TABLE OF CONTENTS

List of figures

TABLE DES MATIÈRES

Liste des figures

La partie II existe exclusivement en version anglaise.

List of figures

List of tables

EXECUTIVE SUMMARY AND SCOPE OF THE REPORT

Implementation of partitioning and transmutation (P&T) technology is intended to reduce the inventories of actinides and long-lived fission products in nuclear waste. Implementation of such technology may decrease the hazards associated with pre-disposal waste management and with physical disturbance of a waste repository, e.g. by human intrusion, but it is not seen universally as a short-term alternative to contemporary fuel back-end policies nor as a means of avoiding the need for a deep geologic repository facility for long-lived waste from contemporary operations.

Nevertheless, there is a widely held view that technologies for reducing these inventories may have the potential for alleviating the waste disposal problems of the fuel cycles designed specifically to exploit them. The associated studies would serve, at least, to advance the science of nuclear energy and attract capable young researchers into the field in preparation for a possible swing of global public opinion towards accepting nuclear power. In addition, studies of laser and accelerator technology, for example, may well provide spin-offs for other fields of science and technology.

Much of the current work in this area is at an early stage of development. It is difficult to predict the outcome, or associated timescales, of current research and development (R&D) or of the costs associated with development and operation of technologies under study. The structure of a total system study and the related requirement for information are outlined in this report but the primary focus, for the present, is on review of the potential technologies for partitioning and transmutation together with a critique of development status and prospects for eventual industrial application.

It is the purpose of this report to provide an authoritative analysis of the technical, radiological and, as far as possible, the economic consequences resulting from the proposed partitioning and transmutation operations on the present and future fuel cycle options. The present report is subdivided in a general part for non specialist readers and a technical systems analysis discussing the issues in partitioning, transmutation and long-term waste management:

Part I: General overview

- Introduction on the international programme co-ordinated by the OECD/NEA.
- A general overview of status and expectation of P&T technology and a comparative analysis of the issues associated with the different fuel cycles.
- A critical evaluation which tries to identify the strong points and weaknesses of the partitioning and transmutation strategy.
- General conclusions and recommendations on future priorities in development.

Part II: Technical analysis and systems study

- An analysis of the potentials of different partitioning methods, either as extensions of the PUREX process or as an autonomous pyrochemical fuel processing method.

- An extended technical discussion on the possible transmutation options for minor actinides (MAs) and selected fission products (FPs) by light water reactors (LWRs), fast reactors (FRs) and hybrid transmutation devices (accelerator-driven transmutation systems (ADS)) including:

  – a feasibility assessment of minor actinide fuel and target fabrication technology and safety;
  – a detailed overview of the issues involved in irradiating minor actinide-containing fuels and targets in current pressurised PWRs (homogeneous and heterogeneous recycling);
  – a comprehensive overview of the transmutation potential of minor actinides in fast reactors as mixed oxides (MOX), metal, and inert matrix fuel forms or targets;
  – a preliminary analysis of the accelerator-driven transmutation systems potential, the issues involved and the current projects;
  – a comparative analysis of the transmutation potential for selected long-lived fission products in different reactor types and accelerator-driven transmutation systems.

- A description of the major research and development programmes carried out in Japan and France.

- A comprehensive discussion of the impact of partitioning and transmutation on risk assessment and waste management, especially the long-term impact of currently proposed methods.

- An analysis of the different transuranic element inventories according to the strategic option taken in the evolution of nuclear power plant (NPP) types (light water reactors, fast reactors, accelerator-driven transmutation systems, etc.) in a future nuclear electricity production grid.

- A short overview of the current waste disposal concepts and the possible impact of partitioning and transmutation on the repository structure and characteristics.

- A discussion of the long-term radiological and radiotoxic impact of the partitioning and transmutation operations.

General overview

A general overview is given of the current fuel cycle status and nuclear materials management as it is practiced today. The expectations of new options that partitioning and transmutation might offer, as well as their limitations, are discussed in an introductory chapter. Additional recycling operations of fissile and fertile materials (uranium, plutonium), minor actinides (neptunium, americium, curium) and some selected fission products (^{99}Tc, ^{135}Cs, ^{129}I, etc.) are presented as an introduction to this complex matter for a non-specialist reader.

A comprehensive analysis is given of the three fuel cycle options: the "Once-Through Cycle" (OTC), the "Reprocessing Fuel Cycle" with uranium and plutonium recycling (RFC) and the "Advanced Fuel Cycle" with partitioning and transmutation (AFC) within the framework of a nuclear electricity production grid of, for example, 100 GWe. Particular attention is devoted to the open issues where management decisions have to be taken in order to minimise the radiotoxic legacy resulting from nuclear energy production.

Reference data on irradiated UO_2 and mixed oxide fuels for a typical range of irradiation conditions are summarised in composite overview tables. Extensive nuclear data tables of the radionuclide inventory of spent fuel are given in Annex E of this document. The data provided are mean values from computer programmes accepted throughout the nuclear community as reference figures for the calculation of radiotoxic inventory over extended periods of time.

Critical evaluation and general conclusions

A series of critical evaluations and general conclusions drafted by the expert group have been added to the general part of the report in order to highlight the key issues which may help decision makers to establish the priorities in future research and development work, to underline the potential of the partitioning and transmutation option in waste minimisation and to draw the limits of this strategy in a future nuclear era.

Technical analysis and systems study

Partitioning

Reprocessing of spent light water reactor fuel is considered as a reference scenario which is a necessary preliminary to undertake additional partitioning of minor actinides and selected fission products in the first step of the advanced fuel cycle.

The aqueous separation techniques, compatible with the well known PUREX process, are briefly described and illustrated by their flowsheets. The possible and desirable improvements for transuranic element extraction in the PUREX process to recover ^{237}Np, and the associated minor actinide extraction processes "TRUEX", "DIDPA", "TRPO", "DIAMEX", "TPTZ", "SESAME" and the latest "CYANEX 301™" processes to recover americium and curium, are briefly described with comments on their merits and limitations. Their impact on the conventional waste treatment processes are briefly discussed, especially the secondary waste generation. An extensive bibliography on partitioning is given in Annex D.

The separation of some fission products requires additional operations within the head-end of the PUREX process since there is segregation between insoluble and soluble fission products. The recovery of ^{129}I and ^{14}C from dissolver off-gas are examples of how the impact of reprocessing on the environment can be reduced. For two other long-lived radionuclides, i.e. ^{99}Tc and ^{93}Zr, their separation from tributylphosphate (TBP) streams could also reduce their environmental impact of the wastes.

Over the last two decades considerable progress has been made in the selective extraction of minor actinides and fission products but the technical maturity of each of these methods, as summarised in an overview table, is very variable.

There are several scenarios which can be considered in the management of separated minor actinides:

- neptunium resulting either directly from the reprocessing process or extracted from HLW could be separated easily from the other nuclides and recycled into MOX fuel or kept in separated form till advanced transmutation-incineration processes (FR, ADS) become industrially available. Alternatively the separated neptunium could be conditioned in a thermodynamically stable phase and kept in a retrievable long-term storage. This matrix could be designed to serve either as irradiation matrix or as final disposal waste form.
- the americium-curium fraction separated from HLW could be treated as a single fraction both in a transmutation strategy and in a disposal option. Both elements behave chemically so similar that very special radiochemical and/or electrochemical techniques have to be used to achieve the americium/curium separation. In a group separation option the americium-curium fraction could be stored as such or conditioned as a ceramic type of matrix and kept in engineered storage till transmutation techniques become available.

Americium could ultimately be separated as individual element and recycled into special LWR-MOX or FR-fuel to be partially transmuted or could be conditioned in a ceramic form for extended transmutation-incineration processes by multi recycling and ADS processes.

The management of separated curium has not yet received convincing answers not only because the curium isolation is difficult but also because this very radioactive nuclide (heat and neutron source) cannot be recycled in LWR- or FR-MOX fuel. Storage of the separated nuclide for a century would alleviate the processing problems and make recycling in an advanced fuel cycle possible.

In a perspective of increased burnups and shorter cooling times, the development of non-aqueous pyrochemical partitioning methods deserves special attention. Pyrochemical methods in molten chloride salt (LiCl-KCl or NaCl-KCl) baths have been tested in laboratory and hot pilot scales in order to separate uranium, plutonium and minor actinides from fission products. A combination of electrolytic separation, chemical precipitation and reductive extraction might be used in the future either as pre-separation steps before aqueous reprocessing or perhaps as an autonomous process for dry reprocessing of high burn-up spent fuel with shorter cooling time. Successful achievement of this technology may shorten the time interval for a complete recycle sequence.

Transmutation

The Transmutation chapter covers a number of activities: fuel and target fabrication, transmutation by neutron capture and “incineration” by fission.

Transmutation can be carried out in thermal reactors, in fast neutron reactors and in accelerator-driven subcritical reactors. The proper choice of each of these transmutation facilities derives from strategy studies which have to provide an optimum combination between the many diverse aspects of fuel fabrication and refabrication, availability of reactor types, transmutation/incineration yields, and last but not least, a significant reduction in overall radiotoxic inventory of the nuclear waste. Extensive studies have been undertaken within the European Union and Japan to provide a first strategic approach.

However these strategy studies are subject to political decisions and economic realities which cannot be forecast over the several decades or centuries necessary to achieve a steady state between input and output of fissile and fertile nuclear materials.

Fuel and target fabrication issues

The report treats in a factual way the problems encountered when introducing minor actinides into the nuclear fuel cycle. Fabrication of mixed oxide fuel with low (<2.5%) neptunium and americium loadings is a feasible option which does not require large investments in the light water reactor mixed oxide fuel (LWR-MOX) fabrication plants. However, the introduction of these minor actinides makes subsequent reprocessing and refabrication of fuel or targets more difficult because a large amount of ^{238}Pu is present in the recycled plutonium stream.

Heterogeneous recycling of minor actinides in light water reactors is highly preferable since the targets can be managed independently of the light water reactor mixed oxide fuel and recycled in dedicated small-scale processing facilities. However the main problem which has not yet received a satisfactory solution is the recycling of ^{244}Cm which is a strong heat and neutron emitter and cannot be handled in regular fuel fabrication facilities. Future work in the field of partitioning is required to provide a workable technology.

Fast reactor fuel has been produced for several decades and experimental fuels with large loadings of americium and neptunium have been fabricated in the frame work of the "SUPERFACT" programme. The irradiation of minor actinides in a fast neutron flux should preferably be achieved in inert matrices (aluminum oxide, magnesium oxide, etc.).

