component NP ("triple strata") system (LWR+FR+MSR) one can really close the fuel cycle with ^{238}U feed only (1 000 kg/GWe per year) – see Table 9.6. In this self-sustaining NP system all surplus neutrons will be spent on actinide transmutation or on incinerating the long-living fission products – ^{129}I and ^{99}Tc, and the equilibrium amounts of MA and Pu are minimal.

The equilibrium amount of TRU can be further reduced only by feeding with ^{232}Th (about 80%) and ^{238}U (about 20%). It is possible to reduce the amount by a factor of 5 and maintain the necessary neutron balance without an external neutron source as has been shown for the AMSTER case.

Table 9.7 presents the various TRU inventories in a quasi-equilibrium fuel cycle for a MSR-burner taking all Pu and MA unloaded from LWRs of VVER type after the first fuel irradiation cycle.

Two cases have been considered:

- Spent fuel is unloaded and immediately after reprocessing TRU comes to the MSR.
- Unloaded fuel is stored for 20 years, then after reprocessing TRU come to MSR.

Preliminary calculations for a MSR of power 1 GWe show that neutron economy in both cases is sufficient not only for stable operation in critical conditions but for incineration of some long-lived fission products (^{129}I and ^{99}Tc). Actinide amounts in both cases are not large, but in intermediate storage the total reaches 20 t/GWe.

In the second case neutron economy is decreased by 0.1 n/fission but remains sufficient for MSR operation without an external neutron source.

Table 9.7. **Amount of TRU in quasi-equilibrium fuel cycle for molten salt reactor (MSR) fed with MA and Pu from LWR with or without intermediate storage (IS), t/GWe**

	MSR fed directly from LWR, without IS	20 years in IS	MSR fed from IS
^{237}Np	0.015	0.81	0.016
Pu (total)	0.81	17.7	0.83
^{241}Am +^{243}Am	0.062	1.42	0.084
Cm (total)	0.22	0.03	0.24
TRU (total)	1.11	20.0	1.18
TRU without Pu	0.30	2.26	0.35
Heavy nuclides (total)	1.11	20.0	1.18

9.2.2 Modular helium reactor and accelerator-driven transmuter with a particle bed fuel – MHR and MHA

A modular helium reactor and accelerator-driven transmuter with TRISO coated ceramic particle fuel has been proposed by General Atomics and is under serious investigations for a simplified, effective scheme of nuclear waste transmutation. In this transmutation scheme, the thermally fissile isotopes are destroyed in a critical but passively safe Gas-turbine Modular Helium Reactor, or GT-MHR, followed by a deep burn-up phase in an accelerator-driven GT-MHA [222,223].

The main idea is to take advantage of the unique feature of TRISO fuel particles, namely:

- Possibly very large burn-ups over 60%.
- Flexible design of TRISO particles for diverse waste destruction requirements (variation of kernel diameter).
- Slow, progressive burn.
- Fast destruction.
- As a final waste form, spent TRISO fuel in graphite blocks can be permanently stored without further processing.

The MHR/A based transmuter system concept has few important components necessary for high transmutation performance. The transmutation system consists of 2 strata: reactor stratum (MHR, so-called Tier-1) and accelerator-driven stratum (MHA Tier-2). Both MHR and MHA have heterogeneous cores with two distinct regions: driver region and transmutation region (see Figures 9.11 and 9.12). Driver regions are loaded with “driver” fuel (DF), containing mainly Pu + Np coming from UREX processes. ^{240}Pu and ^{237}Np in this fuel provide negative prompt feedback. TRISO-coated DF is engineered for 70% burn-up. Dimensions of kernel and fuel block are chosen to provide steady reactivity for 240-days. DF fuel is irradiated for 3 × 240-day periods in the MHR.

Transmutation regions are loaded with “transmutation” fuel containing Am + Cm coming from the UREX process, and TRU coming from a single reprocessing step of spent DF.

Fresh TF is neutronically positive, i.e. it acts as fuel not poison. TRISO-coated transmutation fuel (TF) is engineered to withstand 100% burn-up, with TRISO particles sized so as to provide high destruction rates (no resonance self-shielding). TF fuel is irradiated for 3×240-day periods in the MHR (thermal spectrum with neutrons of 0.01-1 eV) and then up to 6×240-day periods in the MHA (epithermal neutron spectrum of 1-10 eV).

Transmutation is performed in few steps with only two reprocessing phases, the front end UREX reprocessing of LWR spent fuel, and one single reprocessing of TRISO particles to recover TRU-elements and convert them into transmutation fuel – TF (see Figure 9.13). The LWR spent fuel is processed by the UREX process. Pu and Np are formed into "Driver" fuel elements". Higher Actinides (Am, Cm) are formed into "Transmutation" fuel elements. Transmutation fuel fabrication is highly challenging because of the higher actinides are volatile.

Long-lived fission products, Tc and I, are isolated and fabricated into special transmutation elements. It is suggested that Tc and I can be irradiated and transmuted in existing LWRs with minimal impact on operations.

Short-lived fission products, especially Sr and Cs, are isolated and stored in special waste-forms designed to deal with the intense short-term heat generation of the short-lived isotopes. Uranium goes to low level waste.

Driver and transmutation fuel elements are loaded into the MHR-bT critical core (the first stratum – Tier-1). The MHR core is divided into three Regions: Reflector, Driver and Transmutation Regions. The Driver Region (annulus) is divided into three zones of equal volume, and driver fuel (Np, Pu) is loaded into the driver region starting from the innermost zone (see Figure 9.11). The Transmutation Region (inner core) is also divided in three zones of equal volume and transmutation fuel (MA) is loaded in the transmutation region, starting from the outermost zone (see Figure 9.12). Residence of driver and transmutation fuel in the MHR core is three 240-day periods. In the driver region, after each period of operation, the driver fuel is moved outwards to the next zone and new fuel is added in the innermost zone; axial shuffling is also performed, to improve burn-up uniformity. The driver fuel is discharged after it has reached the outermost driver zone and processed to extract the fission products and remaining Pu, Np, MA are added to the transmutation fuel mix.

In the transmutation region, after each period of operation, the transmutation fuel is moved inwards to the next zone and new transmutation fuel added in the outermost zone. After reaching the innermost transmutation zone, the transmutation fuel is discharged. It is suitable for further irradiation in the sub-critical MHA (the second stratum – Tier-2) and no further reprocessing of the TF particles is needed.

The MHA fuel blocks are different in c/f ratio from the MHR fuel, assuring operation in the epithermal neutron spectrum. The MHA core is sub-critical, operating at k_{eff} = 0.7 - 0.8, and a power multiplication of ~10. Like the MHR core it is also divided into three zones of equal volume. Fuel is loaded in the core, starting from the outermost zone, and fuel shuffling is as in the MHR. After irradiation, fuel is sent for final packaging and repository storage.

In the transmutation region, after each period of operation, the transmutation fuel is moved inwards to the next zone and new transmutation fuel added in the outermost zone. After reaching the innermost transmutation zone, the transmutation fuel is discharged. It is suitable for further irradiation in the sub-critical MHA (the second stratum – Tier-2) and no further reprocessing of the TF particles is needed.

Figure 9.11. **Core layout of the MHR-system – Tier-1 Strata.**
Neutrons generated in the MHR Driver (outer) Region from "fresh" spent fuel, rich in Pu, drive the transmutation of the waste actinides in the Transmutation (inner) Region. In the Driver Region, the fuel to graphite ratio and particle size are engineered to provide slowly changing reactivity. In the Transmutation Region, the fuel to graphite ratio and particle size are engineered to provide fast burn rates [222].

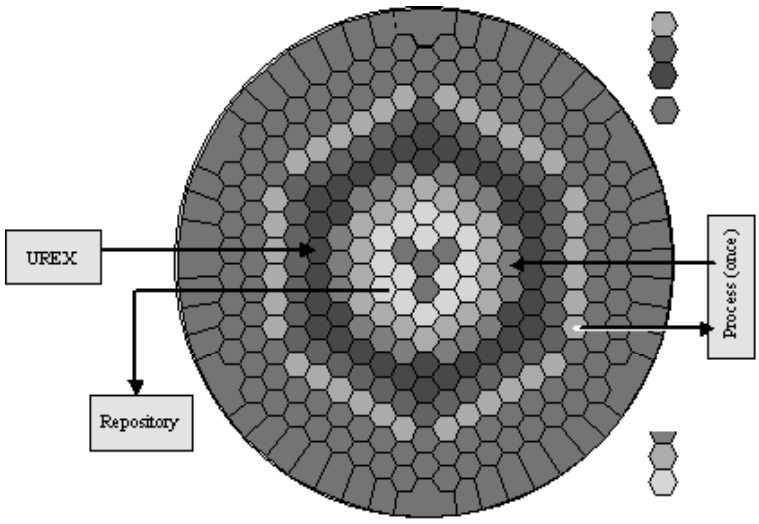


Figure 9.12. **Core layout of the MHA-System – Tier 2 strata.**
Neutrons generated in the spallation target drive the final destruction of the waste actinides in MHA through a progressively harder neutron spectrum (1-10 eV range). In the Outbound Track, going towards softer spectrum, remaining fissile isotopes are reduced. In the Inbound Track, going towards harder spectrum and target neutrons, remaining TRU is deep burned.

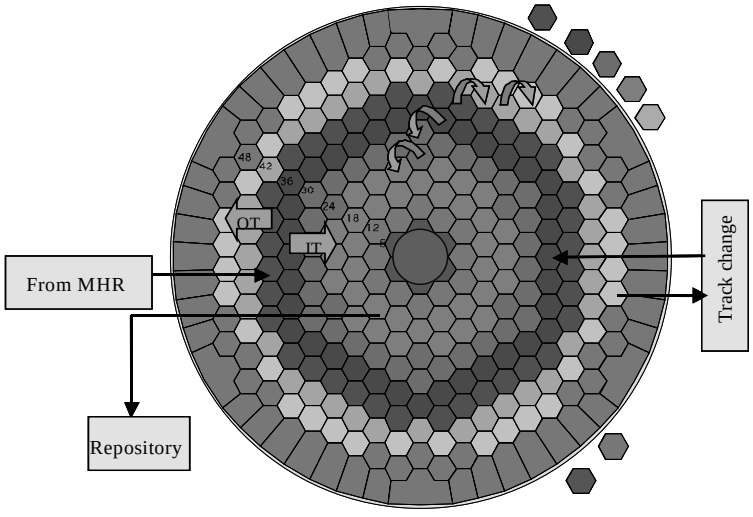
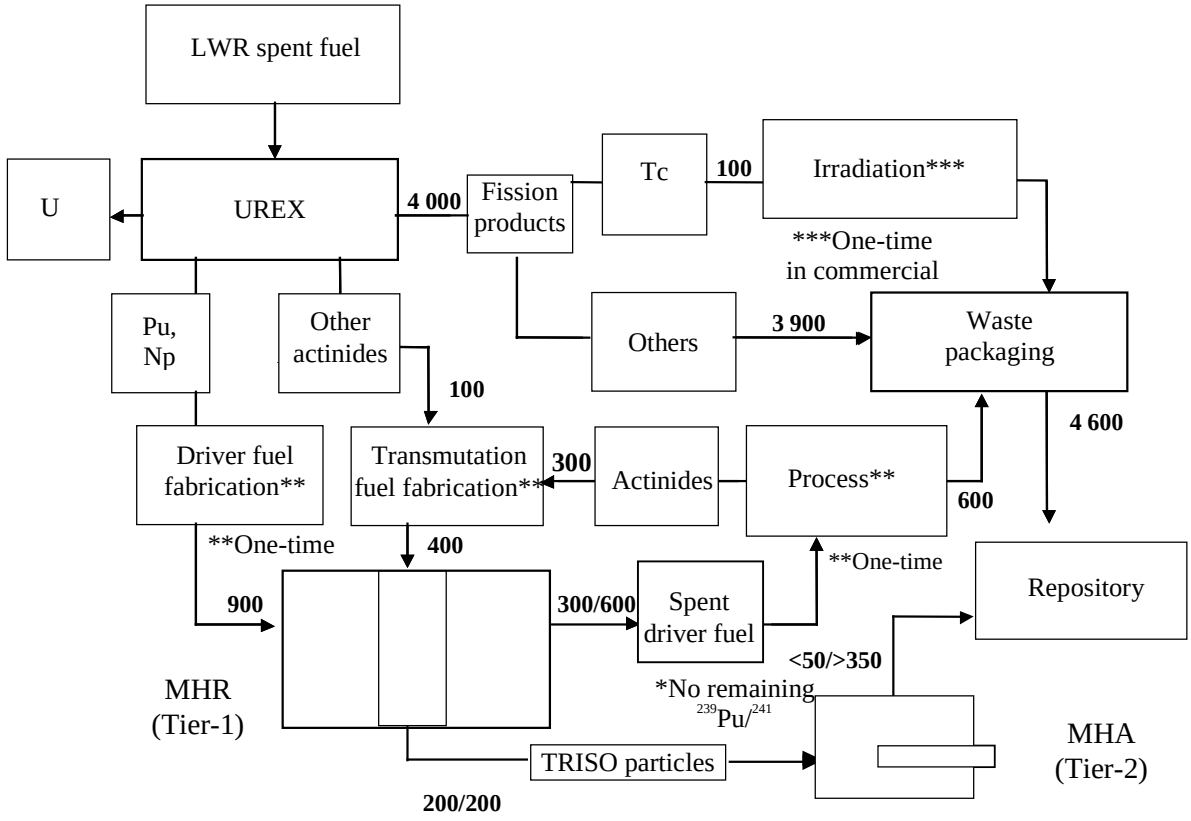


Figure 9.13. **A schematic fuel cycle for MHR/A transmutation system. Material balance in kg/y. One MHA would serve 4-5 MHRs [222] (flows in kg/y)**



The MHA fuel blocks are different in c/f ratio from the MHR fuel, assuring operation in the epithermal neutron spectrum. The MHA core is sub-critical, operating at k_{eff} = 0.7 - 0.8, and a power multiplication of ~10. Like the MHR core it is also divided into three zones of equal volume. Fuel is loaded in the core, starting from the outermost zone, and fuel shuffling is as in the MHR. After irradiation, fuel is sent for final packaging and repository storage.