In order to further increase burnups and transmutation yields, new fuel forms are being studied, in particular metal alloy (of minor actinides, uranium, plutonium and zirconium) and nitride fuel. One of the advantages of using metal fuels is the possibility of forming a compact fuel cycle system based on the pyrochemical techniques, although substantial research and development efforts are still needed for its realisation.

The feasibility of nitride fuel is dependent on the availability of nitrogen-15 enrichment facilities and recovery techniques during recycling procedures. The application of pyrochemical processing to the nitride fuels may also facilitate its recycling.

Transmutation of minor actinides

The physics of transmutation/incineration of minor actinides shows that fast neutrons, either in fast reactors or in accelerator-driven systems, are preferable to a thermal one because of the more advantageous ratio between fission and capture cross-section. Moreover, all minor actinides are to a certain extent fissile in a fast neutron spectrum and contribute during irradiation to the overall energy production.

Comparative tables with calculated transmutation/incineration yields are given for homogeneously and heterogeneously loaded minor actinides in light water reactors fuelled with mixed oxide as well as for fast burner reactors and advanced liquid metal reactors. Most of the advanced reactor concepts are still in the conceptual phase and the calculated yields have yet to be verified in representative conditions.

Minor actinide transmutation in fast reactor cores (liquid metal fast breeder reactors, advanced liquid metal reactors) has no serious drawback in terms of core performance, provided that the ratio of minor actinides to fuel remains small.

The design of fast burner reactors and minor actinide burner reactors (MABRs) needs further research and development because the effects on reactivity coefficients and kinetic parameters cast some doubt on their feasibility with high minor actinide loadings.

Recently hybrid type accelerator-driven sub-critical transmutation systems have been receiving increased attention world-wide as an alternative to conventional nuclear reactors. Accelerators, whether linear accelerators or cyclotrons, produce a proton flux which impinges on a heavy metal target which undergoes spallation. The spallation neutrons impinge in their turn on fissile and fertile nuclear materials, e.g. minor actinides, which undergo fission and capture.

The transmutation of minor actinides cannot be dissociated from the transmutation of plutonium, and any strategy in this connection has to take into account the depletion of minor actinides in targets or fuels and the simultaneous generation of minor actinides from plutonium.

Transmutation of long-lived fission products

Transmutation of long-lived fission products (^{99}Tc, ^{129}I) to short-lived or inactive nuclides has been investigated and led to the conclusion that it is theoretically possible but that higher fuel enrichments are necessary in light water reactors. The transmutation half-life varies from 15-18 years in fast reactors to 40-77 years in light water reactors. Irradiation of selected fission products in accelerator-driven systems or optimised fast reactors would make it possible to decrease the transmutation half-life. Intensive development of target and fuel assembly materials would be required (as in the case of Iodine), as well as refined isotopic separation in the case of some elements (e.g. Se , Zr, Cs, Sn, …).

Current trends in partitioning and transmutation research

Taking into account the progress of the research and development programmes in Japan and in France, a special section and Annexes B and C deal with the current partitioning and transmutation activities in these countries. These programmes, in which many countries participate directly or indirectly, are undoubtedly the most important in the world.

Impact of partitioning and transmutation on risk assessment and waste management activities

In order to set out very clearly the impact of partitioning and transmutation on long-term radiotoxicity, a special section in the report is devoted to the significance and definition of radiotoxic inventory.

The radiotoxic inventory depends on the physical inventory of radionuclides in the various fuel cycles and on the effective dose coefficients of individual radionuclides. Graphs show the natural evolution of radiotoxic inventories corresponding to the once-through cycle, reprocessing fuel cycle and advanced fuel cycle.

The role of partitioning and transmutation in waste management depends primarily on the minor actinide inventories to be handled in the advanced fuel cycle facilities. The section discusses the reactor strategy to be implemented in order to achieve a significant reduction of the radiotoxic inventory. Three representative cases are considered with various combinations of light water reactors loaded with uranium dioxide and mixed oxide fuel, and fast reactors loaded with mixed oxide fuel. A substantial proportion of fast reactors is necessary. In each of the cases examined, the radiotoxic inventory in the waste streams is reduced by a factor of about 100. However, the decrease observed in the waste discharges is accompanied by a steady increase of the radiotoxic inventory of the reactor and fuel cycle facilities operated over a long period of time to accomplish this waste minimisation.

Where all minor actinides are recycled including the curium fraction, the radiotoxic inventory reduction factor, compared with once through cycle, ranges from 77 to 100 after 10 000 years, and in the very long term (10^5 and 10^6 years) from 80 to 150.

If the curium fraction is not recycled, the radiotoxic inventory is reduced only by a factor of 7 to 14 at 10 000 years owing to the decay of ^{244}Cm into ^{240}Pu and ^{243}Cm into ^{239}Pu.

When making a global mass balance of the minor actinides involved in a long-term multiple recycling programme, the radiotoxic inventory of the nuclear materials in the reactors and fuel cycle facilities becomes overwhelmingly more important than the annual waste discharges. Various cases leave equilibrium inventories of several hundred tonnes of transuranic elements.

Based on data from different economic assessments, but principally relying on a comprehensive study carried out within the European Union’s R&D programme on radioactive waste management and disposal, some very preliminary cost data for the advanced fuel cycle with partitioning and transmutation strategy are given in Annex F.

Waste management concepts

Estimates of the radiological benefit to be expected from partitioning and transmutation must take into account the mobilities of the elements in the geosphere, which may substantially modify the simple picture presented by the reduction in radiotoxic inventory alone. Other benefits, e.g. with respect to intrusion, may however be derived from the possibility of especially secure conditioning and emplacement of the long-lived radionuclides.

In order to fit the partitioning and transmutation option into the context of existing waste management strategies, the direct disposal concepts in granite formations of Spain and Sweden are briefly described as examples. A section discusses the issue of criticality safety in the disposal of spent fuel.

In the case of the reprocessing fuel cycle, the impact of the disposal of high- and medium-level waste as scheduled to be realised in Germany, Switzerland and Belgium, respectively in salt, granite and in clay, are given as illustrations. A number of options are being studied on disposal of waste forms resulting from the advanced fuel cycle incorporating partitioning and transmutation. Nevertheless, the report sheds some light on the technical and operational aspects of such a strategy; in particular, intermediate storage management, the fate of irradiated targets and the residual reactor cores from a prolonged nuclear energy production.

In addition to the highly radioactive materials resulting from an advanced fuel cycle, attention should be drawn on the large inventories of depleted uranium produced during the enrichment processes. The depleted uranium stocks can in a short time interval be considered as a strategic material to be reused in case the FRs would become an important fraction of the total electricity generating capacity.

If the FRs do not emerge as nuclear electricity producers, depleted uranium will be considered as a waste material whose radiotoxicity will eventually become equal to that of natural uranium ore.

PART I: GENERAL OVERVIEW

1. INTRODUCTION

1.1 Involvement of OECD/NEA

During the last decade interest in Partitioning and Transmutation (P&T) has grown in a number of OECD Member countries as a result of the initiative taken in 1988 by the Japanese government to launch a long-term research and development programme on the recycling and transmutation of actinides and long-lived fission products. This programme called OMEGA (Options for Making Extra Gains from Actinides and fission products) was initiated by the Atomic Energy Bureau of the Japanese Science and Technology Agency (STA) for "the enhancement of the efficiency of high-level waste disposal and the utilisation of resources in the spent fuel." The programme was conceived as a long-term basic research programme beneficial for future generations and as an incentive for young researchers to use their ingenuity in the scientific aspects of nuclear waste management.

The Japanese government invited OECD/NEA to conduct some form of international project related to actinide separation and use. After some discussions it was decided to set up a programme of technical information exchange which would be the centre-piece of a multilateral effort carried out by the participating OECD/NEA countries in the field of P&T. It was, from the outset of this action, very clear that this programme would not interfere with the established "back-end" policies with and without reprocessing, vitrification of High-Level Waste (HLW) and disposal of waste or spent fuel in geological formations.

The first International Information Exchange Meeting was held at Mito City (Japan) in November 1990. During the meeting a provocative set of papers was presented both on policy orientations and on scientific aspects. The discussion led to the conclusion that several disparate approaches had already been taken, covering a variety of aqueous and non-aqueous chemical procedures and a number of different reactor and accelerator based transmutation schemes [1].

The second International Information Exchange meeting which took place in November 1992 at the Argonne National Laboratory (Illinois, USA) indicated that there was a need for guidance on the research needs and priorities. A number of emerging important issues were identified during the meeting including the legal background, the incentives and the implications of P&T for the whole fuel cycle in different countries with different nuclear policies. It was concluded that a comparison of systems studies in the field of P&T already in progress should form the central part of the P&T activities under the Committee for Technical and Economic Studies on Nuclear Energy Development and the Fuel Cycle (NDC) of the OECD/NEA [2].

The third International Information Exchange Meeting was hosted by CEA and held at the Nuclear Research Centre, Cadarache (France) in December 1994. With more than 80 participants from 11 countries, together with Russia and the IAEA, this meeting showed the wide international interest in this subject and the great progress made throughout the OECD Member countries in understanding the implications of P&T. The meeting provided some insight into the basic elements for pursuing comparative studies on the overall balance of costs and benefits of adding P&T to the nuclear fuel cycle [3].

The fourth International Information Exchange Meeting took place at Mito City, Japan, in September 1996 and was hosted by the Science and Technology Agency in combination with JAERI and PNC. About 100 participants from 11 OECD/NEA Member countries and Russia, as well as from the IAEA and the European Commission participated in the meeting, which, like previous ones, was focused on P&T systems studies. A total number of 40 papers were presented [4]. The meeting showed a far reaching consensus on the directions to be pursued. The necessity for a systems analysis report was stressed by different delegations which offered their services to the realisation of such a report.

The fifth International Information Exchange Meeting took place in Mol, Belgium, in November 1998 and was hosted by the Nuclear Research Center SCK•CEN. About 130 participants attended this meeting with a total of 43 oral presentations and 13 posters [5]. The main essence of this meeting can be characterised by the reported breakthrough in partitioning technology resulting in the Ln/An separation and Am isolation in lab-conditions. Transmutation was also discussed from a more general point of view, addressing the transmutation in LWRs (Am-targets), in FRs and in ADS. The feasibility and main technological challenges of these ADS were highlighted.

1.2 Why P&T?

Nuclear power produces steadily a mass of spent fuel which contains, apart from the short-lived fission products, a significant amount of actinides and fission products with high toxicity and very long half-lives. These nuclides constitute the long-term[a)] radiotoxic inventory[b)] which remains as a hazard far beyond human perception.

Conventional reprocessing recycles most of the major actinides (uranium and plutonium), while the minor actinides (MA) (mainly neptunium (Np), americium (Am), and curium (Cm)) with half-lives of up to 2 million years remain with the fission products which are vitrified before being buried in deep repositories. Partitioning of the minor actinides (Np, Am and Cm) and some selected fission products is a method which would reduce the long-term radiotoxicity of the residual waste components with a factor proportional to the separation yield. The recovered minor actinide nuclides would be recycled into the fuel cycle activities and returned to the reactor inventory of fissile and fertile material for transmutation to short-lived isotopes. Gradually the MAs and some long-lived fission products (LLFP) could be burned out. In principle, this technique would reduce the long-term contamination hazard in the high-level waste and shorten the time interval necessary to keep the actinide containing wastes confined in a deep geologic repository. P&T is, in principle, capable of reducing the radiotoxicity period, although a number of practical difficulties remain to be surmounted.

1.3 Why a P&T systems study?

The implications for the reprocessing fuel cycle of partitioning operations are very variable depending on the effluent or product stream in which these nuclides are carried along. Any change of the separation procedure has widespread consequences on the quality and nature of the separated nuclear products. A thorough analysis of the consequences from partitioning of MAs and FPs requires a systems analysis of the impact of the recycled products on the generated waste and on the safety items associated with the various options.

Once the MAs and LLFPs are separated, the question may be asked what has to be done with the concentrates. Conditioning and long-term retrievable storage is a first option, where matrix stability

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a) In this report, short-term refers to 100 years, medium-term 1 000 years, and long-term beyond 10 000 years.

b) For the definition of radiotoxic inventory, see *Part II, Section 4.1*.

and natural decay play an important role. Transformation into targets for later irradiation is another alternative. Irradiation of targets sometimes produces inactive transmutation products but may also lead to highly radiotoxic nuclides with medium term half-lives.

The report investigates different options to decrease the final radiotoxicity and provides a limited systems analysis of only the main options as a step towards clarifying choices among this complex set of possible alternatives. The preliminary systems analysis starts from the present technical state of the art in the fuel cycle and points to some possible developments in partitioning and transmutation technologies which would result in an advanced fuel cycle with an overall reduction of the radiotoxic inventory and a reduced impact on the biosphere.

1.4 Expert group

A systems analysis expert group representing twelve countries and two international organisations was set up in 1996. The group was commissioned to prepare a P&T systems analysis report, as comprehensive as possible, with the aim of providing decision makers with an authoritative and transparent document describing the impact of P&T on the fuel cycle and waste management policies. Particular points of interest are the consequences of any decision with regard to P&T on the fuel cycle technology, long-term safety and economics.

The expert group (see Annex A) took into consideration the different national and international systems analysis reports [6-11] already published and drew conclusions on the role of P&T in the fuel cycle and on the prioritisation of the different R&D options. Common criteria and constraints were used in order to obtain fair comparisons between the different scenarios and options.

1.5 Objectives of the report

The report as a whole is intended to provide a scientific and technical analysis of the impact of P&T on the back-end of the fuel cycle. The introductory chapters, compiled in Part I: General Overview, are conceived as an address to the decision makers at governmental and industrial levels in order to understand the P&T issues involved without going too much into detail. The technical chapters included in Part II: Technical Analysis and Systems Study, offer an in-depth analysis of the issues involved in introducing P&T into a more elaborate fuel cycle scenario as well as an analysis of the consequences from a series of operational decisions. The technical chapters are: Partitioning, Transmutation, Risk Analysis and Waste Management together with the main R&D programmes in Japan and France. The report emphasises those aspects where P&T could have an impact on the long-term safety within the context of a more elaborate fuel cycle industry and a prolonged nuclear power production strategy. Partitioning as stand alone technology might have merits in a HLW management, such as the improved stabilisation of long-lived nuclides compared with vitrification, the reduction of HLW volume and potential to recover some valuable, rare elements. However in the present report, partitioning is mainly studied as a preparatory step prior to transmutation.

The present report has to be considered as a status and assessment report of P&T with a limited systems analysis of the consequences for the fuel cycle and waste management.

2. STATUS AND EXPECTATION OF P&T TECHNOLOGY

Partitioning, the separation of the MAs and LLFPs from the spent fuel, and transmutation, the transformation of long-lived radionuclides to short-lived or stable ones by nuclear reactions, should be regarded as additions to or improvements of the reprocessing fuel cycle (RFC). In the RFC, plutonium and uranium are separated from the nuclear fuel and recycled in the nuclear fuel cycle, as shown in Figure I.1. In this figure, the RFC is divided in a “front-end” which covers all treatments applied before the use of the fuel in a reactor, and a “back-end” which covers all treatments after use in a reactor. Partitioning and transmutation are essentially back-end processes since their goal is improvement of waste disposal options. In the ideal case, implementation of P&T may lead to an advanced fuel cycle (AFC) in which dedicated technology is applied to obtain optimum benefits (see Figure I.2). However, because recycling in existing reactor types is an option that should be taken into account, P&T can also have implications for the fuel fabrication step in the RFC.

In the present chapter, the status and expectation of P&T technologies are described by considering respectively recycling strategies, reprocessing (separation) options and the technology of fuel and target fabrication. This description is not limited to the minor actinides and long-lived fission products only but also includes the recycling of plutonium and uranium since the benefits of P&T can only be judged relative to those of plutonium recycling.

Figure I.1 **A schematic diagram of the reprocessing fuel cycle for LWRs**

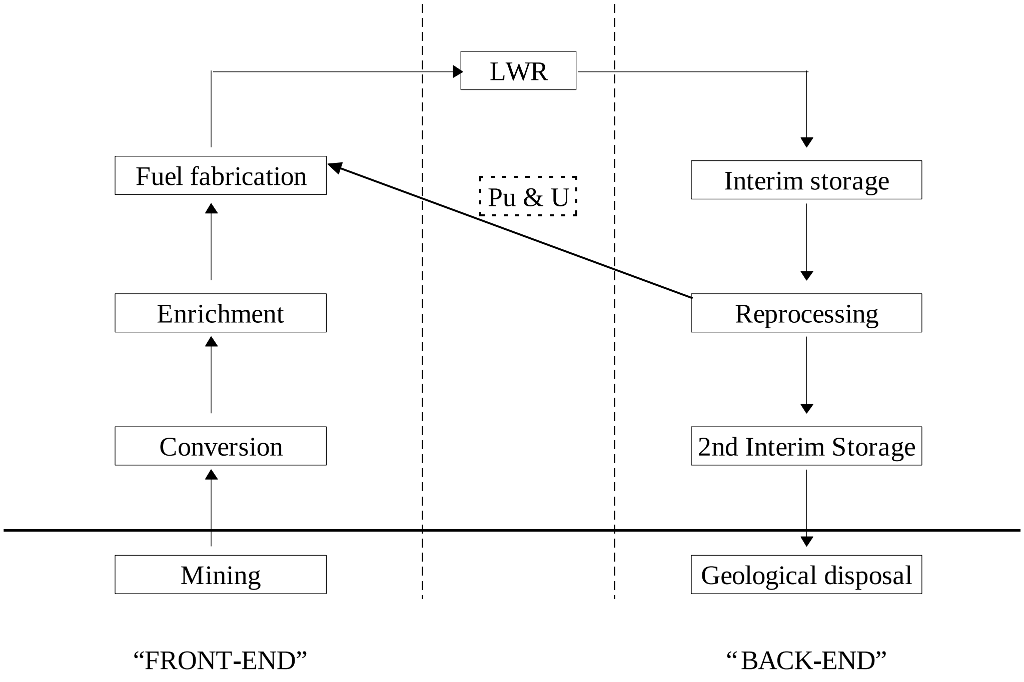
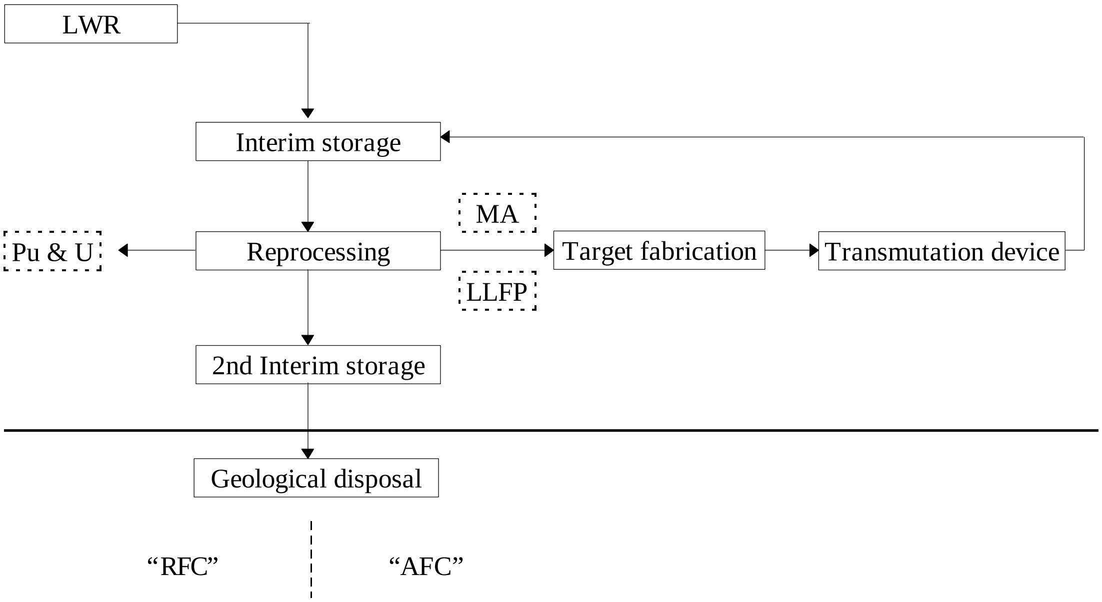


Figure I.2 **A schematic diagram of back -end of an advanced fuel cycle with minor actinide recycling**



2.1 Strategies

2.1.1 Plutonium and uranium

Separation of plutonium from spent fuel leads to a decrease in the long-term radiotoxicity of the remaining waste by a factor of, theoretically, about 10. This number simply originates from the fact that the amount of plutonium present in actual UO_2 spent fuel is about 10 times higher than that of the minor actinides of which the radiotoxicity is more or less equal to that of the plutonium isotopes. Uranium separated from the daughter products has a relatively small contribution to the radiotoxicity. However, recycling of plutonium as LWR-MOX only reduces the radiotoxicity with a factor of 3 taking into account the spent LWR-MOX fuel if considered as waste.