In the latest General Atomics concept a unit park of the MHR/A system comprises 4 × 600 MWth MHRs and one 600 MWth MHA, all designed on the basis of a commercial HTGR reactor with competitive cost/performance features. Table 9.8 gives a preliminary comparison of ALMR and MHR/A performance.

In conclusion it can be said that the MHR/A transmutation concept has very attractive features. In the first stratum 75% TRU transmutation can be achieved and then after a single reprocessing an accelerator-driven MHA increases burn-up to 95% of the initial TRU inventory. These deep burn-up levels are achieved with no plutonium reprocessing and at a much higher rate than in a corresponding fast reactor ALMR based system. This is made possible by encapsulating the waste to be transmuted in coated ceramic microspheres that accommodate large amounts of fission products in spherical expansion volumes.

It remains, however, to be shown that spent TRISO particles are really suitable as a final waste form and that burn-up performance can really reach the limits assumed in the calculations.

Table 9.8. **Comparison of transmutation performances of ALMR and MHR/A systems**

	ALMR* (3 000-MWth parks) each park is 6 modules	**MHR/A (4 × 600-MWth R-units + 1 × 600-MWt A-unit)**
TRU initial	700 t	700 t
TRU inventory (per unit or park, Core + Process)	12 000 + 4 000 = 16 000 kg	3 000 + 200 = 3 200 kg
Breeding ratio	0.76	0.0
TRU destruction rate (75% avail.)	200 kg/y	1 000 kg/y
TRU destruction ratio (rate/inventory)	1.5 %/y	30 %/y
Decontamination factor	200	200
TRU loss rate to processing/ fuel fabrication	40 kg/y	1 kg/y
Campaign time to destroy 700 t	~3 500 park-y	~700 park-y
Residual TRU-inventory:		
– TRU waste to repository	3.5 t	35 t
– TRU lost in processing	~100 t	~1 t
– TRU in last unit	12 t	~0 t
Total	**115.5 t**	**36 t**

9.3 Conclusions

Attention should be paid to a category of actinide transmutation systems using alternative technologies, e.g. thermal neutrons and liquid fuels. However, most of these should be seen in a very long-term perspective as the respective fuel and reprocessing technologies as well as the systems themselves are essentially based on even more advanced concepts, compared with the solid-fuel/fast-spectrum systems on which the present study is focussed. This applies especially to the molten salt systems which, from a sustainability viewpoint, appear to be the most interesting systems in this category.

10. CONCLUSIONS

10.1 Introduction

The principle of sustainable development requires the fuel cycle of future nuclear energy systems to be closed for plutonium as well as minor actinides to ensure the production of fission energy with limited amounts of natural resources (i.e. uranium) and long-lived radioactive waste. It also requires a safe and cost-effective nuclear energy production. The resource efficiency and waste reduction goals together can ultimately only be reached by the introduction of advanced reactor systems with a significant fraction of fast reactors. For well-known reasons, however, a massive substitution of existing LWR-based, by such advanced, reactor and fuel cycle technology is not a realistic near-term scenario.

Partitioning and transmutation (P&T), which could address the high-level radioactive waste issue now and prepare the ground for a more resource-efficient nuclear energy system in the future, may become an attractive and appropriate intermediate strategy on the way to the ultimate goal of the sustainable nuclear energy system. In this context, the accelerator-driven system (ADS) can play an interesting role as a minor actinide or transuranics (TRU) burner. The interest in such burners is, of course, coupled with the P&T system and will diminish with an increase in the fraction of fast reactors in the park because, in a FR-dominated reactor park, dedicated burners will no longer play an essential role.

The scope of the present study comprises the clarification of the roles and merits of the fast reactor and the fast-spectrum accelerator-driven system (fast ADS) with regard to their application as actinide and fission product burners as well as the assessment of the development status of the ADS with emphasis on reactor technology and safety, fuel cycle technology, trends in electricity cost, and general feasibility. By concentrating on transmutation strategies with fully closed fuel cycles, i.e. the particularly effective transmutation strategies, the study complements the P&T status and assessment study published in 1999. The essential differences between the variety of proposals for implementing such strategies are evaluated with the help of a set of representative “fuel cycle schemes”, which are analysed in a consistent manner using reactor and fuel cycle parameters agreed by the Expert Group.

Technical conclusions have already been given at the end of each technical chapter. In this chapter these conclusions are combined and rearranged in agreement with the objectives of the study as follows:

- Role of ADS in actinide transmutation strategies.
- Fuel cycle technology.
- ADS technology and safety.
- Cost of actinide transmutation.
- Fission product transmutation.
- R&D needs.

For the benefit of policy makers, the detailed technical conclusions are preceded by a set of "general conclusions". These combine principal messages from the study with generally known facts which deserve to be re-emphasised on this occasion. However, the results and merits of the study can only be fully comprehended by consulting the following detailed technical conclusions.

10.2 General conclusions

Principal messages which could influence policy decisions are:

- While P&T will not replace the need for appropriate geological disposal of high-level waste, the study has confirmed that different transmutation strategies could significantly reduce, i.e. a hundred-fold, the long-term radiotoxicity of the waste and thus improve the environmental friendliness of the nuclear energy option. In that respect, P&T could contribute to a sustainable nuclear energy system.
- Very effective fuel cycle strategies, including both fast spectrum transmutation systems (FR and/or ADS) and multiple recycling with very low losses, would be required to achieve this objective.
- Multiple recycle technologies that manage Pu and MA either together or separately could achieve equivalent reduction factors in the radiotoxicity of wastes to be disposed. The study shows that pyrochemical reprocessing techniques are essential for those cycles employing ADS and FRs where very high MA-content fuels are used.
- In strategies where Pu and MA are managed separately, ADS can provide additional flexibility by enabling Pu-consumption in conventional reactors and minimising the fraction of dedicated fast reactors in the nuclear system.
- In strategies where Pu and MAs are managed together, the waste radiotoxicity reduction potential by use of FRs and ADS is similar and the system selection would need to be made based on economic, safety and other considerations.
- Further R&D on fuels, recycle, reactor and accelerator technologies would be needed to deploy P&T. The incorporation of transmutation systems would probably occur incrementally and differently according to national situations and policies.
- Fully closed fuel cycles may be achieved with a relatively limited increase in electricity cost of about 10 to 20%, compared with the LWR once-through fuel cycle.
- The deployment of these transmutation schemes need long lead-times for the development of the necessary technology as well as making these technologies more cost-effective.

10.3 Technical conclusions

The following sections list the detailed conclusions for the different technical areas.

10.3.1 Role of ADS in actinide transmutation strategies

- All transmutation strategies with closed fuel cycles could, in principle, achieve high reductions in the actinide inventory and the long-term radiotoxicity of the waste, and these are comparable with those of a pure fast reactor strategy. With respect to these reductions, the potentials of the FR and the ADS are very similar.

- Under the assumptions used in the study, these strategies can achieve a more than hundred-fold reduction in the long-term waste radiotoxicity and even higher reductions in the heavy metal and TRU losses to repository, compared with the once-through fuel cycle.
- The reduction factors are primarily determined by the fuel burn-up and the reprocessing and fuel fabrication losses. An ambitious goal for the recovery of all actinides (99.9 %, as already achieved for uranium and plutonium) must be set, if the quoted reduction factors are to be realised.
- Multiple recycling of plutonium without minor actinide transmutation is useful for the management of plutonium, but cannot qualify as a transmutation strategy because it reduces the long-term waste radiotoxicity by only a factor of about five.
- With regard to actinide waste production and technological aspects, the TRU burning in FR and the double strata strategies are similarly attractive. The former can gradually evolve to a pure fast reactor strategy, but requires high initial investment in fast reactor and advanced fuel cycle technology. The latter confines the minor actinides to a small part of the fuel cycle, but calls for particularly innovative technology for this part of the fuel cycle.
- The sub-critical operation of an actinide burner with a fast neutron spectrum offers interesting additional parameters of freedom in the core design. In particular, the possibility of operating such a burner with a uranium-free (or thorium-free) fuel supply allows the fraction of specialised transmuters in the reactor park to be minimised.
- A further advantage of the sub-critical operation mode is the tolerance of the system against degradations in the safety characteristics of the core. Both of these advantages are of particular relevance for systems which burn pure minor actinides, e.g. minor actinide burners in a double strata strategy.
- Transmutation systems with partially closed fuel cycles, e.g. systems in which minor actinides are separated from the fuel and recycled in special “target” pins, are technologically less demanding and do not require an ADS, but cannot achieve the high transmutation effectiveness of systems with fully closed fuel cycles.
- Physical limitations associated with the production and destruction of in-pile and out-of-pile fuel inventories imply very long time constants for the start-up and final shut-down phase of new fission-based nuclear technologies. This implies that transmutation technology, with or without ADS, can fulfil its promises only, if it is introduced with the intention of using it for at least a century.
- For a nuclear energy scenario with a finite time horizon, the full benefit from transmutation can be realised only if, in the shut-down phase, the TRU inventory is burnt and not put to waste. Due to the low power-specific heavy metal inventory of the respective burner, the TRU burning in ADS strategy features a lower steady-state TRU inventory and, in the shut-down phase, can burn this inventory more quickly than the other investigated strategies.

10.3.2 Fuel cycle technology

- Actinide transmutation implies the handling of fuels with very high decay heats and neutron source strengths. A significant effort is required to investigate the manufacturability, burn-up behaviour and reprocessability of these fuels. This applies particularly to fuels with high minor actinide content, which can probably be reprocessed only with the help of pyrochemical methods. These methods have to be further developed to tolerate from ten to

more than twenty times higher decay heat levels than those encountered in the pyrochemical reprocessing of fast reactor fuels.

- The introduction of pyrochemical processing techniques at the industrial level will require the development of new process flowsheets and the use of potentially very corrosive reagents in hostile environments. These processes will generate chemical and radiological hazards which will have to be mitigated.
- The PUREX aqueous reprocessing can be considered as valid for the FR-MOX fuel in the plutonium-burning and double strata schemes. Reprocessing of this fuel within short cooling times and with the required high recovery yields, however, will require the plutonium dissolution yield to be improved and the PUREX flowsheet to be modified.
- Due to the high radioactivity of FR-MOX fuel, its handling will require measures to be taken to reduce the radiation doses in the fabrication plant and during the transportation of the fuel assemblies. The increased requirements for shielding, and preference for short transportation paths, of multiple recycled fuels also favour the pyrochemical reprocessing method.
- All transmutation strategies which include LWRs in the reactor mix produce large streams of depleted and irradiated uranium. If this uranium is not considered as a resource for future fast reactors, its long-term radiological impact has also to be taken into account.

10.3.3 ADS technology and safety

- For all ADS-based transmutation strategies, important technological challenges exist with regard to the accelerator, the target, the sub-critical reactor and new types of safety issues.
- On the whole, the development of accelerators is well-advanced, and beam powers of up to 10 MW for cyclotrons and 100 MW for linacs now appear to be feasible. However, further development is required with respect to the beam losses and especially the beam trips to avoid fast temperature and mechanical stress transients in the reactor.
- Various problems related to the accelerator-reactor coupling have still to be investigated. Thereby, special attention has to be given to the target and especially the beam-window, as these components are subjected to complex stress, corrosion and irradiation conditions which are not encountered in normal reactors.
- While the reactor physics of sub-critical systems is well-understood, the issues regarding the dynamic response to reactivity and source transients require further investigation because they are the area of greatest difference between critical and sub-critical systems.
- The presence of an external neutron source which can vary very rapidly, in combination with very weak reactivity feedbacks, especially from the Doppler effect, implies fast and (depending on the sub-criticality level) large responses to control actions which puts additional demands on the control actuators, the fuel behaviour, and the heat removal processes. In particular, the fuel should be capable of adiabatic heat storage to buffer any sharp changes.
- If hypothetical core disruptive accidents have to be taken into account in the safety analysis of an ADS, a prompt negative feedback mechanism for quenching such accidents has to be developed.

10.3.4 Cost of actinide transmutation

- Fully closed fuel cycles may be achieved with a relatively limited increase in electricity cost of about 10 to 20%, compared with the LWR once-through fuel cycle. In case of partially closed fuel cycles, e.g. only closed for plutonium, the cost increase is about 7%.
- Among the fully closed fuel cycle strategies investigated in the present study, TRU burning in FR and the double strata strategy feature the lowest increases in system-wide electricity cost relative to the LWR once-through fuel cycle.
- Fuel cycle strategies which involve the use of ADS-technology show an overall economic benefit by burning as much plutonium as possible in less-expensive, more-conventional systems, i.e. MOX-LWRs and MOX-FRs.
- Especially the TRU burning in ADS strategy is sensitive to accelerator beam costs. Reducing the accelerator cost by a factor three would halve the electricity cost increase for this strategy.
- The economic incentive to increase the burn-up fraction in the minor actinide and TRU burners beyond a value of 15% becomes marginal. Further reductions in fuel losses to repository at an acceptable system-wide energy costs, therefore, are to be obtained preferentially by increasing the fuel recovery rate.
- For the closed fuel cycle strategies, the advanced technology contribution to the system-wide electricity cost is in the range of 10 to 50%. If all non-LWR technology is considered as advanced, the advanced technology cost contribution for the closed fuel cycle strategies lies in the range of 30 to 50%.
- The cost analysis confirms the long-term potential of P&T for reducing the transuranic inventory and the radiotoxicity of the waste with a rather limited increase in the electricity generation cost, despite that this cost increase may not be acceptable in today's market environment. It also means that transmutation may become affordable with only a limited cost increase, if a reduction in the actinide waste radiotoxicity becomes important for the society.