The simplest method to recycle plutonium is the use of mixed oxide (MOX) fuel assemblies in LWRs. A MOX assembly contains 35 kg of plutonium before the irradiation and 25 kg after. This amounts to a consumption of 10 kg per fuel assembly, which should be compared to a production of about 5 kg plutonium in a standard uranium oxide assembly. At present, a number of reactors in France, Germany, Belgium and Switzerland are being operated with up to 30% of MOX fuel assemblies in the core. In that case $\left[(2\times 5)-(1\times 10)\right]\cong$ 0 kg plutonium is produced, a near zero balance. Increasing the number of MOX assemblies in the reactor core (up to 100%) will thus lead to a net consumption of plutonium, but it requires changes in the moderator/fuel ratio. Higher consumption rates of plutonium can be achieved by mixing the plutonium with a neutron inert material, the so-called inert-matrix, since the source of the plutonium production (^{238}U) is no longer present. However, the presence of an inert-matrix will affect the Doppler coefficient of the fuel due to the absence of ^{238}U resonance. Another possibility under investigation is to mix plutonium with thorium.

The number of recycling steps in an LWR is limited due to the build-up of plutonium isotopes that are not fissionable in a thermal neutron spectrum. Therefore devices producing large fluxes of fast neutrons (fast reactors, accelerator driven sub-critical systems) remain necessary to incinerate the degraded plutonium resulting from plutonium recycling in LWRs. Fuel management calculations have shown that such a device can be operated in symbiosis with a park of LWRs. In Europe, the CAPRA project has been defined to design a fast reactor which will consume as much plutonium as possible. In the current CAPRA reference design, a mixed oxide core is foreseen with MOX fuel containing up to 45% plutonium oxide. However, this fuel does not dissolve in nitric acid solution as used in the conventional PUREX process and alternative fuels are therefore being considered. The present focus in Japan but also in Europe is on nitride fuels, since PuN easily dissolves in HNO_3, the aqueous medium in the PUREX process. This option requires the use of fully enriched ^{15}N reagents for PuN production in order to avoid ^{14}C formation.

Presently, reprocessed uranium (REPU) is not being significantly recycled as a result of the low price of natural uranium and the fact that it contains some ^{236}U which is a neutron poison and decreases the reactivity of reprocessed uranium. Delay or absence of recycling of REPU in reactors will lead to the build-up of the very radiotoxic decay products of ^{232}U and ^{234}U particularly ^{228}Th and ^{208}Tl. Both stocks of REPU and of the depleted uranium from enrichment will need to be taken into account in overall strategies for radioactive waste management. In a very long-term perspective, the total radioactivity of depleted uranium if considered as waste material exceeds that of neptunium.

2.1.2 Minor actinides

If the recycling of plutonium can be achieved effectively on an industrial scale, the recycling of americium should be considered next because of the following reasons:

- americium has the second highest contribution to the radiotoxicity in spent fuel;
- in the performance assessment of underground repositories, americium dominates the radiotoxicity during the first 1 000 years;
- ^{241}Am is the precursor of long-lived ^{237}Np which generally dominates the normal evolution scenarios in the performance assessment because of its long half-life;
- plutonium recycling increases the Am production.

As for plutonium and uranium, the most favourable transmutation reaction for the minor actinides is fission since capture or (n, 2n) reactions generally produce other long-lived actinides. The fission of the minor actinides can best be achieved in a fast-neutron flux in which most actinide isotopes are fissionable. Even in this case, capture followed by fission is still an important process. In thermal spectra, extra neutrons are required to convert the non-fissile into fissile isotopes (e.g. ^{241}Am into ^{242m}Am) requiring extra fuel enrichment.

It is not surprising that the strategies for minor actinide transmutation in fast flux devices are similar to those for Pu transmutation: they can either be mixed homogeneously in MOX fuel to yield the so-called MINOX fuel, or loaded in special fuel assemblies as inert-matrix fuels, based on oxides or possibly on nitrides and carbides. From a reactor physics point of view, both options are feasible. However, limitations are set by the fuel fabrication.

Instead of recycling, one could adopt a strategy in which as many minor actinides as possible are incinerated in a single, extended irradiation, followed by final disposal, a so-called "once-through" option. This can be done in a high flux thermal irradiation facility or in a moderated subassembly of a fast reactor. By selecting an appropriate inert-matrix, which immobilises the residual waste formed, the radiological effects of storage might be further reduced, since an extra barrier is introduced. Geologically stable minerals like zircon ($ZrSiO_4$) or monazite ($CePO_4$) are being considered for this purpose.

2.1.3 Fission products

From the point of view of reduction of radiotoxicity, transmutation of fission products is of very little interest. The majority of the fission products has decayed after about 250 years, and the 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 underground disposal. In addition, the treatment of spent fuel results in releases through gaseous and liquid effluents which also contributes to the long-term radiological effects of nuclear power generation. Separation and/or capture from the waste streams, followed by transmutation could be means to reduce the long-term radiological hazards. The fission products that deserve most attention in this respect are technetium (Tc), caesium (Cs) and iodine (I), since ^{99}Tc and ^{135}Cs are the dominant isotopes in risk analyses of spent fuel disposal and ^{129}I, which is not incorporated in the vitrified HLW, is the dominant isotope in the radiological effects of reprocessing effluents or even from spent fuel in certain geological formations.

Technetium present as a single isotopic species (^{99}Tc) can be transmuted by single neutron capture into the stable noble metal ruthenium (^{100}Ru). However, transmutation of ^{99}Tc in thermal reactors such as present-day LWRs will be difficult because of the very long transmutation half-lives and the large inventories required. Better results can be obtained in heavy-water reactors, moderated subassemblies of fast reactors and presumably in future accelerator-driven high-flux reactors. In all cases, additional enrichment of the fuel is required unless isotopic separation techniques are applied.

Unlike technetium, caesium separated from spent fuel is not a single isotope but a mixture of the long-lived ^{135}Cs, the short-lived ^{137}Cs and the stable isotope ^{133}Cs, all present in about equal quantities. As a consequence parasitic neutron capture in especially ^{133}Cs will occur during irradiation. Taking also into account the relatively low neutron absorption cross-section of ^{135}Cs, transmutation of caesium cannot be considered feasible.

Though the iodine separated from spent fuel is a mixture of ^{129}I and ^{127}I, the fraction of the latter is tolerable (16%). Transmutation of ^{129}I can theoretically be achieved by single neutron capture, yielding the noble gas xenon (^{130}Xe). But also for the transmutation of iodine in thermal reactors, large reactor loadings and long irradiation periods are required.

2.2 Reprocessing

2.2.1 Plutonium and uranium

Reprocessing of spent fuel from LWRs is being done on an industrial basis in several countries (France, United Kingdom, India and Russia). In these plants, fuel rods from light water reactors are treated to recover uranium and plutonium. MOX fuel elements can also be processed when diluted with standard UO_2 elements to comply with dose and Pu inventory limitations. Reprocessing of fast reactor fuel with high Pu-content, such as that used in the current CAPRA designs, cannot be done in the existing reprocessing facilities since it does not dissolve quantitatively (i.e. completely) in the standard nitric acid solution.

Reprocessing of spent fuel is adopted in several OECD Member countries, India and Russia. Important exceptions are Canada, Spain, Sweden and USA, where direct storage of fuel is foreseen. In all reprocessing plants, the recovery is being done with the PUREX process, a wet chemical extraction process based on nitric acid dissolution of the fuel and solvent extraction of uranium and plutonium by tri-butyl-phosphate (TBP). The present state of the art in reprocessing allows the separation of uranium and plutonium from LWR fuels with efficiencies of 99.88%. The remaining 0.12% is being incorporated in the vitrified waste or encapsulated together with hulls and end-fittings and embedded in concrete.

As the fuel remains property of the utility, all waste products of the reprocessing (vitrified and concrete waste) are returned to the country of origin. Per 1 000 kg of fuel of 33 GWd burn-up, 955 kg are reprocessed uranium, 10 kg plutonium and about 2 m^3 vitrified and concrete waste is produced. The present prospects show that this waste volume will be reduced even more in the future by optimising the reprocessing process and the high-level waste management.

The costs of reprocessing depend of course on the timing, burn-up, etc., but reasonable estimates by OECD/NEA show that it is of the order of 720 ECU/kg-U. This amount should be considered together with the costs of the other steps in the back-end of the fuel cycle and compared to those of direct storage. Savings are made in the mining and milling of fresh uranium and in the enrichment services. However, the savings in demand of natural uranium and of enrichment services for new fuel are the most important incentives for plutonium recycling and the use of LWR-MOX fuel elements.

In some countries, the reprocessing of spent fuel by pyrochemical processes in molten salts is also being investigated. The major advantage of this technology is the much higher radiation resistance of the molten salt as a result of which shorter cooling times can be applied. The molten salt technology was developed for the Integral Fast Reactor/ALMR concept in the USA, and continues to be investigated by the Japanese institute (CRIEPI).

2.2.2 Minor actinides

Presently, the separation of the minor actinides neptunium (Np), americium (Am) and curium (Cm) is not being done in the commercial reprocessing facilities and the elements are incorporated in the vitrified high-level waste, together with the fission products. This is not only due to the fact that up to now there was very little incentive to separate these elements, but also due to the impossibility of separating the trivalent actinides (Am, Cm) in the PUREX process. The recovery of neptunium, in contrast, can be achieved effectively using a modified PUREX process: by adjusting oxidation state Np is extracted with U-Pu and can be separated from the heavy metal stream, either as single Np element fraction, or co-extracted with uranium, or routed towards the plutonium stream.

A lot of research is being done to discover efficient methods to separate the trivalent actinides from the PUREX waste stream and several processes have been developed in recent years: DIAMEX in France, DIDPA in Japan, TRPO in China and TRUEX in the USA and Japan. The most difficult point to address there is the separation of trivalent actinides (Ans) from trivalent lanthanides (Lns). Since the ratio between the lanthanides and the actinides varies between 10 and 20 according to the spent fuel burn-up, purification of trivalent MA (Am-Cm fraction) is only possible when high separation factors are achieved. Solutions to this problem are under study in the framework of the development of DIDPA and TRUEX processes. Moreover, other processes are under study based on the use of new extracting molecules. A great effort is being devoted to the fundamental study of extractants and complex systems capable of discriminating between actinides and lanthanides. Recent developments in China, Germany, France and Japan on "CYANEX 301" compounds are apparently very promising.