10.3.5 Fission product transmutation

- Excess neutrons produced by critical and sub-critical burners can, in principle, be utilised to transmute fission products. With the neutron fluxes available in these systems, it is theoretically possible to transmute the long-lived fission products; the transmutation of the more abundant short-lived fission products, however, is impracticable due to insufficient transmutation rates. This means that transmutation, in principle, allows the mitigation of the long-term risk from fission products in a geologic repository, but cannot significantly reduce the heat generation and mass of the disposed fission products.
- Maximising the supplier-to-burner reactor support ratio can result in an insufficient neutronic potential for transmuting the long-lived fission products of the entire reactor park. If the transmutation would be limited to ^{129}I and ^{99}Tc, all TRU burning strategies could, theoretically, accomplish the task.
- In practice, the necessity of isotopic separations and difficulties in the preparation of targets present difficult obstacles for the fission product transmutation, which currently reduce the number of candidate nuclides to only one or two, i.e. ^{99}Tc and, possibly, ^{129}I. So far, the feasibility has been established only for ^{99}Tc. This means that, for the remaining long-lived

fission products, partitioning followed by immobilisation in a specially stable matrix may remain the only realistic method for reducing their radiological impact.

10.3.6 R&D needs

- *Basic R&D* is needed for the new FR and ADS in the fields of nuclear data and neutronic calculations, fuel technologies, structural materials, liquid metals, reprocessing technologies, target materials and high power accelerators (the last two only for ADS).

- *Experimentation on fuels is a priority.* No concept can be considered seriously, if the appropriate fuels are not defined and proven, i.e. characterised, fabricated, irradiated and reprocessed.

  - Since fuels play a central role in all scenarios of waste minimisation and nuclear power development, an international share of efforts around nitrides, oxides and metals should be organised in order to ensure an optimum use of resources in the few existing laboratories which can handle very active fuels.
  - In this context, *the availability of irradiation facilities, in particular fast neutron facilities which can produce high damage rates in the specimens, is a key issue and major concern.* Again, an international initiative could be envisaged to harmonise programmes and to allow the best use of existing resources to be made. Identification of the experimental irradiation needs in such a shared international fast-spectrum facility would be a worthwhile undertaking.

- *Demonstration at appropriate scale of the performance of pyrochemical processes* (level of losses, secondary waste, etc.) is needed in order to assess in more detail the technico-economic viability of certain fuel cycle options.

- In the field of basic R&D supporting FRs as well as ADS, the discussion around the *coolants for fast-spectrum systems* would benefit from a better international agreement on pro and cons of the different options.

- *Improved modelling tools to simulate the materials behaviour* under (mixed) irradiation conditions (and possibly high temperatures) may prove to be a very valuable approach and a sharing of expertise and benchmarking within an international context may be advocated.

- *Safety analysis of ADS* should identify the possible paths to exclude hypothetical core disruptive accidents (HCDA) in ADS. If such a HCDA has to be taken into account in the safety analysis of an ADS, a prompt negative feedback mechanism for quenching such an accident has to be developed.

- In addition to this R&D, countries embarking on an ADS-based fuel cycle strategy should envisage a *demonstration experiment* which allows the ADS concept to be validated from operation and safety viewpoints.

- And last but not least, *Performance assessment studies for a geological disposal site using a P&T source term* are necessary in order to seek clarification of the cost/benefit analysis of such advanced fuel cycles, including geological disposal.

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Annex A

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Annex B

ACRONYMS

ADS	Accelerator-driven system (sub-critical)
ADTF	Accelerator-driven test facility
AGR	Advanced gas-cooled reactor, developed from the Magnox type (UK)
ALMR	Advanced Liquid Metal (-cooled) Reactor
AMSB	Accelerator molten-salt breeder (conceptual)
AMSTER	Actinide molten-salt transmuter
amu	Atomic mass unit
An	Actinide, i.e. U, Pu, Np, Am or Cm. "Actinides" stands for any combination of U, Pu, Np, Am and Cm.
ANL	Argonne National Laboratory (US)
ATW	Accelerator Transmutation of Waste, US programme of partition and transmutation
ATWS	Anticipated transients without scram
BDB	Beyond design basis (of an accident needing emergency measures)
BOEC	Begin of equilibrium cycle
BOL	Begin of life
BOP	Balance of plant (e.g. electric generating equipment)
BU	Burn-up
BWR	Boiling water reactor
CANDU	Canadian reactor type cooled and moderated by heavy water
CAPRA	Fast reactor operated to burn rather than breed plutonium
CCDTL	Continuous current drift-tube linear accelerator
CDB	Cost database
CEA	Commissariat à l'énergie atomique (France)
CER	Cost-estimating relationships
CER-CER	Ceramic in ceramic
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
CIRCE	Circuito Eutettico
CNRS	Centre National de la recherche scientifique
COE	Cost of energy
CONFIRM	Collaboration on Oxide & Nitride Fuel Irradiation & Modelling
CRBR	Clinch River Breeder Reactor
CRIEPI	Combined Research Institute of the Electric Power Industries (Japan)
CRS4	Center for Advanced Studies, Research and Development
CSMSR	Cascade sub-critical molten-salt reactor (with two-stage neutron amplification)
CW	Continuous wave (distinction from pulsed mode)
D&D	Decommissioning and dismantling
DC	Direct (continuous unidirectional) current
DF	Driver fuel
DFR	Dounreay (experimental) fast reactor (UK)
DOE	Department of Energy (US)
DTL	Drift-tube linear accelerator

EBR	Experimental breeder reactor
EdF	Electricité de France
EFFTRA	Experimental Feasibility of Targets for Transmutation (EC)
EFR	European fast reactor concept
ENEA	Ente per le Nuove tecnologie, l'Energia e l'Ambiente
EOEC	End of equilibrium cycle
ETGCFR	Gas-cooled fast reactor
FCA	Fast Critical Assembly (Japan)
FCF	Fuel conditioning facility
FCR	Fixed charge rate
FFTF	Fast flux test facility
FR	Fast reactor (operating at criticality)
FZJ	Forschungszentrum (research centre) Jülich (Germany)
FZK	Forschungszentrum Karlsruhe (Germany)
GeV	Giga-electron-volt (10^9 electron volts)
GWd	Gigawatt-day (usually thermal, i.e. without allowance for conversion losses)
GWe	Gigawatt (electrical)
HCDA	Hypothetical core-disruptive accident
HLW	High-level waste (fission products etc.)
HM	Heavy metal (uranium, plutonium etc), usually as before irradiation
IBA	Ion Beam Applications
IFR	Integral Fast Reactor, directly associated with reprocessing facilities
INFCE	International Nuclear Fuel Cycle Evaluation
INFN	Instituto Nazionale di Fisica Nucleare
IPHI	Injecteur de protons de haute intensité
IPPE	Institute for Physics & Power Engineering
ISTC	International Center for Technical Co-operation (Russia)
JAERI	Japan Atomic Energy Research Institute
JNC	Japan Nuclear Cycle Development Institute
KAERI	Korea Atomic Energy Research Institute
KALLA	Karlsruhe lead laboratory (Germany)
KCR	Known conventional resources
KEK	High Energy Accelerator Research Organization (Japan)
LANL	Los Alamos National Laboratory (US)
LANSCE	Los Alamos Neutron Science Centre
LBE	Lead-bismuth eutectic mixture
LEDA	Low energy demonstration accelerator
LiSOR	Liquid Solid Reaction experiment
LLFP	Long-lived fission product(s)
LMFBR	Liquid metal (cooled) fast breeder reactor
Ln	Lanthanide
LOD	Line of defence
LOFWS	Loss of flow without scram
LOHSWS	Loss of heat sink without scram
LWR	Light water reactor (either pressurised or boiling)
MA	Minor actinides, principally neptunium, americium and curium

MAB	Minor actinide burner
Magnox	A magnesium alloy used as fuel cladding, the fuel itself, or the reactor type using it (UK)
MASURCA	MAquette de SURgénérateur à Cadarache (France)
MCA	Multi-criteria analysis
MDT	Mean down time
MEGAPIE	MEGAwatt Pilot Experiment
MHA	Modular helium accelerator-driven transmuter
MHR	Modular helium (-cooled) reactor
MHz	Megahertz (million cycles per second)
MOX	Mixed oxide fuel, uranium and plutonium unless otherwise specified
MSBR	Molten salt breeder reactor
MSR	Molten salt (fuelled and cooled) reactor
MSRE	Molten Salt Reactor Experiment (US)
MTBF	Mean time before failure
MTR	Material-testing reactor
MUSE	Neutron-multiplying experiment (France)
MW	Megawatt (million watts)
MYRRHA	Prototype multi-purpose accelerator-driven neutron multiplier
NC	Normally conducting
NEA/NSC	Nuclear Energy Agency Nuclear Science Committee
NFC	Nuclear fuel cycle
nsc	Nuclear cycle scheme number in present report
O&M	Operation and maintenance
OFC	Open fuel cycle
OMEGA	Options Making Extra Gains from Actinides (Japanese initiative)
ORNL	Oak Ridge National Laboratory (US)
P&T	Partition and transmutation
PFR	Prototype fast reactor (UK)
POD	Point of departure
PRISM	Advanced fast reactor concept
PSA	Parametric systems analysis
PSI	Paul Scherrer Institute (Switzerland)
PUREX	Fuel reprocessing scheme based on solvent extraction and reduction of plutonium to an inextractable form for separation from uranium
PWR	Pressurised water (cooled and moderated) reactor
RAR	Reasonably assured resources (uranium)
RF	Radio frequency
RFQ	Radio frequency quadrupole
RIAR	Research Institute of Atomic Reactors (Russia)
RVACS	Reactor vessel auxiliary cooling system (?)
SA	Sub-assembly (composite fuel element)
SC	Superconducting
SCK•CEN	Nuclear Research Centre (Belgium)
SCNES	Self-contained nuclear energy system
SINQ	Swiss Spallation Nuetron Source (Spallation Neutronen Quelle) (Switzerland)
SPIN	French programme on partition and transmutation

SPIRE	Spallation and Irradiation Effects (EC)
SSC	Separated sector cyclotron
Sv	Sievert (unit of radiation dose)
TASSE	Thorium-based accelerator-driven system with simplified fuel cycle for long-term energy production
TECLA	Technologies, Materials and Thermal-Hydraulic for Lead Alloys (EC)
TERM	Test experiment at the Riga mercury loop
TESLA	Superconducting electron-positron collider
TF	Transmutation fuel
TOPWS	Transient over-power without scram
TRISO	Triply-coated ceramic particle fuel
TRU	Transuranic elements, i.e. Pu, Np, Am and Cm.
TUI	Transuranium Institute (Germany)
TWhe	Terawatt-hour (electrical), i.e. 10^{12} watt-hours after conversion losses
UOX	Uranium oxide fuel
UREX	Reprocessing scheme designed to recover plutonium by solvent extraction and discard uranium as low-level waste
UTC	Unit total (capital) cost
VIPAC	Vibratory packing of fuel particles into cladding
VVER	Russian pressurised water reactor
XADS	Experimental accelerator-driven system

Annex C

HISTORY OF P&T STUDIES IN OECD/NEA MEMBER COUNTRIES AND INTERNATIONAL ORGANISATIONS

1. *First generation systems studies* on P&T as a new waste management issue were initiated in the 1970s in different OECD/NEA Member countries:

   - In 1973, the Japan Atomic Energy Industry Forum published a report titled "A closed system for radioactivity" [1]. This report pointed out the importance of R&D for P&T of long-lived nuclides as long-term efforts. JAERI started the development of the partitioning process for high-level liquid waste and the design study of a transmutation system in mid 1970s.

   - In the United States, many individual researchers and small groups were conducting studies related to P&T since Steinberg's seminal work in 1964 [2]. Oak Ridge National Laboratory investigated P&T during the 1970s from a theoretical and assessment perspective [3-4]. Claiborne [5] demonstrated in 1972 the neutron-physical feasibility of transmuting "by-product actinides" in LWRs. Argonne National Laboratory (ANL) performed very interesting work on closing the fuel cycle with Pu-recycling and having potential for P&T. Specifically the pyrochemical reprocessing methods were initially developed in ANL and these methods remain of high value in future P&T-schemes. This work by ANL was however almost exclusively based on own funding without specific governmental support.

   - Simultaneously, the German Research Centre of Karlsruhe, the CEA in France and the European Commission at the Joint Research Centre of Ispra [6-7] started a comprehensive theoretical and experimental R&D programme. In France, the Castaing Commission [8] conducted a general investigation in 1981-82 on the different approaches possible in the fuel cycle and included the P&T option as a mandatory route for further R&D. The studies were conducted during about ten years and were summarised in overview reports which showed the complexity of the issue and the discrepancy between the waste management "risk" approach on long-term disposal and the P&T-approach aiming at the reduction of the radiotoxic inventory by recycling long-lived nuclides into fission reactors.

2. Three major "final assessment" reports were published in the late 1970s and early 1980s, which led to following conclusions:

   - The conclusions of the EC programmes on P&T in 1977 and 1983 [6-7] were that the impossibility of total actinide recycling and the impact of the process flowsheets complexity on waste streams were the main limitation of the potential benefits from the proposed P&T scenarios for long-term hazard reduction. Partitioning would become worthwhile as a HLW management scheme if advanced fuel cycles such as recycling of plutonium and MAs through FBRs and LWRs were implemented, provided that the loss factors for fuel isotopes could be kept very low ($<5 \times 10\text{-}4$). Transmutation of MAs was considered theoretically feasible from the point of view of neutron physics and fuel cycle technology but it was not obvious whether the potential long-term risk reduction for the waste disposal site compensates the increase in short-term risks for the workers and the environment.

   - Taking into account the potential long-term hazard associated with the disposal of spent fuel, the Castaing report (France) in 1982 concluded that it was worthwhile to investigate the benefits of advanced reprocessing techniques with separation and conditioning of Pu and MAs for intermediate storage and tentatively for destruction by neutron irradiation. This

long-term programme is to be conducted simultaneously with investigations of the waste disposal technology in experimental underground facilities.

- The ORNL studies in 1977 and 1980 [3-4] concluded that there were no cost or safety incentives P&T of actinides in High-Level Waste (HLW) for waste management purposes since the long-term risk is mainly associated with long-lived fission products ^{99}Tc and ^{129}I and not with the actinides. The reduction of the radiotoxic inventory of waste is theoretically possible but needs the development of advanced partitioning methods and the use of other types of reactors than the available LWRs.

3. The period of active investigation on P&T starting in early 1970s was terminated around 1982-1983 as no international consensus was obtained on the benefits of P&T as an alternative or complementary waste management option.

4. During the 1980s, a growing awareness of the inherent difficulties in creating and licensing large nuclear waste repositories, and growing delays in the repository R&D projects, particularly in the development of underground pilot repository facilities, led the international community to reconsider the potential benefits of P&T as a complementary waste management option and these resulted in second generation system studies. This renewed interest was also based on technological developments in several fields making the P&T option seemingly more feasible.

5. In October 1988, the Japanese government by way of the Atomic Energy Commission (AEC) launched the ambitious "OMEGA" R&D programme [9]. The R&D programmes were stimulated by the collaborative efforts of JAERI and the former PNC (now JNC). In the public sector, CRIEPI has also been carrying out R&D on this subject. The "OMEGA" programme is proceeded in two steps: the phase-I was intended to cover a period up to about 1996, and the phase-II to about 2000. The basic studies and tests were to be conducted in the phase-I, and engineering tests of technologies or demonstration of concepts are planned in the phase II. The first check and review of the phase-I of the programme by the Atomic Energy Commission was started in February 1999. After 2000, pilot facilities would be built to demonstrate the P&T technology. Following items are being studied:

   - Physical and chemical properties of MAs and FPs.
   - Partitioning of radioactive elements from high-level liquid waste by reprocessing process and recovery of useful metals.
   - Nuclear and fuel property data of MAs.
   - System design studies.
   - Reactor fuel and accelerator target.
   - Development of high power accelerator for transmutation.

6. It was during this second era of P&T activities that the NEA became involved in studying this subject. In 1988, next to launching the "OMEGA" programme, the Japanese government also invited the international community, through the OECD/NEA, to participate in the assessment of a broad range of P&T developments. This initiative was the starting point of a world-wide renewal of interest and work in the P&T field. Large scale R&D programmes are still being conducted in Japan (JAERI, JNC, CRIEPI) and in France (CEA) in co-operation with several European countries under sponsorship of the European Commission. Important experimental programmes were conducted in the United States at the Argonne National Laboratory (ANL).

7. As a result of this increasing interest, the need was felt to re-examine the validity of the P&T option in the light of the more recent results. In France, a National Evaluation Commission was appointed in 1993 in order to supervise the R&D activities in the field of radioactive waste management. Reports were issued [10-12] in 1995, 1996 and 1997. In the field of P&T, the following recommendations were made:

   - Priority should be given to separation of Am-Cm from rare earths followed by Am/Cm separation.
   - Among the fission products priority should be given to Cs and Tc.
   - On the subject of transmutation a distinction should be made between short-term projects based on transmutation in present PWRs and long-term R&D on future reactor systems e.g. fast reactors and accelerator-driven transmutation.
   - Two options (partitioning-transmutation and partitioning-conditioning) should be studied at the same level of priority and a priority listing of the critical radionuclides should be made for each option.
   - The separation processes DIAMEX and SESAME should be demonstrated as soon as possible in the hot facility ATALANTE.
   - Accelerator-driven transmutation is a new venture, which should be studied on the national level within a co-ordinated CEA-CNRS-EDF R&D effort (GEDEON).

8. In Japan, the ongoing "OMEGA" project covered the activities on P&T where comparable national evaluation and assessment reports have not been openly published. However, the Japanese evaluations and assessments have been included in the OECD/NEA activities and publications as part of the NEA assessment studies.

9. A series of American reports was published in the meantime. On the basis of the ORNL retrospective assessment of P&T [13], the Electric Power Research Institute (EPRI) started a detailed evaluation programme [14] on the concept of transuranic burning using liquid metal reactors (LMR) and included, in their overview, the waste management consequences resulting from "alternative spent fuel separation processes". A study of the impact of P&T on the disposal of high-level waste was prepared by Lawrence Livermore National Laboratories [15] and the main conclusions of these US reports were:

   - The toxicity of high-level waste during the first thousand years cannot be reduced by transmutation since the cross-sections of the isotopes ^{90}Sr, ^{137}Cs, ^{3}H and ^{85}Kr are too small.
   - The cost of alternative reprocessing in order to reduce the actinide content to a level below 100 nCi/g (3 700 Bq/g) is very high and requires the construction of advanced aqueous reprocessing facilities and/or the development and construction of pyrochemical reprocessing units.
   - The use of LMRs for burning plutonium and actinides would require the construction of an aqueous reprocessing capacity of ~2 000 tHM/year and the deployment of 30 GWe LMR capacity creating a cost penalty of $0.5 billions to $2 billions per year.
   - The decentralised structure of the US electricity production, the absence of economic incentive for reprocessing and the changes in the regulatory requirements (NRC and EPA) for disposal facilities would make the acceptance of P&T as a waste management scenario very improbable under the then present economic conditions.

10. The most recent published and most comprehensive national assessment report on P&T was issued in 1996 by the National Academy of Science of the US under the chairmanship of N.C. Rasmussen [16]. The report covers all aspects of the problem from an American point of view. The principal recommendations listed in the report are:

    - None of the P&T system concepts reviewed eliminates the need for geological disposal.
    - The current policy of the "once-through-cycle" should be continued.
    - Fuel retrievability should be extended to ~100 years.
    - R&D should be conducted on selected topics of P&T.

11. Since the beginning of the 1990s, an emerging interest has been oriented towards renewed P&T technologies, e.g. accelerator-driven systems (ADS) and pyrochemical partitioning, which induced new R&D activities in several OECD/NEA Member countries. Especially ADS has been the attraction pole for many new researchers in the field and new international collaborations are being set-up in this domain. Those OECD/NEA Member countries conducted in addition studies on the P&T potential and giving overviews of national and international R&D activities in this field. This growing community of researchers in different OECD/NEA Member countries (in Europe about 250 researchers) published multiple reports on P&T during the past five years, where an overview of all these is out of the scope of this note.

12. The IAEA assessment report on P&T in 1995 [17] investigated the technical feasibility and the radiological impact. Conclusions indicated that partitioning is indeed feasible but considerable R&D would be required to implement a realistic flowsheet operable at industrial scale. The reduction in long-term risks achievable by P&T of actinides is less than expected and long-lived FPs which are not amenable to any form of P&T, also contribute to the very long-term risk. All in all, the implementation of P&T would be an immense undertaking, involving a large proportion of a country's nuclear power program, but providing at best a rather small reduction in potential long-term radiological hazard. The IAEA undertook several complementary activities with respect to OECD/NEA's work:

    - A survey of research activities related to P&T in non-OECD countries was undertaken upon recommendation by a Technical Committee Meeting and the report was published in 1997 [18].
    - Participants of a Special Scientific Programme on "Use of High energy Accelerators for Transmutation of Actinides and Power Production" held in Vienna in 1994, in conjunction with the 38th IAEA General Conference recommended the IAEA to prepare a status report on ADS. The general purpose of the status report was to provide an overview of ongoing development activities, different concepts being developed and their status, as well as typical development trends in this area and to evaluate the potential of this system for power production, Pu burning and transmutation of minor actinides and fission products. The document [19] includes the individual contributions by experts from six countries and two international organisations.
    - Other activities involve Co-ordinated Research Projects (CRP) on the potential of Th-based fuel cycles to constrain Pu and to reduce long-term waste toxicities examining the different fuel cycle options in which Pu can be recycled with Th to get rid of the Pu, or replace the Pu with materials that are less unacceptable to the public.

- A Technical Committee Meeting was organised on the feasibility and motivation for hybrid concepts for nuclear energy generation and transmutation where programmes and concepts on ADS development were presented [20].

13. The European Commission was partly supporting research work on partitioning and transmutation of radioactive waste under the Fourth Framework Programme (1994-1998). This work included nine research projects. Five strategy studies were evaluating the capabilities of various burners and fuel cycles to limit the production and even destroy the stock of actinides (plutonium and minor actinides). Two experimental projects were aimed at developing techniques for the chemical separation of actinides and two others were dealing with the investigation of transmutation of americium and long-lived fission products. Within the Fifth Framework Programme (1998-2002), strategy studies on P&T are foreseen to investigate its benefits and compare different methods such as critical and sub-critical systems taking into account the whole fuel cycle. New efficient and selective processes will be developed for the separation of the critical long-lived radionuclides form high level and medium level waste. Basic nuclear data essential for transmutation and the development of ADS will be measured and computed. The radiation damage induced by spallation reactions in materials will be investigated. It is foreseen to develop and test fuels and targets for actinide and long-lived fission product incineration. The preliminary study of an ADS is also considered in the programme with supporting research work on sub-critical mock-ups, safety, coolants, the confinement of the accelerator/reactor window and high power accelerators. Finally, new specific matrices could be also developed for the conditioning of long-lived radionuclides, which cannot be transmuted.

14. The European Commission (EC) published in 1997 a report on the perspectives and the deemed costs of P&T [21]. Main conclusions in this report were the potential reduction of waste radiotoxicity by a factor of 40 to 100 compared with the open fuel cycle scenario and depending on the moment considered in the cooling period. Recycling the FPs was reported not to entail any gain on their radiotoxicity where neptunium recycling results in a gain after roughly one million years of decay. Nevertheless, in terms of residual radiotoxicity, recycling these elements may be an advantage because of their mobility in a geological repository environment. For the first level of P&T, based on technologies derived from existing techniques, the cost supplement over recycling plutonium alone was estimated at about one-third of the fuel cycle cost. Partitioning and fuel fabrication accounting roughly equal fractions of this cost supplement. At the second level, where P&T is implemented based on completely new technologies and aiming at complete separation of the MAs and some FPs, the partitioning involves an additional cost estimated at half the cost of the conventional fuel cycle operations.

15. Today, several national projects and bilateral or multilateral programmes are being undertaken. The most important projects involved are:

    - The Japanese “OMEGA” Programme is currently ongoing and the activities cover the development of a wet partitioning process, design study of an actinide burner reactor (ABR) and an ADS, the development of nitride fuel cycle technologies, and basic research such as nuclear data and fuel property data measurements. Development of a high-intensity proton linac has been carried out under the Neutron Science Project of JAERI which aims at construction of a superconducting proton linac of 8-MW for a 5-MW spallation neutron source for a neutron scattering facility and for an ADS experimental facility.
    - Besides the European countries own national projects and the EC Fifth Framework programme, a trilateral activity was launched by France, Italy and Spain. The Advisory Group and Technical Working Group on Accelerator-driven Systems, chaired by Prof. Rubbia, aims to investigate the potentialities of ADS for the transmutation of waste and to co-ordinate between governmental

agencies and industrial bodies. Their prime objective however is to construct a demo ADS-plant on a 10 years time schedule. Some other European countries joined this informal initiative in order to exchange information and create a European Network.

- The ATW-programme in the US has recently been reviewed by US-DOE and a roadmap for developing this technology has been published in October 1999 [22]. This roadmap proposes a six-year science-based R&D Programme to be established in order to reduce the technical risks and to assess the technical viability of the ATW technology. The total cost of this six-year R&D Programme amounts to $281 M
- The ISTC-framework has included specific projects related to ADS-technology and especially technological issues (Pb-Bi technology, …).

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Annex D

DOSE CONVERSION FACTORS BASED ON ICRP-1990 RECOMMENDATIONS

Nuclide	Sv/Bq	Nuclide	Sv/Bq	Nuclide	Sv/Bq	Nuclide	Sv/Bq
Rb^{86}	2.8 E-09	I^{129}	1.1 E-07	Pb^{209}	5.7 E-11	U^{232}	3.3 E-07
Rb^{87}	1.5 E-09	I^{131}	2.2 E-08	Pb^{210}	6.8 E-07	U^{233}	5.0 E-08
Sr^{89}	2.6 E-09	I^{135}	9.3 E-10	Pb^{211}	1.8 E-10	U^{234}	4.9 E-08
Sr^{90}	2.8 E-08	Cs^{134}	1.9 E-08	Pb^{212}	5.9 E-09	U^{235}	4.6 E-08
Y^{90}	2.7 E-09	Cs^{135}	2.0 E-09	Pb^{214}	1.4 E-10	U^{236}	4.6 E-08
Y^{91}	2.4 E-09	Cs^{136}	3.0 E-09	Bi^{210}	1.3 E-09	U^{238}	4.4 E-08
Zr^{93}	2.8 E-10	Cs^{137}	1.3 E-08	Bi^{212}	2.6 E-10	Np^{237}	1.1 E-07
Zr^{95}	8.8 E-10	Ba^{135m}	4.5 E-10	Bi^{213}	2.0 E-10	Np^{238}	9.1 E-10
Nb^{93m}	1.2 E-10	Ba^{140}	2.5 E-09	Bi^{214}	1.1 E-10	Np^{239}	8.0 E-10
Nb^{95}	5.8 E-10	La^{140}	2.0 E-09	Po^{210}	2.4 E-07	Np^{240}	8.2 E-11
Mo^{99}	7.4 E-10	Ce^{141}	7.1 E-10	Fr^{223}	2.3 E-09	Pu^{238}	2.3 E-07
Tc^{99}	7.8 E-10	Ce^{143}	1.1 E-09	Ra^{223}	1.0 E-07	Pu^{239}	2.5 E-07
Ru^{103}	7.3 E-10	Ce^{144}	5.2 E-09	Ra^{224}	6.5 E-08	Pu^{240}	2.5 E-07
Ru^{105}	2.6 E-10	Pr^{143}	1.2 E-09	Ra^{225}	9.5 E-08	Pu^{241}	4.7 E-09
Ru^{106}	7.0 E-09	Pr^{144}	5.0 E-11	Ra^{226}	2.8 E-07	Pu^{242}	2.4 E-07
Rh^{105}	3.7 E-10	Nd^{147}	1.1 E-09	Ra^{227}	8.4 E-11	Pu^{243}	8.5 E-11
Rh^{106}	1.6 E-10	Pm^{147}	2.6 E-10	Ra^{228}	6.7 E-07	Pu^{244}	2.4 E-07
Pd^{107}	3.7 E-11	Pm^{148}	2.7 E-09	Ac^{225}	2.4 E-08	Am^{241}	2.0 E-07
Ag^{111}	1.3 E-09	Pm^{148m}	1.8 E-09	Ac^{227}	1.1 E-06	Am^{242}	3.0 E-10
Cd^{113}	2.5 E-08	Pm^{149}	9.9 E-10	Ac^{228}	4.3 E-10	Am^{242m}	1.9 E-07
Cd^{115m}	3.3 E-09	Pm^{151}	7.3 E-10	Th^{227}	8.9 E-09	Am^{243}	2.0 E-07
In^{115}	3.2 E-08	Sm^{147}	4.9 E-08	Th^{228}	7.0 E-08	Cm^{242}	1.2 E-08
Sn^{123}	2.1 E-09	Sm^{151}	9.8 E-11	Th^{229}	4.8 E-07	Cm^{243}	1.5 E-07
Sn^{125}	3.1 E-09	Sm^{153}	7.4 E-10	Th^{230}	2.1 E-07	Cm^{244}	1.2 E-07
Sn^{126}	4.7 E-09	Eu^{154}	2.0 E-09	Th^{231}	3.4 E-10	Cm^{245}	2.1 E-07
Sb^{124}	2.5 E-09	Eu^{155}	3.2 E-10	Th^{232}	2.2 E-07	Cm^{246}	2.1 E-07
Sb^{125}	1.1 E-09	Eu^{156}	2.2 E-09	Th^{234}	3.4 E-09	Cm^{247}	1.9 E-07
Sb^{126}	2.4 E-09	Eu^{157}	6.0 E-10	Pa^{231}	7.1 E-07	Cm^{248}	7.7 E-07
Sb^{126m}	3.6 E-11	Tb^{160}	1.6 E-09	Pa^{233}	8.7 E-10		
Te^{123}	4.4 E-09			Pa^{234}	5.1 E-10		
Te^{125m}	8.7 E-10						
Te^{127}	1.7 E-10						
Te^{127m}	2.3 E-09						
Te^{129m}	3.0 E-09						
Te^{132}	3.7 E-09						