As an alternative to aqueous processing methods a great effort has been accomplished on the development of pyrochemical separation methods in the USA (ANL) and Japan (CRIEPI). These dry reprocessing techniques are suited for the treatment of high burn-up, shortly cooled spent fuel since they are not sensitive to radiation damage and display much less severe criticality constraints. Metallic alloy transuranic elements (TRU) type fuel can be treated directly by a combined electro-refining and reductive extraction process in molten salt and molten metal (Cd) bath. This technology has been studied at the conceptual design and the experimental laboratory level. The use of high-temperature processes and corrosive reagents make this approach more difficult for large scale industrial application. However, it may be anticipated that multiple recycling of high burn-up fast reactor fuel will be very difficult with aqueous methods, leaving the pyrochemical techniques as a valid alternative for the future.

2.2.3 Fission products

In the PUREX process, the fission products are distributed over three different waste streams:

- gaseous effluents that are released in the air and principally contain the noble gas ^{85}Kr;
- low-level liquid effluents that are released into the sea or ocean and contain a majority of ^{129}I;
- high-level liquid waste (HLLW) in which all other fission products are concentrated.

During the dissolution step in the PUREX process, iodine in the fuel is oxidised to elemental iodine. Depending on the process conditions, it can be released from the solution to the off-gas or it can be concentrated in the solution. The former method permits a very efficient separation route but this needs to be followed by a trapping from the off-gas. This is normally done by counter-current scrubbing and by the use of silver impregnated molecular sieves, which can reach decontamination factors of about 10^4.

Technetium occurs after reprocessing partially in an insoluble species together with the other platinum metals (Ru, Rh, Pd) and in the soluble species $^{99}\mathrm{TcO}_4^-$. The separation of soluble technetium is technically feasible with some adaptations of the PUREX process. It can be separated as pertechnetic acid ($HTcO_4$) using monoamide extraction molecules or active carbon absorption method. A precipitation method by reducing nitric acid concentration is also effective for Tc separation from HLLW. Prior to any transmutation, both fractions have to be recombined.

Caesium is left in the HLLW during the PUREX process. An adsorption method with inorganic ion exchanger has been developed for Cs separation and successfully demonstrated with real HLW. Recently, a new method to separate Cs from this solution has been developed, on a laboratory scale, using functionalized macrocyclic molecules (calixarenes) as carrier in supported liquid membranes. By Cs permeation through the membrane, more than 99.8% of the caesium can be removed from acidic solutions.

2.3 Fuel and target technology

2.3.1 Plutonium and uranium

The fabrication of MOX fuel assemblies is current technology, available on a commercial basis. The present capacity of the fabrication plants in Belgium, France, United Kingdom and Japan is about 200 tHM/year but extension of the capacity to more than 300 tHM/year in the year 2000 is foreseen. MOX fabrication is about four times more expensive than the regular uranium oxide fabrication ($275~300 /kg-U) owing to increased safety requirements which principally concern the radiological protection of the workers by performing the fabrication processes in glove-boxes and the safeguards system. On the contrary, recycling of plutonium will lead to savings in the front-end of the fuel cycle, specifically mining and enrichment, which are the major contributors to the dose rates of the workers and to the costs of the front-end of the fuel cycle.

Inert-matrix fuels are a recent development to obtain optimal incineration rates. In such fuels, the compound(s) to be transmuted is (are) mixed with a neutron-inert material. This can be done as a dispersion (macroscopic scale) or as a solid solution (microscopic scale). Typical materials that are being considered as inert-matrix are oxides such as spinel ($MgAl_2O_4$), yttrium oxide (Y_2O_3), yttrium aluminium garnet ($Y_3Al_5O_{12}$), metals such as tungsten or vanadium, or silicon carbide (SiC). However, additional research needs to be done on the fabrication and characterisation of such fuels, especially with respect to the irradiation behaviour and their compatibility with current reprocessing techniques.

In addition to oxide fuels, nitride and metal fuels are being considered for the incineration of plutonium. Both new fuel types are compatible with liquid sodium and can therefore be considered for fast burner reactors. Nitride fuels, as proposed for CAPRA type burner reactors, have been produced on a laboratory scale in the 1960s. As mentioned before, the present interest in nitride fuels originates partly from the fact that oxide fuels with high plutonium content do not dissolve in nitric acid, whereas nitride fuels do. Another advantage of nitride fuels is their much higher thermal conductivity, which will lead to lower central fuel temperatures and, hence, increased safety margins. The major disadvantage of nitride fuels is the ^{14}C production as a result of (n,p) reaction on ^{14}N. Enrichment in ^{15}N of the nitrogen used for the fuel fabrication is therefore a requirement and the recycling of enriched nitrogen would be facilitated by using pyrochemistry as investigated in Japan.

In the USA, metal fuels consisting of an alloy of uranium, plutonium and zirconium (Zr) was investigated for the IFR concept and production has been achieved on a pilot scale. Like nitride fuels, metal fuels have much better thermal conductivity than oxide fuels, in addition, they have good radiation stability as a result of which very high burn-ups can be reached. In the IFR/ALMR concept, the metal fuel would be reprocessed by molten salt techniques.

2.3.2 Minor actinides

For neptunium, recycling in MINOX fuel seems to be a feasible solution as it can be done in existing MOX fabrication facilities with minor adaptations. These adaptations principally concern improvement of the biological shielding and enhancement of the level of automation to cope with the higher radiation levels. This will lead to an increase of about 20% of the fabrication costs. Fuels with the composition $(U_{0.55}Pu_{0.40}Np_{0.05})O_2$ have been prepared successfully at the Institute for Transuranium Elements (ITU) in Karlsruhe for the CAPRA irradiation campaign.

Only very small amounts of americium can be added to UO_2 fuel due to dose limitations during fabrication. The high γ-dose is not only due to americium itself but also due to the fact that some lanthanides will be present as impurities. As a consequence, incineration of americium on a large scale can be done most effectively in an inert-matrix fuel which has to be prepared in specially designed facilities with a very high level of shielding and remote handling. Similar materials as for the fuels for plutonium incineration (see above) are being considered as inert-matrix for americium. However, the properties of americium oxide (AmO_2) are not very favourable: it has a poor thermal conductivity, a high oxygen potential and reacts with liquid sodium.

Recycling of metal and nitride forms containing or not an inert support material might, therefore, be more advantageous for the incineration of americium and possibly other MAs. This option is being studied at JAERI.

Recycling of pure curium in dedicated systems does not seem feasible at the moment because of the very high α, γ and neutron radiation due to decay and spontaneous fission. One option is interim storage of curium for about 100 years, after which the relatively short-lived curium isotopes (^{242}Cm, ^{243}Cm and ^{244}Cm) have decayed to plutonium isotopes which can then be recycled as described above. However, an effective separation method of Am from Cm is a major prerequisite for some scenarios. But recycling of a mixture of Am and Cm is still being considered.

2.3.3 Target selection for fission products

If transmutation of the fission products technetium and iodine is considered, they will most likely be irradiated in special targets. Research on the selection of suitable materials as well as pilot irradiation experiments have been done in the frame of the European EFTTRA collaboration. The preliminary results of these studies have shown that metallic technetium can be used as target material: a fabrication route for casting the metal into rods has been developed and irradiation experiments in a thermal spectrum (to a burn-up of about 6%) did not show any evidence for the swelling or disintegration of these rods.

Iodine cannot be transmuted in its elemental forms due to the volatility and chemical reactivity. Iodide metal compounds are therefore being considered. The experiments performed in the frame of the EFTTRA co-operation have shown that sodium iodide (NaI) is the best candidate if transmutation of iodine is being considered.

3. DESCRIPTION OF THE FUEL CYCLES

Definition of partitioning and transmutation

Partitioning is a complex series of chemical and/or metallurgical operations, intended to separate selected radiotoxic nuclides or groups of nuclides from the bulk of radionuclides occluded in the spent fuel elements which are periodically discharged from nuclear reactors. The separated nuclides or group of nuclides can be stored as such or transformed into new fuel elements or irradiation targets.

Transmutation is a general term covering as well elementary nuclear conversion through single neutron capture as fission of heavy nuclides, spallation and other nuclear reactions involving neutrons. The aim of transmutation in the context of this study is to reduce the long-term inventory of radiotoxic nuclides by converting the initial nuclides either into short-lived radionuclides or into stable nuclides.

3.1 The nuclear fuel cycles

The nuclear fuel cycles include all the operations necessary to supply fresh fuel to the power plants and to manage the spent fuels discharged from the reactors. Figure I.3 shows the main steps of the reprocessing fuel cycle (RFC), which are the following for light water reactors:

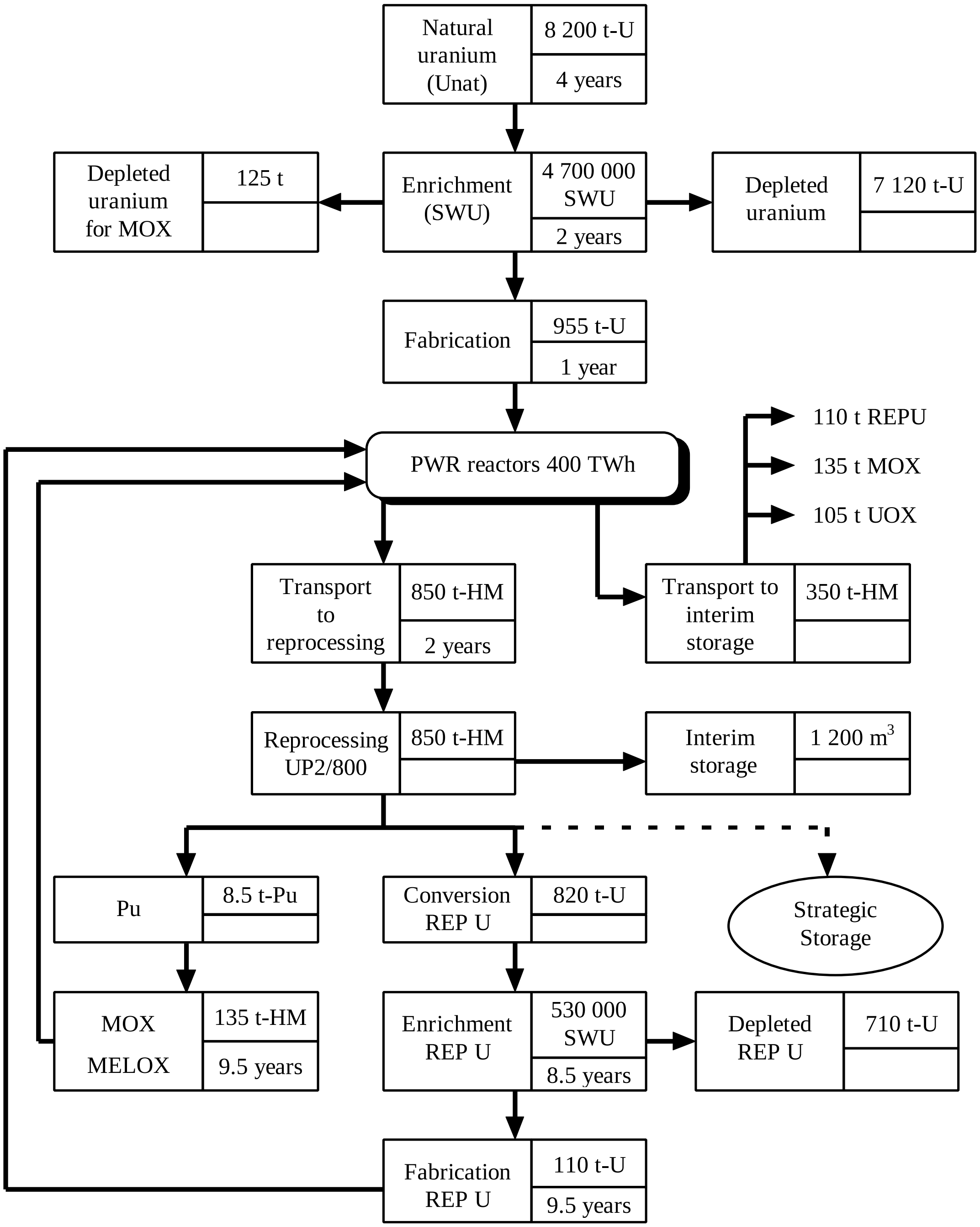
- the front-end of the cycle covers operations ranging from uranium ore prospecting to the transport of fuel elements to the power plant for refuelling: ore extraction and treatment to produce a uranium concentrate, conversion of the concentrate to hexafluoride sent to uranium enrichment plants to raise the content of the isotope ^{235}U from its natural level (0.72%) to a level of 3 to 5%, and fuel element fabrication.

- the back-end of the cycle comprises two complementary alternatives today:
  - the closed cycle based on spent fuel reprocessing, which is designed to separate and recycle the energy materials (mainly plutonium) which they contain, and to optimise waste management for disposal;
  - the so-called “open cycle” with direct disposal of the irradiated fuels in a geological repository after an interim storage period of variable length.

Figure I.4 shows an example of the annual material flows in a fuel cycle with a mixed PWR-FR park. Figure I.5 shows a fuel cycle where only FRs are used for energy productions.

In the present report, four types of fuel management are considered.

Figure I.3 **A schematic diagram of the reprocessing fuel cycle for a 400-TWh LWR park (French case study)**



In this chart it is assumed :

- an adaptation of the reprocessed quantitis of UO_2 fuel to the recycling possibilities of Pu in Mox fuels, and
- a full capacitiy of UP2 La Hague for reprocessing (850 t/y) and MELOX plant (135 t/y) for MOX fabrication.

Figure I.4 **A materials flowsheet for a 700-TWh mixed PWR-FR nuclear electricity grid (an example calculated by JNC, Japan)**

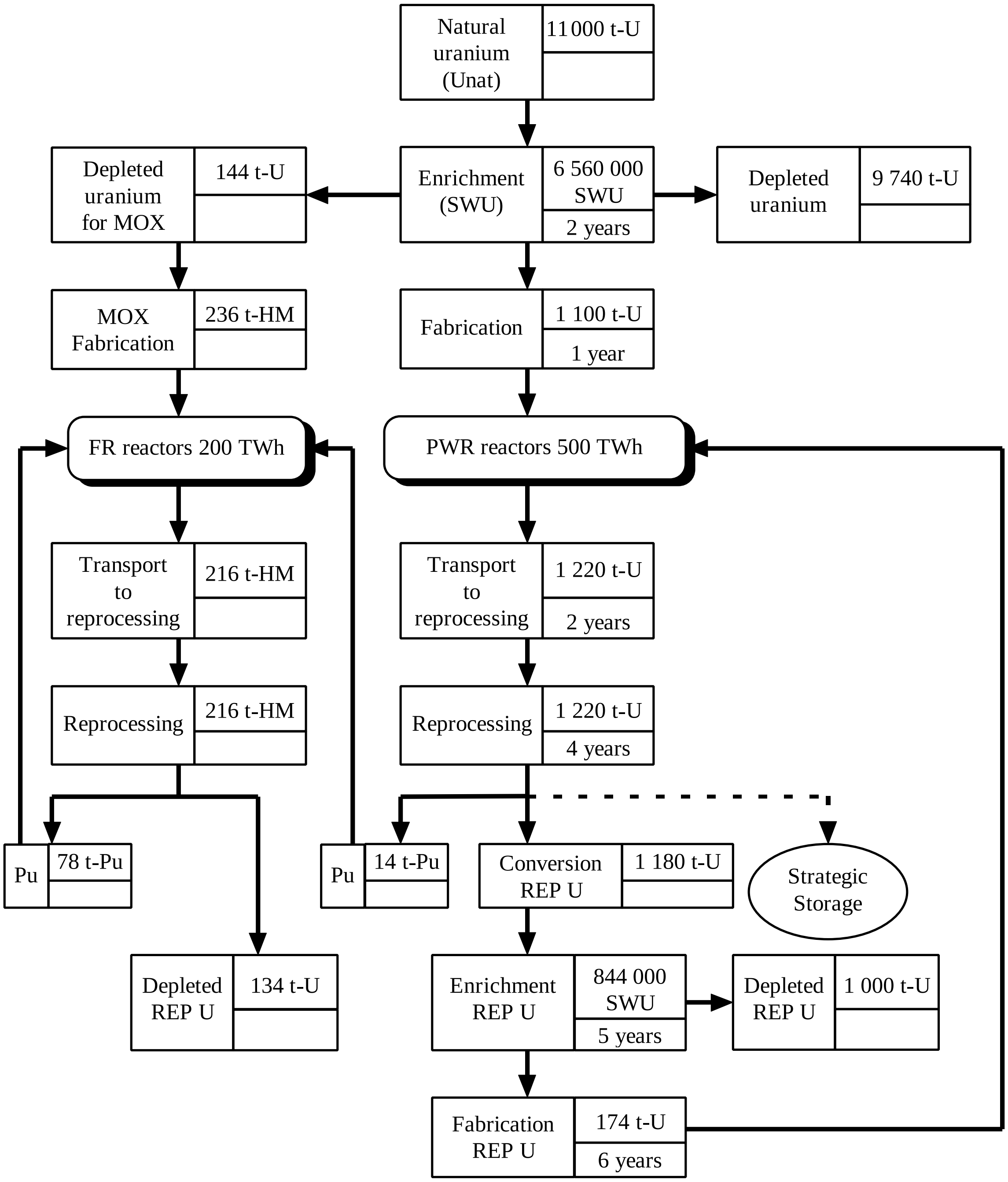
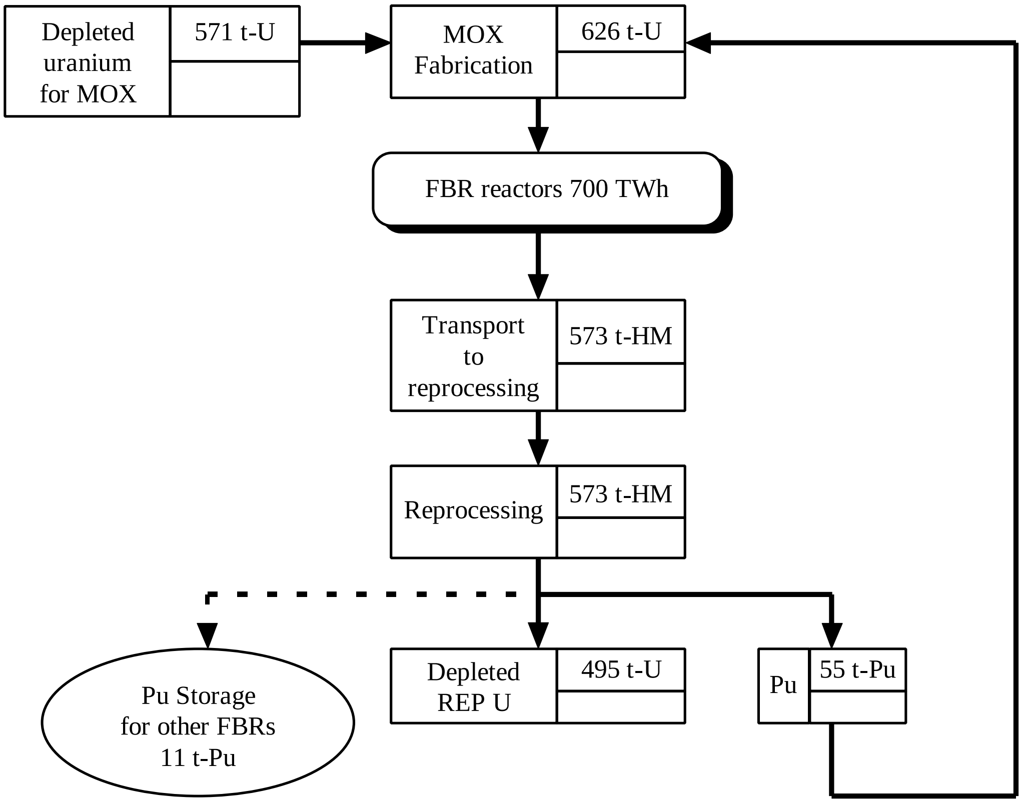


Figure1.5 **A notional materials flowsheet for a 700-TWh FR park (an example calculated by JNC, Japan)**



3.1.1 Once-Through Fuel Cycle (OTC)

The OTC is the scenario by which all spent fuel discharged from a nuclear reactor is stored in engineered facilities till sufficiently cooled to be transferred to a permanent disposal site in deep geological formations.

3.1.2 Reprocessing Fuel Cycle (RFC)

The RFC scenario incorporates the reprocessing step which aims at the recovery of uranium and plutonium from dissolved fuel elements and the transfer of FPs and MAs (Np, Am, Cm) to the High-Level Liquid Waste (HLLW) which is stored for a number of years until vitrification of this highly active solution. The glass-blocks are stored in engineered facilities until their transfer as HLW to a geological repository.