Annex E

COMPARISON OF MA TRANSMUTATION EFFECTIVENESS IN DIFFERENT FUELS AND COOLANT SYSTEMS

The MA transmutation characteristics (in terms of transmutation effectiveness) in conventional MOX-type fuel, sodium-cooled fast reactor cores were described in detail in the OECD/NEA P&T Phase 1 report "Status and Assessment Report of Actinide and Fission Product Partitioning and Transmutation" (1999) [1]. Recently, a feasibility study for different fuels and coolant systems was performed in Japan. The objective of the feasibility study was to establish FR and related fuel cycle technologies with the following targets:

- Economic competitiveness as an energy production system.
- Effective utilisation of uranium resources.
- Reduction of radioactive waste.
- Security of non-against proliferation.

In this addendum, the MA transmutation effectiveness in FRs with different types of fuels and coolants is compared quantitatively.

E.1 Fuel-types

Three types of FR fuel, oxide, nitride and metal were considered. In this kind of comparison, it is desirable to make a consistent and fair evaluation. Since MA transmutation in power FRs is assumed here, the performance as power generation system should be made equivalent for each fuel-type core. The core parameters fixed here were: reactor thermal power, operation cycle length, refuelling batch number, spent fuel burn-up, core height and blanket thickness. The design criteria which must be satisfied from a realistic viewpoint are: reactivity at the end of equilibrium cycle, positive burn-up reactivity loss, maximum linear heat rating of fuel pin, and pressure drop at fuel bundle.

Figure E.1 shows the 1 000 MWe-class MOX-fuelled FR core as the reference of the study. Fuel-types changed were $(U,Pu)O_{1.98}$ as oxide, $(U, Pu)^{15}N$ as nitride, and U-Pu-10Zr as metal fuel. The core specifications of each fuel-type core are summarised in Table E.1.

The MA composition to be loaded is assumed to come from the reprocessing of LWR-spent fuel, 35 GWd/t, after 5-year cooling, i.e.:

$^{237}Np/^{241}Am/^{242m}Am/^{243}Am/^{243}Cm/^{244}Cm/^{245}Cm = 49.1\%/30.0\%/0.08\%/15.5\%/0.05\%/5.0\%/0.26\%$

Table E.2 shows the evaluated MA transmutation effectiveness of each fuel-type core where MA was loaded at 5 weight% of total fuel. The total transmutation effectiveness of nitride and metal-fuelled cores is 9.9% and 9.7% per year, respectively, both a little better than that of oxide-fuelled core. The difference can be attributed to the harder neutron spectrum of the new fuel-type cores. Figure E.2 shows the dependence of MA transmutation on loading with MA for each fuel-type core. The slight superiority of nitride and metal fuel to oxide can be seen again, but the difference is rather insignificant, compared with the dependencey to on MA loadinged.

Figure E.1. **1 000 MWe MOX-fuelled FR core**

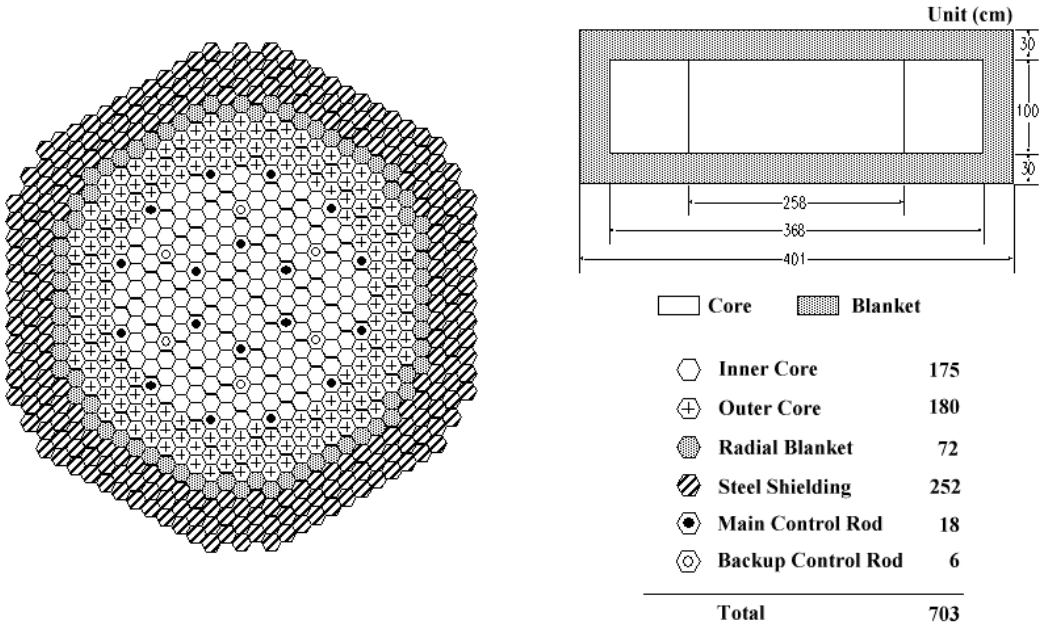


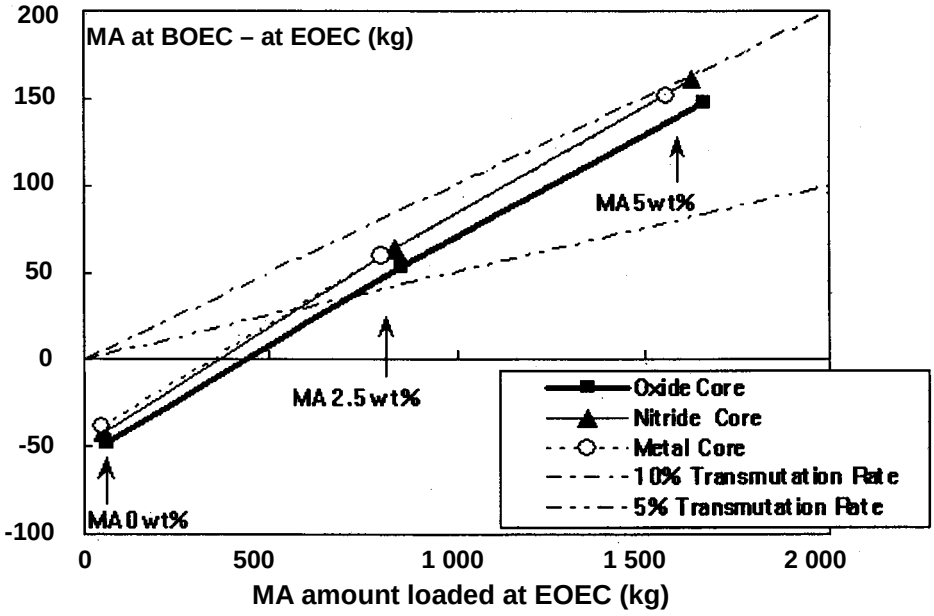
Table E.1. **Major specifications of different fuel-type cores**

Item	**Specification**		
	Oxide core	Nitride core	Metal core
Thermal out-put	2 600 MWth	2 600 MWth	2 600 MWth
Operation cycle length	12 EFPM	12 EFPM	12 EFPM
Refuelling batch number	3 batches	3 batches	3 batches
Core height	100 cm	100 cm	100 cm
Core equivalent diameter	368 cm	346 cm	348 cm
A/B thickness (upper/lower)	30 cm/30 cm	30 cm/30 cm	30 cm/30 cm
Fuel type	$(U, Pu)O_{1.98}$	$(U, Pu)^{15}N$	U-Pu-10Zr
Fuel smear density	87.6%TD	80%TD	75%TD
Fuel pin outer diameter	8.3 mm	8.7 mm	8.5 mm
Cladding thickness	0.4 mm	0.42 mm	0.41 mm
Number of fuel pins per fuel sub-assembly	271	271	271
Fuel sub-assembly pitch	179.8 mm	197.0 mm	198.2 mm
Number of fuel subassembly (inner/outer)	175/180	126/130	126/130
Pu enrichment (inner/outer)	15.3%/19.0%	12.3%/16.2%	12.2%/16.5%
Spent fuel average burn-up rate	73.6 GWd/t	73.2 GWd/t	75.4 GWd/t
Burn-up reactivity loss per cycle	1.96%dk/kk'	0.31%dk/kk'	0.57%dk/kk'
Peak linear heat rate (without 3-dim. Effect)	381 W/cm	530 W/cm	508 W/cm
Breeding ratio	1.22	1.35	1.34
Pressure drop at fuel bundle	3.4 kg/cm^2	2.3 kg/cm^2	2.4 kg/cm^2

Table E.2. **MA transmutation performance of different fuel-type cores (Loaded MA: 5 weight% of total fuel)**

Element	Oxide core			Nitride core			Metal core		
	Transmutation			Transmutation			Transmutation		
	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)
Np	764	112	14.7	756	114	15.1	728	105	14.4
Am	746	72	9.7	728	83	11.4	695	78	11.3
Cm	150	-38	-25.4	148	-36	-24.4	139	-32	-23.1
MA total	1 659	147	8.8	1 631	161	9.9	1 562	151	9.7

Figure E.2. **MA transmutation effectiveness**



E.2 Coolant types

Three types of coolant (sodium, lead and gas) were compared with respect to their impact on MA transmutation effectiveness. Since it is quite difficult to make the parametric comparison strictly due to their different characteristics of density and thermal conductivity, existing typical reactor designs with these coolants were considered in the survey: a sodium-cooled fast reactor of commercial size, a lead-cooled reactor of BREST-300 type [2] and a CO_2 gas-cooled reactor of ETGBR-type [3]. Major design specifications of these cores are summarised in Table E.3. Although there are many differences in design parameters including thermal power, operation cycle length and fuel-type etc. besides coolant type, a rough evaluation of MA transmutation characteristics may be possible by normalising the results with respect to thermal power and operating period. MA composition loaded in this survey is the same as in the fuel-type survey above. Figure E.3 is the comparison of neutron spectrum among

these different coolant-type cores. On the whole, all cores have a fast reactor spectrum with a peak energy around several hundred keVs, where some differences can be found caused by coolant-type. The gas-cooled reactor has the hardest spectrum among the three coolants, because of its very low moderating capability. On the other hand, the lead-coolant core shows special features. Above 1 MeV, the neutron flux reduces owing to the large inelastic cross-section of lead. Below 100 keV, the neutron spectrum is also smaller than that of sodium, as lead shows a lower moderating capability due to the heavy atomic mass. From the comparison of neutron spectrum, a gas-cooled core might be more favourable from a MA transmutation viewpoint, and sodium and lead-cooled cores might be equivalent.

Table E.3 summarises the MA transmutation effectiveness of each coolant-type core. After normalisation, the MA transmutation effectiveness of these cores is almost identical with a value of 7.5-7.7% per year. Figure E.4 shows the dependence of MA transmutation effectiveness on MA loading for each coolant- type after normalisation. The ratio of transmuted MA to loading is a little worse in the case of lead-coolant, but the difference is rather small compared with the dependency to other core parameters like core-fuel inventory which is not directly connected with coolant-types. As a conclusion, the effect of coolant choice in FR design will be negligible from the viewpoint of the MA transmutation.

Table E.3. **Major specifications of different coolant-type cores**

Item	**Specification**		
	Sodium-cooled core (Commercial type)	Lead-cooled core (BREST-300 type)	CO_2 gas-cooled core (ETGBR type)
Thermal out-put	3 800 MW_{th}	700 MW_{th}	3 600 MW_{th}
Operation cycle length	540 days	284 days	344 days
Refuelling batch number	5 batches	5 batches	5 batches
Core height	120 cm	110 cm	150 cm
Core equivalent diameter	457 cm	230 cm	456 cm
Fuel type	Oxide	Nitride	Oxide
Pu vector ($^{238}Pu/^{239}Pu/^{240}Pu/^{241}Pu/^{242}Pu/^{241}Am/^{242m}Am/^{243}Am$)	3/52/27/9.5/1/ 5/0/0	0.5/64/28/3.1/1.7/ 2.1/0.1/0.5	1.9/53/26/9.9/7.9 1.5/0/0
Fuel pin outer diameter (inner/middle/outer)	9.7/-/9.7 mm	9.1/9.6/10.4 mm	8.2/–/8.2 mm
Pu enrichment (inner/middle/outer)	17.8/–/19.8%	14.0/14.0/14.0%	18.7/–/26.7%
Number of fuel pins per fuel subassembly (F/S)	271	114	169
Fuel subassembly pitch	195.4 mm	149.6 mm	180.6 mm
Number of F/S (inner/middle/outer)	264/–/198	57/72/56	334/–/216
Spent fuel average burn-up rate	15.43 GWd/t	62.4 GWd/t	115.1 GWd/t
Burn-up reactivity loss per cycle	2.92%dk/kk'	0.04%dk/kk'	2.35%dk/kk'
Peak linear heat rate (without 3-dim. effect)	370 W/cm	313W/cm	320 W/cm
Breeding ratio	1.04	1.03	1.01
Pressure drop at fuel bundle	3 kg/cm^2	1 kg/cm^2	4 kg/cm^2

Table E.4. **MA transmutation performance of different coolant-type cores (Loaded MA: 5 weight% of total fuel)**

Element	Sodium-cooled core (Commercial type)			Lead-cooled core (Brest-300 type)			CO_2 gas cooled core (ETGBR type)		
	Transmutation			Transmutation			Transmutation		
	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)
Np	1 058	201	19.0	326	28	8.5	961	118	12.3
Am	1 262	148	11.7	364	25	6.8	1 176	81	6.9
Cm	321	–52	–16.2	64	–8	–12.1	228	–34	–15.0
MA total	2 641	297	11.3	754	45	5.9	2 366	165	7.0
Normalised MA transmutation	695 kg per GWth	53 kg per GWth per year	7.6% per year	1 077 kg per GWth	83 kg per GWth per year	7.7% per year	657 kg per GWth	49 kg per GWth per year	7.5% per year

Figure E.3. **Comparison of neutron spectrum among different coolant type cores**

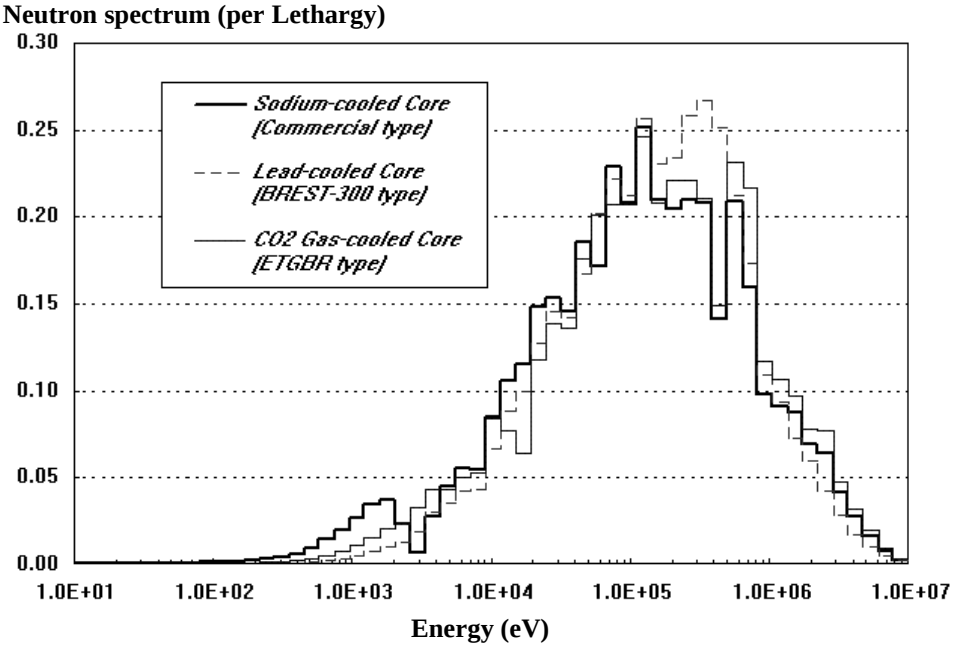
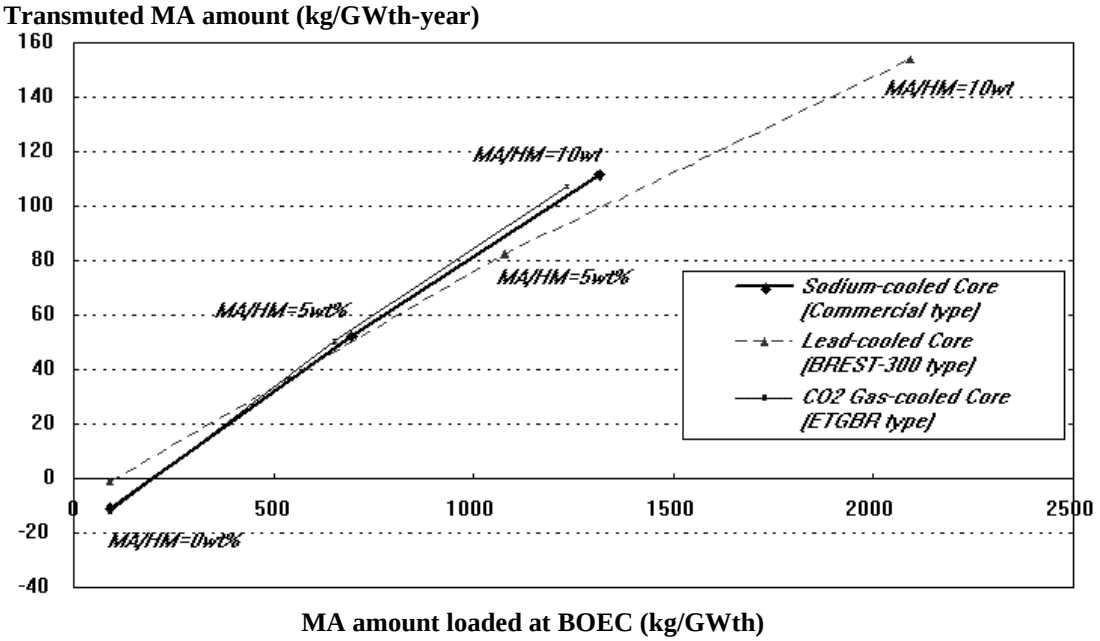


Figure E.4. **Dependence of MA transmutation effectiveness on MA loading for different coolants**



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Annex F

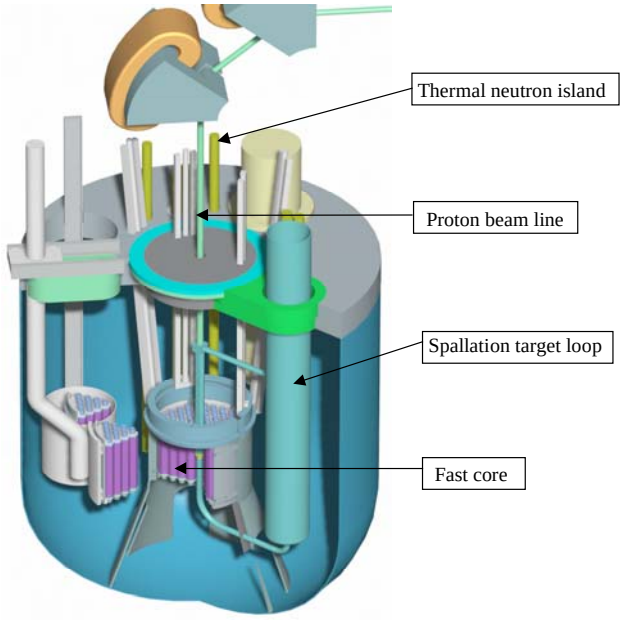
OVERVIEW OF NATIONAL AND INTERNATIONAL ADS PROGRAMMES

Belgium

SCK•CEN is at the present time finalising the pre-design of an ADS prototype called MYRRHA (see Figure F.1). MYRRHA is intended to be a multipurpose R&D irradiation facility. One of its main purposes is investigating the feasibility of actinide transmutation. Other ADS-related research topics concern materials and fuel behaviour, the utilisation of liquid metals and the associated issues, the reactor physics and safety of sub-critical systems, and the production of radioisotopes. The planned design period till 2003 will continue at the initial pace of the previous years. Construction of the Myrrha pre-prototype depends on a specific authorisation by the government.

The accelerator part of the device, presently designed by IBA, is to deliver a 350 MeV, 5 mA proton current. A neutron yield slightly higher than 3 per proton is expected at this energy. The spallation source would be Pb-Bi, windowless design, with an outer diameter of about 72 mm. The sub-critical core (k_{eff} 0.95) consists of an annulus, around the spallation source, of Pb-Bi cooled FR-type MOX assemblies (active length: 50 cm) with high Pu content (up to 30% in some zones). The fast zone is to be further surrounded by thermal "islands" in separate in-pile sections, with low neutron flux coupling to the fast core. The total power of MYRRHA should not exceed 30 MWth. Fast fluxes (E>0.75 MeV) up to 1 015 n/cm^2·s are to be attained in irradiation positions near the spallation source intended for minor actinide transmutation.

Figure F.1 **Conceptual view of MYRRHA**



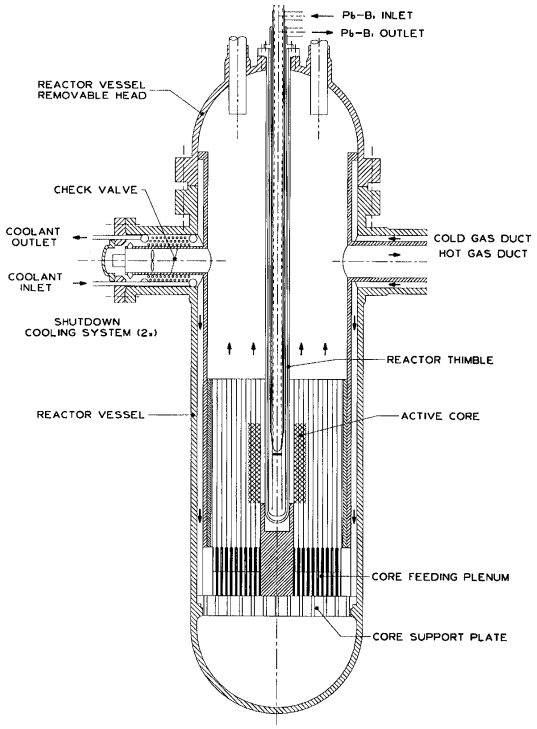
France

A research group (GEDEON) made up of CNRS, CEA, EdF and Framatome was launched in 1996 to co-ordinate the activities within France related to P&T. This co-operation was a means to intensify and co-ordinate the research requested by the 1991 law. The main options for the XADS have been defined in 1998 by a French working group leaded by the Ministry of Research and grouping CEA, CNRS, EDF and Framatome. The main technical options are as follows:

- A proton beam with energy between 400 MeV and 1 GeV, impacting on a heavy metal spallation target.
- A sub-critical core in a fast neutron spectrum.
- A solid fuel for the transmutation of the radioactive wastes.
- A maximal power for the sub-critical core lower than 200 MW thermal.
- A physical separation ("window") between the accelerator and the spallation target.
- A physical separation between the spallation target and the reactor housing the sub-critical core.

Based on these main options, a XADS concept has been proposed by France at the European TWG. The concept is still preliminary and studies should be performed in the frame of the Fifth European Framework Programme to consolidate the proposed design. Gas has been chosen as cooling medium of the sub-critical core (See Figure F.2). It had been judged that this option should be investigated in order to propose an alternative to the liquid metal concepts using sodium, lead or lead-bismuth. Helium has been preferred owing to its thermal characteristics, and because the risk of chemical interactions, radiolysis and radioactive activation can be intrinsically excluded.

Figure F.2. **Gas-cooled XADS concept**



Activities have been performed in several areas including neutron cross section measurements, integral experiments at MASURCA, development of the IPHI accelerator and system studies. Today, a report to promote the development of a demonstration-ADS is being prepared.

A multi-purpose irradiation facility, called CONCERT (COmbined Neutron Centre for European Research and Technology), has been proposed by CNRS. This facility would use the secondary beams, produced by high energy protons in a spallation target, for research in muon science, nuclear/particle physics, neutron science, radioactive beams, materials research and also nuclear transmutation research. A five years feasibility and detailed engineering study is proposed while the total construction period for the installation and the experiments would take another 10 years.

Germany

At the Technical University Munich the design of a separated-orbit cyclotron, with super-conducting channel magnets and super-conducting RF cavities for a 1 GeV proton beam of up to 10 MW beam power, is under development (TRITON). The distinguishing feature of this type of cyclotron is the strong transverse and longitudinal focusing. Recently it was demonstrated that the principle works as anticipated with operation well above the design values.

In Germany, some small activities related to the application of ADSs for the back-end of the fuel cycle have been in progress for several years. The first main objective was to establish reliable calculation procedures in order to be able to compare ADS capabilities with those of critical reactors. Exploratory investigations have been performed for thermal systems with dispersed fuel in lead coolant at FZJ Julich and for Phénix-like fast systems at FZK Karlsruhe.

Italy

ENEA and INFN set up a basic R&D programme TRASCO aiming at the study of physics and technologies needed to design an ADS for nuclear waste transmutation. The programme consists of research sub-programmes on accelerator, neutronics, thermal-hydraulics analysis, beam window technology, and material technology and compatibility with Pb and Pb-Bi. An industrial programme was also set up to issue a reference configuration description of a low power ADS prototype.

ANSALDO has embarked, together with Framatome, on a design of a prototype gas-cooled or LBE-cooled ADS (XADS), which was proposed to the European Technical Working Group. Since early 1998, the Italian ENEA, INFN, CRS4 and Ansaldo have set up a team, led by Ansaldo, to design an 80 MWth XADS, a key-step towards assessing the feasibility and operability of an ADS prototype. The results obtained so far [1], though preliminary and not exhaustive, allow outlining a consistent XADS configuration (see Table F.1 and Figure F.3).

The concept is still preliminary and further studies will be performed in the frame of the Fifth European Framework Programme to consolidate the proposed design.

In support to the these ADS design activities, ENEA has decided to build CIRCE [2] – a Pool Test Facility based on LBE – which will allow to test the key operating principles of the LBE XADS. The basic configuration of CIRCE, including the first test section, has been completed and commissioned at the site of Brasimone (Italy) in 2001. The CIRCE facility will be shortly described in Chapter 7.