3.1.3 Advanced Fuel Cycle with TRU and selected FP recycling (AFC)

The AFC is a series of chemical, metallurgical and nuclear operations by which all the actinides (Pu, Am, Cm, Np) and some selected fission products (^{99}Tc, ^{129}I) are separated from the main stream and recycled as targets into the NPPs or into dedicated nuclear reactors and/or ADSs to obtain a significant reduction of their radiotoxic inventory.

3.1.4 Double strata fuel cycle

A combination of the RFC and the AFC leads to the set-up of a mixed energy producing and waste incinerating reactor park. In the Double Strata Fuel cycle concept, the P&T cycle, as an additional cycle to a commercial fuel cycle, is dedicated for the management of HLW produced only in the reprocessing process of the first cycle. The features of this concept are the independence of the P&T cycle from the commercial fuel cycle and the significantly less throughput of heavy metals in the P&T cycle.

The quantities and characteristics of the nuclear fuel materials and unloaded fuel assemblies are shown in Annex E. In order to improve the understanding, these data refer only to PWR irradiation. For BWR, CANDU and other types of reactors, these data are different but have not been included to avoid a multiplication of tables.

3.2 Overview of the fuel cycle and associated issues

3.2.1 The Once Through Cycle (OTC)

The OTC scenario is the main alternative for Canada, Spain, Sweden, USA and some other countries. This scenario gives, with the present low uranium prices, the cheapest nuclear energy production. However, it implies that the residual fissile material content (1% Pu and 0.8% ^{235}U) as well as the remaining fertile material (^{238}U) of the spent fuel will not be recovered.

The long-term potential radiotoxicity of spent nuclear fuel is associated mainly with the actinide elements particularly the transuranium nuclides (TRU = Pu, Np, Am, Cm …). These constitute over a very long time period (hundred thousand years) a significant radiological source term within a spent fuel repository. However, the intrinsic insolubility of actinides in deep geological formations contribute to the effective isolation of TRU.

The FPs are, in the short term, the most limiting factor in designing the repository facilities due to the γ-radiation and the decay heat emission that increases proportionally with the burn-up. After some 300 to 500 years, the major part of the FPs have decayed except for some long-lived nuclides (^{135}Cs, ^{99}Tc, ^{129}I, ^{93}Zr …). Some of these are relatively mobile in the geosphere and may contribute to the dose to man.

The long-term radiological impact of the OTC can be controlled by a man-made system and natural barriers which should provide protection for as long time as the life of the radiological source term they confine. The long-time periods involved require a careful analysis of the processes involved and of the long-term consequences for conceivable scenarios.

At the present time, there is no world-wide agreement on the time intervals for confinement of high-level radioactive wastes in a geologic repository. Periods of 1 000, 10 000, 100 000 years or even longer have been considered as a target but no internationally accepted confinement period has been established.

A few specific regulatory and safety aspects are associated with the OTC scenario:

- the potential for criticality has to be addressed in the licensing process of a spent fuel repository because substantial quantities of fissile material will be deposited.
- the heat emission, beyond 500 years, of spent fuel due to the total actinide content is significantly higher than of the fission products alone and has to be accounted for in the design of a repository. The absolute value of that long-term heat output is, however, significantly lower than that of the initially loaded fission products.
- in the USA, the maximum inventory of spent fuel is limited to 70 000 tHM per repository. Thus for a large country with a large number of nuclear reactors, like e.g. USA, a new repository may have to be installed about every 30 years.

3.2.2 The Reprocessing Fuel Cycle with U and Pu recycling (RFC)

Since natural uranium contains only 0.72% of fissile ^{235}U isotope, the recycling of uranium and plutonium from spent fuel through the RFC has been from the beginning of the nuclear era the standard scenario of nuclear energy production. There has been reduced support for this approach in many OECD countries in recent years owing to economic factors and concerns of non-proliferation policy.

By proceeding this RFC scenario the major fraction (99.9%) of the uranium and plutonium streams are extracted and only a very minor fraction of the so-called "major actinides" are transferred to the HLLW (and consequently to the HLW) and eventually to the geologic repository.

Partitioning of MAs from HLLW in order to reduce further the radiological potential of HLW has been studied since the 1970s. Initially, the R&D activities were focused on the complete removal of MAs in order to eliminate the need for any long-term storage or final disposal in geologic formations. This option was abandoned because it is unrealisable.

However, if the public and/or political acceptance of very long-term disposal of HLW could not be obtained, the removal of MAs from HLLW is a technical solution which might reduce the residual radiotoxicity of the HLW. Moreover, with increasing burn-up, the generation of MAs becomes more and more important. The addition of a MA partitioning module to the standard reprocessing plant would, in such a case, be the most obvious change to the current RFC. Countries with a reprocessing infrastructure (France, UK, Japan, India, Russia and China) and their associated partners could in the medium term realise a partial partitioning scenario by which the HLW would be practically free from long-lived TRUs.

However, the question arises what to do with the recovered U, Pu, and MA fractions. The countries which chose to reprocess their spent fuel did this with the main purpose of recovering the major actinides (U and Pu), to save fresh uranium purchase (20%) and to use the residual fissile components of the spent fuel (ca. 1% ^{235}U, 1% Pu), corresponding to about 25% of the regular SWU expenses in the uranium enrichment step.

For a number of decades the plutonium recycling was envisaged in a fast reactor (FR) option but for economic and political reasons, this long-term option of nuclear energy production has been slowed down and sometimes even put to an end. The stock of plutonium already accumulated at the reprocessing plants and which was intended to be used in LMFBRs became redundant in a cheap uranium market economy.

Some years ago, the recycling in LWRs of plutonium recovered in the reprocessing operations became an industrial practice. The use of LWR-MOX in a RFC option got industrial significance in Western Europe where increasing quantities of PuO_2 were transformed into LWR-MOX fuel and irradiated in specially licensed reactors in France, Germany, Switzerland and Belgium. The reuse of plutonium is to a certain extent a first step in a global P&T scenario which has to be brought into broader perspective of reuse of resources and reduction of the long-lived waste produced during the nuclear age.

The recycling of plutonium as LWR-MOX is, a posteriori, a step to recover the expenses made during the conventional reprocessing step by producing a fraction of the electricity in a subsequent reactor core. But from radiotoxic point of view, the overall gain is rather limited since only ~25% of the recycled plutonium is fissioned and about 10% is transformed into a long-term radiotoxic MA source term. For the present, recycling of spent LWR-UO_2 fuel as MOX provides an overall reduction of mass (a factor of about 5) of this radiotoxic source term, but this recycling does not significantly reduce total radiotoxicity. Multiple recycling of LWR-MOX is theoretically possible in LWRs but the resulting radiotoxicity increases throughout the subsequent recycling campaigns.

If it were required to reduce further the global radiotoxic inventory, it would be necessary to transfer the TRU content of spent LWR-MOX fuel into a FR-MOX fuel cycle scenario which becomes a part of the next-generation scenario: the AFC. In the meantime, retrievable storage of spent LWR-MOX fuel is the most appropriate fuel cycle option until a fast neutron “incinerator” technology becomes industrially available. Reprocessing of spent LWR-MOX fuel with a view to recycling the TRUs in a FR producing energy is conceptually possible in the present plants.

The HLLW produced during reprocessing of LWR-MOX fuel would require an additional TRU separation module to reduce significantly its radiotoxicity in comparison with non-reprocessed spent LWR-MOX fuel. The same technology as that proposed for HLLW from LWR-UO_2 would be adequate.

The recovery of uranium from spent fuel was, from the beginning of the nuclear era, an obvious option which was realised by its extraction in the PUREX process carried out with TBP. For many decades uranium has been recovered during reprocessing but very little of this stockpile has been reused in subsequent reactor loading. Reprocessed uranium contains some troublesome radioisotopes like ^{232}U which is the parent of natural decay chains with radiotoxic daughter nuclides, or like ^{236}U which forms ^{237}Np by irradiation.

3.2.3 The Advanced Fuel Cycle with TRU recycling (AFC)

Any AFC scenario must rely on the use of any form of reprocessing of spent fuel as an indispensable first step.

A comprehensive AFC scenario with P&T comprises the following steps:

- reprocessing of LWR-UO_2 fuel;
- separation of MAs from HLLW resulting from LWR-UO_2 reprocessing;
- fabrication of MA targets for heterogeneous irradiation in LWRs;
- quantitative recycling of U and Pu into LWR-MOX fuel (single or multiple recycling);
- reprocessing of spent LWR-MOX fuel in adequate facilities (higher Pu inventory);
- separation of MAs from HLLW and conditioning of individual elements (Np, Am, Cm);
- fabrication of FR-MOX, -metal or -nitride fuel with limited MA content;

- irradiation of FR-fuel in Fast Burner Reactors (FBuR) or dedicated hybrid facilities (very high burn-up);
- reprocessing of spent FR-fuel in specially (aqueous or non-aqueous) designed and licensed facilities;
- quantitative separation of all TRUs from the spent FR fuel processing during multiple recycling;
- multiple recycling of FR-MOX fuel with major TRU content until significant depletion;
- separation of certain fission products with long half-lives if required for the disposal step;
- revision of the fission product management: ^{99}Tc separation (head-end, HLLW); and
- platinum metals separation and recovery.

The most important of these steps will be briefly commented.

P&T has to play an essential role in the future AFC which is intended to reduce as much as possible the long-term radiotoxic inventory. However, modifications to be implemented in present facilities or to be designed for future reprocessing plants with incorporated partitioning steps are of prime importance for the successful implementation of the P&T option in the AFC fuel cycle.

3.2.3.1 Impact of reprocessing of LWR-UO$_2$ fuel

The current separation efficiency of (99.9%) obtained for major actinides is in a first approach sufficient to reduce their content in HLLW to an acceptable level. The only improvement which might have a significant influence on the long-term risk and waste management is the reduction of medium-level waste (MLW) which is not vitrified before disposal and occupies a rather large volume compared to HLW.

3.2.3.2 Separation of MAs from HLLW resulting from spent LWR fuel

During the reprocessing operations most of the MAs are transferred to the HLW. Am and Cm (together with shorter-lived TRUs, Bk, Cf …) are quantitatively (>99.5%) transferred to HLLW. Np goes directly or indirectly to the HLLW apart from a proportion (usually small) to the plutonium product. The partitioning of Np needs further chemical extraction steps to ensure a quantitative transfer to one single waste or product stream, from which further separations could be performed.