Table F.1. **XADS configuration**

Plant area	Reference solution
Plant power	80 MWth sub-critical system controlled by a 600 MeV, 6 mA proton beam
Target/Window	Two options: a) Proton window b) Windowless target
Core	0.97 (at beginning of cycle)<k_{eff} 0.94 (at end of cycle), at full power
Fuel	U and Pu MOX
Primary system	Pool configuration with four integrated IHXs
Primary coolant circulation	Circulation enhanced by gas injection in a natural-circulation reactor configuration
Secondary system	Two low vapour pressure organic diathermic fluid loops rejecting heat by means of air coolers
Thermal cycle	300°C at core inlet, 400°C at core outlet
Reactor roof	Metallic plate
Main vessel and safety vessel	Hung from a cold annular beam
Structural materials	Vessels and internals: 316L Target and fuel SA's: 9Cr 1Mo
In-vessel fuel handling	One rotating plug, one fixed arm, one rotor lifting machine
Secondary fuel handling	Flask, encapsulator, canister, lifting and translating equipment, water pool
Nuclear island	Common basement on anti-seismic support
Plant safety	Fully passive system

Japan

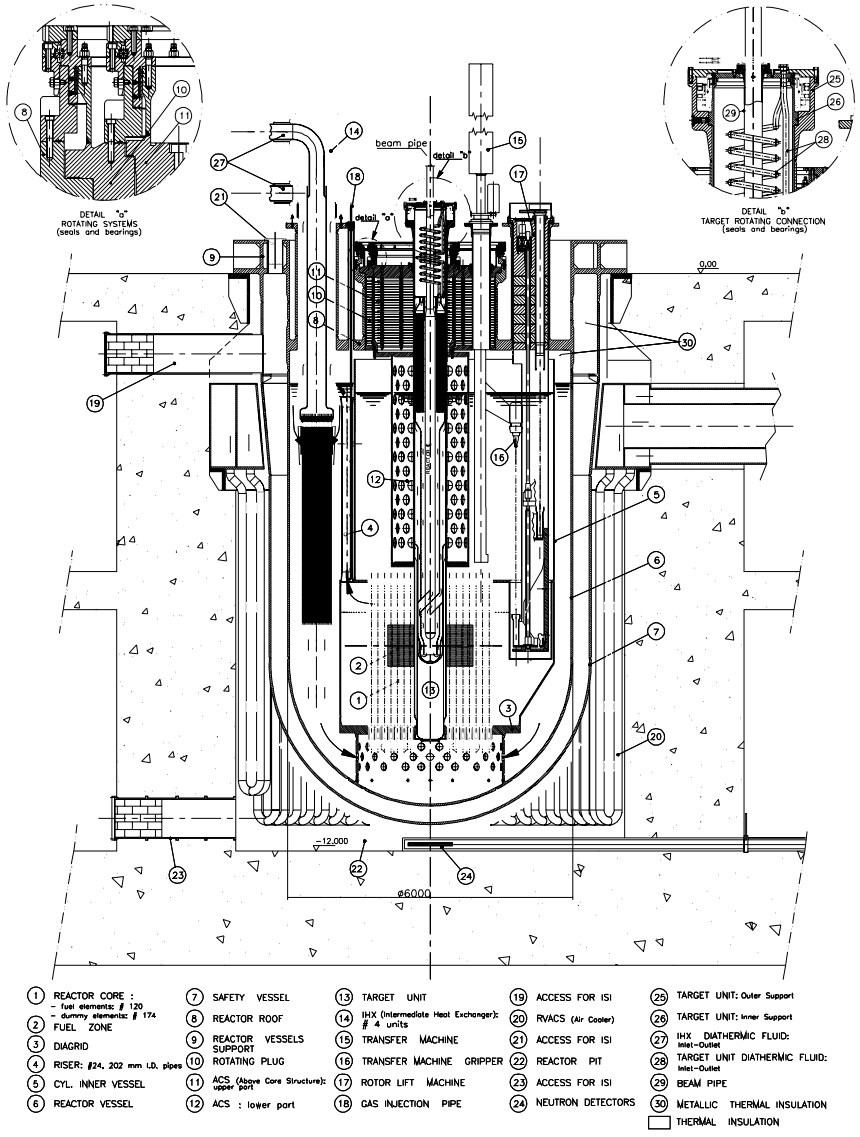
A preliminary design study of an 800 MWth lead-bismuth cooled accelerator-driven system (ADS) with nitride fuel has been directed towards a dedicated transmutation system to be deployed as the second stratum of a double-strata fuel cycle scheme. The plant has a pool-type configuration and a power conversion system operating on a saturated cycle (see Figure F.4).

An experimental program to develop and demonstrate accelerator-driven transmutation technology has been carried out under the project plan of the High Intensity Proton Accelerator and the OMEGA Program at JAERI. A pre-conceptual design study is being prepared for a transmutation experimental system. There are several technical challenges unique to the accelerator-driven transmutation system. The major areas of technology to be tested and demonstrated are sub-critical reactor physics, system operation and control, transmutation, thermal hydraulics, and material irradiation.

The typical sub-critical core configuration is based on that of the FCA (Fast Critical Assembly at JAERI) facility where various experiments can be conducted with changing core structure layout. The proton beam power will be 10 W with a proton energy of 600 MeV in pulses at a frequency of 25 Hz. The core thermal power is limited to 500 W, owing to the heat removal by forced air circulation.

Main integral measurements will be the reaction rate ratio and distribution, neutron spectrum, effects of high-energy neutrons, and sub-critical factor. System operation will be demonstrated and control experiments performed on beam trip effects, restarting operation and maintainability.

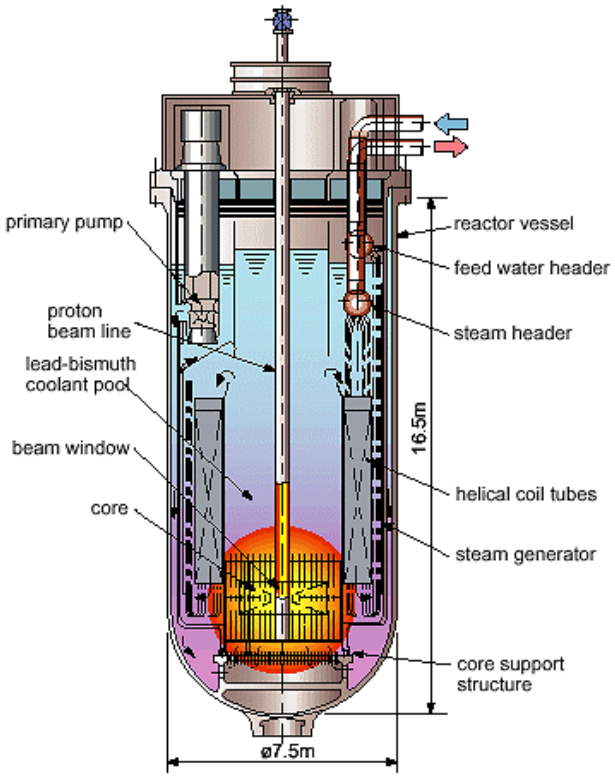
Figure F.3. **Scheme of a LBE-cooled XADS**



Furthermore, the engineering experiment facility is intended for research and development on beam window materials and thermal-hydraulic properties by using lead-bismuth target/loop equipment. As one of the most critical issues, corrosion/erosion in the lead-bismuth target/coolant system will be tested at operational temperature under proton and neutron irradiation. Material irradiation experiments will be performed with the proton beam power of 200 kW with 600 MeV and 25 Hz pulses in the first phase.

Most of the other important target and core technologies will be demonstrated through the experiments. In the planned scenario for developing the accelerator-driven transmutation system, the experimental program will proceed in a stepwise manner, according to the available power and the operating mode of the accelerator beam.

Figure F.4. **JAERI's design of a lead-bismuth cooled ADS with nitride fuel**



South Korea

A study on transmutation was initiated in 1992 at the Korea Atomic Energy Research Institute (KAERI). However, until 1995 the research was not very active. During this period, a sort of feasibility study was performed and some basic guidelines were set up to decide the research direction for transmutation. On the basis of these feasibility studies, an accelerator-driven sub-critical reactor was found to be the most promising candidate for incinerating nuclear waste from nuclear power plants. KAERI is setting up a long-term research programme called HYPER (HYbrid Power Extraction Reactor) and the schedule was drawn up in July 1997. The whole development schedule is subdivided into two phases. The basic key technologies are to be developed in Phase I (1997-2001) and a small bench scale test facility (~5 MWth) is to be designed and built in Phase II (2002-2006). Phase II will start only on condition that the Phase I research produces successful results. Therefore, a strict review will be performed within KAERI and the government just after Phase I. The expected major activities are: 1) developing a theoretical model to analyse coolant system behaviour, fuel system behaviour, and physics behaviour for the system design based on the experiments conducted in Phase I; 2) detailed design for the bench scale facility; 3) constructing a small scale facility; 4) doing performing a system safety analysis to obtain construction and operating permission from the regulatory body.

KAERI is also trying to launch a programme to develop a 1 GeV-20 mA multi-purpose linear proton accelerator called KOMAC. The design goal of the KOMAC is to generate protons of 1 GeV, 20 mA. It will be used for basic science research, radioisotope production, and transmutation technology development. The external review for the KOMAC system was done by international accelerator specialists in 1997. The user's program for the accelerator application was determined to

be developed in parallel with the KOMAC program in that review workshop. The development schedule consists of two phases in combination with the HYPER program. The Injector, RFQ (3 MeV, 20 mA), DTL (20 MeV, 20 mA) and some low energy beam utilisation technology will be developed in Phase I (1997-2001) and the whole accelerator facility will be completed in Phase II (2002-2006).

Spain

CIEMAT launched in 1997 a P&T research program. The aim of the program is the study of ADS, with close attention paid to their applications in nuclear waste transmutation. The program has three main research lines. The first one is dedicated to the study of transmutation of long-lived radionuclides, including development of concepts, designs, operation models and computer simulation tools together with the participation in experiments on this field of research. The second line includes the partitioning of radionuclides, by hydrometallurgical and pyrometallurgical processes. The third line is dedicated to the study of materials that could be used in this type of systems, including in particular the use of Pb-Bi as coolant. Besides, several universities are participating in these projects. These research and development projects are supported by the Spanish Agency for Radioactive Waste Management (ENRESA).

Sweden

Research on P&T is mainly supported by the Swedish Nuclear Fuel and Waste Management Co. (SKB). The main activities occur at the Royal Institute of Technology in Stockholm, where physics, safety and other aspects of ADSs, are studied. The different research groups have strong international co-operative links and participate in projects supported by EU.

Switzerland

Nuclear energy research in Switzerland is concentrated at the Paul Scherrer Institute (PSI). Recent activities in the field of accelerator-driven systems and transmutation comprise investigations of the role and potential of accelerator-driven systems in advanced fuel cycles and possible benefits for the management of radioactive wastes, the validation of models in nucleon-meson transport codes by means of proton irradiation experiments, advanced fuel development, high-current cyclotron development, including conceptual design studies, and material technology development for liquid-metal spallation targets in the framework of the MEGAPIE initiative. Analytical studies in the reactor physics and safety area are planned.

The MEGAPIE experiment is currently set up at PSI as an international project with participation of CEA, FZK, CNRS, ENEA, SCK·CEN, JAERI, KAERI and US-DOE. The purpose of the experiment is to demonstrate the safe operation of a liquid metal target at a beam power in the region of 1 MW. The minimum design service life is one year (6 000 mAh). The target is scheduled for installation in the SINQ facility in 2004 or 2005. The major objectives of the MEGAPIE initiative are:

- Full feasibility demonstration of a spallation target system.
- Evaluation of radiation and damage effects of structures and beam window in a realistic spallation spectrum.
- Effectiveness of the window cooling under realistic conditions.

- Liquid-metal/metal interactions under radiation and stress.
- Post irradiation examinations.
- Demonstration of decommissioning.

A SINQ target irradiation experiment, in which miniature specimens of candidate structural and target materials are irradiated in special target rods, and the LISOR experiment, which allows liquid-metal/metal reactions under radiation and stress to be simulated in a liquid lead-bismuth loop set up at a 72 MeV proton accelerator, provide R&D back-up to the MEGAPIE experiment. Additional experimental R&D support to MEGAPIE is provided by the KALLA laboratory at FZK and by ENEA using the CIRCE loop at Brasimone.

The PSI activities in the field of accelerator-driven systems and transmutation are embedded in projects of the OECD/NEA and the fifth framework programme of the European Commission (EC). The respective EC projects are SPIRE, TECLA, HINDAS, CONFIRM, and PDS-XADS (for more information on these projects, see Chapter 7).

USA

In 1999 the US Congress directed the US-DOE to study the Accelerator Transmutation of Waste (ATW) project and to prepare a "roadmap" for developing this technology. In response to the congressional mandate, DOE developed, through the work of a steering committee and the national laboratories, an ATW roadmap that identified the technical issues to be resolved, proposed a schedule and programme, assessed the impact of ATW technology on the civilian spent fuel programme and estimated the costs of such a programme as well as identifying areas of development in other sectors and with other countries.

The roadmap exercise finally advised the US congress that an initial six-year programme of trade studies and science-based R&D on key technology issues, costing 281 million US$, would be prudent to increase the knowledge base to support future decisions.

Transmutation R&D in the US has been focused initially on accelerator-driven systems and has involved a series of trade-off studies. In all cases, it has been assumed that uranium remaining in civilian spent fuel elements would be recovered, probably by a modified Purex process called UREX. Initial studies of the UREX process have shown that the uranium product will meet US. Class C requirements and could be disposed of as low level waste or be stored for possible future use in a nuclear fuel cycle. The remaining process streams would be chemically separated into transmutation fuel material, long-lived fission product transmutation targets, and a waste stream that could be converted into durable waste forms capable of disposal in a high-level nuclear waste repository.

Various combinations of proton accelerator designs, spallation neutron sources, and transmutation target have been evaluated for technological readiness, and assumed irradiated targets have been studied for the effectiveness of chemical processing to recycle untransmuted long-lived isotopes. These evaluation have resulted in a base-line design which includes a linear proton accelerator (or Linac), a lead-bismuth spallation target, and sodium-cooled non-fertile elements of metallic or ceramic dispersion construction as transmutation targets or /blanket fuel. Other alternative designs have included cyclotrons as proton source, nitrides as transmutation targets, and tungsten spallation targets cooled by sodium, pressurised helium, or water.

Another interesting transmutation system design currently being evaluated consists of a "dual strata" approach which would involve a thermal critical reactor within which plutonium and minor actinides would fission, and ^{99}Tc and ^{129}I would be subjected to a thermal neutron flux. Technetium would probably be in metallic form and iodine as an iodide of sodium, silver or other stable cations. The thermal-spectrum reactor would be effective in burning plutonium-239 along with other actinides with high thermal fission cross-sections. Higher actinide isotopes would be produced by non-fissioning neutron capture, and after post-irradiation chemical processing, they would be the primary targets of an accelerator-driven transmutation system. Chemical processing of such targets after irradiation would result in actinide recycle to the ATW unit and recycle of ^{99}Tc and ^{129}I to the thermal reactor. High-level waste streams for repository disposal would be produced by the initial processing of civilian spent fuel, the recycle processing of spent fuel from the thermal reactor, and the ATW recycle process.

Since transmutation produces a net energy gain, it has been of interest to design systems capable of producing electric power to off-set transmutation expenses. One concern has been the current high "trip" rate of present generation accelerators, which may experience several unplanned cut-offs each day. Quite apart from safety considerations of thermal shock in the transmutation system, such interrupted power would have much lower value than conventional base-load systems. Early analysis indicates that more than ninety percent of the energy release in the "dual strata" would occur in the thermal reactor, so it may be possible to design the ATW system as a low-temperature actinide burner with much less stringent requirements for accelerator power and stability. Materials and corrosion problems in the ATW system would also be minimised. Studies of the concept are continuing.

Advanced Accelerator Applications (AAA)

The ATW program during the fiscal year 2001 involves approximately a doubling of the Fiscal Year-2000 funding. This will allow an expansion of experimental programs, and DOE's Office of Nuclear Energy, Science and Technology (NE) is actively seeking opportunities for collaborative research with foreign ADS programs. Meanwhile, the program is being reorganised to combine the objectives of the DOE Defence Programme's Accelerator Production of Tritium program with those of NE's ATW efforts. The combined programme is known as Advanced Accelerator Application, and it will be administered by NE. Congress has requested a report by March 1, 2001 on how the new activity will be carried out. It will be a public document, available on the World Wide Web as well as in hard copy.

One objective of the new program will be to help strengthen the nuclear science infrastructure in America. To accomplish this, graduate thesis projects related to the program objectives will be sponsored at many universities. Another objective will be to strengthen nuclear test facilities, and an Accelerator Driven Test Facility is under active consideration. The need to make better use of limited test facilities throughout the world is also one of the reasons why DOE will be seeking to increase international ADS/ATW collaboration. The coming years may see a considerable expansion of the international quest for effective transmutation systems.

Russia

Several research institutes in Russia are involved in a P&T programme directed by MINATOM. Most of the activities relevant to ADS are carried out within the framework of ISTC projects. The main research institutes involved in R&D in the field of ADS are: Institute of Theoretical and Experimental Physics (ITEP), Institute for Physics and Power Engineering (IPPE), All-Russian Scientific Research Institute of Experimental Physics (VNIIEF), Joint Institute of Nuclear Research

(JINR), Institute of Nuclear Energy (IAE), Institute of Nuclear Research of the Russian Academy of Sciences (INR RAS), Experimental Design Bureau GIDROPRESS (OKB GP), and the Petersburg Nuclear Physics Institute (PNPI). The main activities performed are: theoretical research, accumulation of experimental data for the justification of the physical processes in ADS, and design studies on ADS and its sub-systems.

European Commission

The Fifth Framework Programme (1998-2002) of the European Atomic Energy Community (EURATOM) has two specific programmes on nuclear energy, one for indirect research and training actions and the other for direct actions with the Joint Research Centre of the European Commission. The first one, "Research and training programme in the field of nuclear energy", includes a key action on nuclear fission and comprises four areas: (i) operational safety of existing installations; (ii) safety of the fuel cycle; (iii) safety and efficiency of future systems and (iv) radiation protection. In the safety of the fuel cycle, waste and spent fuel management and disposal, and partitioning and transmutation (P&T) are two large activities, whereas the decommissioning of nuclear installations is a smaller one.

To implement the key action on nuclear fission and the generic research on radiological sciences, a first call for proposals was made in 1999. In the area of partitioning and transmutation, 20 proposals were received, requesting about 3.8 times more than the available budget. By taking due account of the advice of the evaluators, the Commission services selected 10 proposals for funding at a level lower than requested due to budget limitations.

The selected projects are subdivided into three clusters: (i) partitioning, (ii) transmutation – technological support and (iii) transmutation – basic studies. The cluster on partitioning includes three projects. The first one on pyrometallurgical processing assesses salt/metal extraction and electrorefining for the separation of actinides and lanthanides, while the two others will develop aqueous processes for the chemical separation of minor actinides from high level waste. In the cluster on technological support, four projects will address (i) experimental work on neutron and proton irradiation damage to a spallation target, (ii) corrosion of structural materials by lead alloys used as a spallation target and as a coolant for an accelerator-driven system (ADS) and thermal hydraulic experiments with liquid lead alloys, (iii) fuel issues for ADS (fabrication and irradiation of nitride fuel) and (iv) irradiation of thorium fuel. Finally, three projects are grouped in the cluster on basic studies: one on the experimental investigation and code interpretation of sub-critical neutronics and two on nuclear data, one at medium and high energy required for the ADS engineering design including the spallation target, and one encompassing the lower energy in resonance regions required for transmutation.

Extended Technical Working Group (TWG)

In 1998, the Research Ministers of France, Italy and Spain, recognising the potentialities of Accelerator-Driven System (ADS) for the transmutation of long lived waste, had decided to set up a Group of Advisors (Ministers' Advisors Group – MAG) in order to define a common R&D European platform on ADS. On its meeting on May 1998, the MAG recommended a European demonstration programme over a 10-year time scale. A Technical Working Group (TWG) under the chairmanship of Prof. C. Rubbia was established with the task of identifying the critical technical issues in which R&D is needed, in view of a demonstration programme. In October 1998, the TWG issued an Interim Report [3] which, in particular, highlighted:

- The need of a demonstrator.
- The basic components and the different options for the proposed DEMO facility.

- The R&D directly relevant to the realisation of the demonstrator.

This report was endorsed by the MAG on its meeting of March 1, 1999 and, in the same context, the following main issues were brought forward:

- Extension of the European participation beyond the three countries initiative.
- Role of ADS transmutation R&D within the Fifth European Framework Programme.
- Recognition of the ASAP (As soon as possible)-DEMO as a European goal.

As a consequence, a MAG "ad hoc" meeting open to all the interested EU member states was held in Rome on April 21, 1999. Representatives of eleven countries (Austria, Belgium, Denmark, Finland, France, Germany, Italy, Portugal, UK, Spain and Sweden) participated in that meeting which gave rise to the following main conclusions:

- It was agreed that transmutation represents an attractive approach to radioactive waste disposal, being complementary to geological disposal.

- All participants appreciated the proposal to extend the participation in the initiative to other European countries besides France, Italy and Spain, particularly considering that similar approaches were being undertaken in the USA and Japan.

- The interim report of the TWG issued in 1998 was accepted as a good basis for future work to be carried out by an Extended (actually European) Technical Working Group (ETWG), under the chairmanship of Prof. C. Rubbia.

In September 1999, the ETWG – composed by representatives of Austria, Belgium, Finland, France, Germany, Italy and Spain – issued a new technical report [4] aimed at providing an overview of the different ongoing activities on ADS in various European countries, along with an examination of the proposals to be submitted to the Fifth FWP. The report, presented to and endorsed by MAG on its meeting of September 17, 1999, also identified a number of open points and gave recommendations for the future development of the activities. In particular, the ETWG strongly recommended an increased support – even by European Commission – and co-ordination of ADS-related activities at multinational level.

Early 2000, the ETWG (further enlarged to representatives of JRCs, Portugal and Sweden), issued a so-called four-page document [5] on strategy of implementation of the ADS programme in Europe. In particular, the document calls for the urgent definition of a "roadmap" towards demonstration of feasibility of an European waste transmutation facility and recognises its potentially-relevant implications on the 6th European Framework Programme. The four-page document was submitted to the MAG at its last meeting on February 25, 2000 and received positive comments: consequently, the TWG was committed and encouraged by MAG to proceed in the forthcoming months in defining the above-mentioned roadmap.

The report, entitled "A European Roadmap for Developing Accelerator Driven Systems for Nuclear Waste Incineration" [6], was issued by the ETWG on April 2001.

After reviewing historical background and identifying motivations for developing ADS technology in the field of P&T, the Roadmap defines and proposes a detailed technical programme, comprehensive of planning and cost estimates, which will lead to the construction of an Experimental ADS (XADS) within 12 years, covering the 6th and 7th European Framework Programmes. This is

considered, by the ETWG, as an essential prerequisite to assess the safe and efficient behaviour of such systems for a large-scale deployment for transmutation purposes in the first half of this century.

The document also reviews and assesses the status of current scientific and technology programmes and facilities relevant to ADS research in the EU and worldwide, and – by means of three specific reports - presents a comprehensive overview of the status and future developments in the field of high-power proton accelerators [7] and innovative fuels and reprocessing technology [8,9].

At last, the Roadmap identifies possible synergies that the ADS programme could have within the scientific community, indicates potential spin-offs, shows how competence can be maintained in the currently stagnating field of nuclear energy research.

As a result of a mandate given to the ETWG, the Roadmap was directed, in the first instance, to MAG on May 2001. The document – being of interest, however, to policy makers throughout Europe – was also addressed on July 2001 to members of the European Parliament, to the relevant Directorates General of the European Union, as well as to national and international organisations involved with ADS research and development within the EU and worldwide.

IAEA

In compliance with its statutory mandate, one of IAEA's roles is to provide all Member States with an international source of balanced and objective information on advances in nuclear technology, and to provide an international forum for information exchange and co-operative research.

Accelerator-driven transmutation of long-lived waste has increasingly become of interest in many Member States, and could be an important component of strategies to deal with international requirements in managing nuclear materials.

To respond to the Member States' needs, the IAEA has established the project on "Technology Advances in Fast Reactors and Accelerator Driven Systems for Actinide and Long-lived Fission Product Transmutation" [10].

Within the framework of this project, a status report "Accelerator-driven Systems: Energy Generation and Transmutation of Nuclear Waste" [11] was published, providing an overview of ongoing development activities, different concepts being developed and their status, as well as typical development trends in this area, and evaluating the potential of these systems for power production, plutonium incineration and transmutation of minor actinides and long-lived fission products. It is intended to update this status report at regular intervals, and establish it as a "living document" on the project's Web Site [10].

Among the most important collaborative R&D activities of the project, mention must be made of the "Coordinated Research Project (CRP) on the Use of Thorium-based Fuel Cycles in Accelerator Driven Systems (ADS) to Incinerate Plutonium and to Reduce Long-term Waste Toxicities", concluded at the end of 2000 [12]. The last stage of this CRP was centered on experimental benchmarks based on the YALINA experiments (a sub-critical, thermal facility set up in Minsk, Belarus in the frame of ISTC project #B070). It is planned to start in 2002 a follow-up CRP on "Benchmark Analyses on Data and Calculational Methods for Accelerator-driven System (ADS) Source Related Neutronic Phenomenology with Experimental Validation", addressing all major physics phenomena of the spallation source and its coupling to the sub-critical system. The participants will apply integrated calculation schemes to perform computational and experimental

benchmark analyses. Also to start in 2002, the CRP on "Studies of Advanced Reactor Technology Options for Effective Incineration of Radioactive Waste" will focus, in its first stage, on analyses of safety-relevant parameters of ADS. The main thrust will be on long time-scale effects of transients initiated by strong perturbations of the neutron source or of the sub-critical core. Benchmark models based on various designs of the sub-critical core, as well as extreme cases (sub-critical cores "dedicated" to transmutation, i.e. fuelled with transuranics in a fertile-free matrix) will be considered. This CRP will also seek to perform experimental benchmark studies.

Last but not least, an important ongoing activity is to implement a "Database of Experimental Facilities and Computer Codes for ADS Related R&D" (so called "ADS R&D Database"). Presently, a WWW-based version of the database is being tested in-house and will be operational shortly [10].

OECD/NEA

Back in 1989, the OECD/NEA started a comprehensive programme of work in the field of partitioning and transmutation (P&T) [13]. This programme was initiated by a request from the Japanese government which was launching a programme on P&T (OMEGA project) and invited the OECD/NEA to co-ordinate an international information exchange programme on P&T. This has since materialised in several activities, among them the Information Exchange Meetings and state-of-the-art systems studies besides a diverse set of activities oriented towards more basic science. NEA has recently reorganised the P&T activities as a horizontal project between the Nuclear Development and Nuclear Science Committees, and while a restructuring of the science programme under the umbrella of a new Working Party on Scientific Issues in P&T, covering specifically ADS aspects, has recently been started. This Working Party will envelop the scientific aspects of P&T and comprises four sub-groups:

- Group on Accelerator Utilisation and Reliability:

  This group emerges from previous workshops on Accelerator Utilisation and Reliability, will synthesise the improvements made and draw conclusions from each workshop held and continue to organise such workshops. The group will also deal with target and window performances, for instance, issues on spallation products and thermal stress and radiation damage, respectively.

- Group on Chemical Partitioning:

  The existing expert group on Pyrochemistry moves under this WPPT and will first focus on drafting a state-of-the-art report on Pyrochemistry. Despite its name, the group will also look into aqueous processing issues.

- Group on Fuels and Materials, as the new proposed transmutation systems will demand specific materials to be validated or developed for use in more challenging irradiation conditions.

- Group on Physics and Safety of Transmutations Systems:

This group will organise theoretical and experiment-based benchmarks to validate nuclear data as well as calculation tools needed for simulating advanced transmutation systems, and investigate safety aspects of transmutation systems such as the beam trip problem of ADS.

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