Recovery of ^{237}Np from the U-Pu product stream is technically possible in the PUREX process. The separation of ^{241}Am is also a prerequisite for a significant reduction of the long-term radiotoxicity due to ^{237}Np.

Partitioning of all MAs from HLLW is presently under investigation in many laboratories throughout the world (Japan, France, China and some other countries) and was studied formerly in the US national laboratories (ANL, ORNL, Hanford …). Several processes have been studied at the conceptual level and tested in hot facilities; among the most important are: the TRUEX, DIDPA, TRPO, DIAMEX, and CYANEX 301™ processes. Non-aqueous partitioning is being investigated in Japan.

Separation of Am (plus Cm) from HLLW is the first priority from the radiotoxic point of view but the Am (Cm) fraction contains all the rare earth (RE) elements which are in terms of quantity, about 10 to 20 times more important than actinides depending on the burn-up. At 45 GWd/tHM, the ratio is 16 (13.9 kg RE compared to 0.870 kg Am-Cm). In order to obtain a TRU fraction with 90% purity, a decontamination factor of more than one hundred is required for the RE fraction. A 99% purity involves a separation factor of more than 1 000, which is at the technical limit for elements such as the lanthanides and the actinides with very similar chemical properties. Recent developments in chemical separation techniques have improved the prospects for solving that problem.

The adaptations to be made to the current flowsheets and their translation into technological realities are a difficult task which is outside the scope of this report. Future reprocessing plants will possibly include MA and LLFP partitioning rigs from the design phase on.

3.2.3.3 Fabrication of MA targets for heterogeneous irradiation in LWRs

In the medium term, only thermal reactors and particularly LWRs are available for irradiation of MA-loaded fuel or targets. Fabrication of irradiation targets with industrially representative quantities of MAs is difficult to accomplish even in pilot-scale hot-cell facilities.

Experience has been gained in the production of isotopic heat sources, but the present radiologic context and the ALARA limitations to be expected from regulatory bodies on industrial activities render industrial recycling of MAs very different from what has been done in the past for military and space applications.

The presence of large quantities of ^{241}Am accompanied by 1 to 10% RE will require fully gamma shielded and remotely operated fabrication facilities. The presence of 5% ^{244}Cm in an ^{241}Am-^{243}Am target will amplify the degree of technical complexity due to the additional neutron shielding resulting from the spontaneous fission rate and from the α-n reaction in oxide-type isotopic targets.

3.2.3.4 Separation of long-lived fission and activation products

A number of radiologically important fission/activation products play a potentially important role in the assessment of a geologic repository and have been considered in a P&T option. The following nuclides have to be assessed: the fission products, ^{99}Tc, ^{129}I, ^{135}Cs, ^{79}Se, ^{93}Zr and ^{126}Sn, and the activation products, ^{14}C and ^{36}Cl.

^{99}Tc is a fission product with a half-life of 213 000 years which occurs as Tc metal and TcO_2 in the insoluble residues and as pertechnetate ion in the HLLW solution. Its radioactive concentration in spent fuel of 45 GWd/tHM is 6×10^{11} Bq/tHM and its radiotoxicity is characterised by an annual limit of intake (ALI) by ingestion of 3×10^{7} Bq/year. In order to effectively address the long-term radiotoxicity problem both fractions ought to be combined before any nuclear action is taken towards depletion by transmutation. The extraction of soluble Tc is relatively easy. The similarity between Tc and the platinum metals in insoluble waste and the nature of the separation methods (pyrochemical techniques) makes this partitioning operation very difficult. Tc displays only a radiotoxicological hazard when submitted to oxidative underground conditions (Tuff, Yucca Mountains). In reducing deep underground aquifers the migration of ^{99}Tc is negligible.

^{129}I is separated from the HLLW during the conventional reprocessing operations. It occurs in the scrub liquids of the dissolver. The radioactive concentration in spent fuel is 1.6×10^{9} Bq/tHM and its ALI is 2×10^{5} Bq/year. The separated fraction can either be stored on a specific adsorbent or discharged into the ocean. Since ^{129}I has a half-life of 16 million years it may very probably enter into a world-wide dispersion in the geosphere or biosphere. ^{129}I is because of its high radiotoxicity one of the critical nuclides when considering land-based repositories of spent fuel. In a world-wide dispersion scenario its radiotoxic importance is rather limited.

^{79}Se is a fission product with a half-life of 65 000 years which occurs in the HLW. Chemically this nuclide behaves as SeO_4^{2-} and will be incorporated in vitrified waste. Its radioactive concentration in spent fuel is expected to be around 2×10^{10} Bq/tHM, and its ALI is 10^7 Bq/year. Separation from liquid HLW is not obvious taking into account the very small chemical concentration in which it occurs, in comparison with natural sulphur compounds.

^{93}Zr and ^{135}Cs are two long-lived (1.5 and 2 million years half-life, respectively) nuclides occurring in spent fuel. Separation of these radionuclides from the other fission products for eventual transmutation is almost excluded since they are accompanied by other radioisotopes which are very radioactive (^{137}Cs) or are present in much larger quantities (0.73~1 kg ^{93}Zr with 3.3~5.0 kg Zr per tHM). In order to effectively reduce the radiotoxic potential by neutron irradiation, a series of isotopic separation processes ought to precede any target fabrication and this route is presently considered as an almost impossible endeavour from the economic point of view.

^{126}Sn has a half-life 100 000 years and is partly soluble in the HLLW and occurs partly in the insoluble residues. Its concentration in HLW ranges around 4×10^{10} Bq/tHM and its ALI limit of 3×10^6 Bq/year. The radioactive species ^{126}Sn is accompanied by a series of stable isotopes (^{116}Sn, ^{118}Sn, ^{119}Sn, ^{120}Sn, ^{122}Sn, ^{123}Sn and ^{124}Sn) which makes it difficult to consider its transmutation.

^{14}C, with a half-life of 5 730 years, is a difficult case because it can potentially enter into the biosphere through its solubility in groundwater and play an important radiotoxicological role because of its incorporation into the biochemical life cycle. According to the nitrogen contamination of the initial UO_2 fuel, its concentration in spent fuel is about 3×10^{10} Bq/tHM and its ALI limit 4×10^7 Bq/year. Its role in the long-term radiotoxicity is dependent on the physico-chemical conditions occurring in deep underground aquifers or in water unsaturated geospheres. The capture cross-section in a thermal neutron spectrum is very small

^{36}Cl: UO_2 and MOX fuel as well as zircaloy cladding contain about 20 ppm Cl. During irradiation this ^{35}Cl is transmuted into ^{36}Cl with a half-life of 300 000 years. This activation product arises partly in the dissolver liquid and partly remains within the washed Zircaloy hulls. At 45 GWd/tHM about 2×10^6 Bq/tHM are calculated to be globally present in the HLW and MLW. Due to its chemical characteristics this nuclide is easily dissolved in groundwater and could contaminate water bodies around a repository. The ALI by ingestion is 2×10^7 Bq/year. This radionuclide cannot be considered in a transmutation scenario since contamination of waste concentrates with natural ^{35}Cl would generate additional ^{36}Cl.

Some radionuclides discussed in this section ought to be examined in depth in order to establish their risk and potential radiotoxic role in comparison with the TRUs. Their radiotoxicity is between 1 000 and 100 000 times less important than TRUs but their contribution to the long-term risk is predominant because migration to the biosphere may be much more rapid and generate in the very long term a non-negligible radiation dose to man.

3.2.3.5 Quantitative recycling of U and Pu into LWR-MOX fuel

Plutonium separated during reprocessing of LWR-UO_2 fuel is in some countries transferred to the reactor as LWR-MOX. However, the irradiation of LWR-MOX increases the radiotoxic inventory of the fuel because of the increased production of MAs. Multiple recycling of plutonium in LWR-MOX is not efficient.

In the recent past, consideration was given to using the separated plutonium as a feedstock for FR-fuel where the "incineration" of even as well as uneven numbered isotopes resulted in an increased fission rate compared to that in LWRs. The reduction of the radiotoxic inventory of nuclear materials can only be accomplished by using transmutation in fast spectrum devices (FRs or ADS).

Recycling of reprocessed uranium in LWR-fuel is done industrially but entails an increase of the fissile material enrichment in the fresh fuel. From pure radiotoxic point of view, the stockpiling of depleted uranium and of reprocessed uranium has a greater impact than e.g. neptunium.

3.2.3.6 Reprocessing of LWR-MOX

Up to now the reprocessing of LWR-MOX has mainly been done by diluting the LWR-MOX fuel with LWR-UO_2 fuel according to the ratio in which it occurs in the reactor-core (UO_2/MOX = 2). Reprocessing of spent LWR-MOX without dilution in UO_2 fuel has been demonstrated at Cogéma La Hague UP2 plant in 1992 as a special campaign (~5 t) and can be performed industrially if the reprocessing plant has been licensed for the treatment of increased plutonium concentrations and a much higher total plutonium inventory.

The radiotoxic inventory of spent LWR-MOX fuel is about 8 times higher than that of spent LWR-UO_2. Conventional reprocessing will remove U+Pu which accounts for about 30% of the total α-activity and the residual 70% enters into the HLLW made up of Np, Am and Cm. However, the Cm and Am isotopes constitute the overwhelming majority of this α-activity.

In a perspective of P&T, it would be indispensable to remove the TRUs from the HLLW before vitrification. The techniques to be used are in principle the same as for the LWR-UO_2 fuel (see 3.2.3.2 Separation of MAs from HLLW resulting from spent LWR fuel), but the higher α-activity level will interfere with the extraction because of increased radiation damage.

Another option is to store the spent LWR-MOX fuel for example during 50 or more years and to let ^{244}Cm decay (18 year half-life) to ^{240}Pu before carrying out the reprocessing. The chemical extraction processes are much easier to perform after the extended "cooling" period as the α-decay heat is reduced by a factor of 7 or more, depending on the isotopic composition. Multiple recycling of LWR-MOX is possible if sufficient fresh plutonium from reprocessing of LWR-UO_2 with moderate burn-up is available.

3.2.3.7 FR-MOX fuel fabrication with limited MA content

The largest industrial experience has been gained in the FR-MOX fuel fabrication since for several decades FR programmes were undertaken in many nuclear countries [12]. The fabrication of FR-MOX fuel with 15 to 25% Pu has been realised routinely and on a commercial basis. But the plutonium quality used for these purposes was derived from low burn-up UO_2 fuel with low ^{238}Pu and ^{241}Pu contents.

In the meantime the burn-up of spent LWR-UO_2 and LWR-MOX has reached 50 GWd/tHM. The isotopic composition of plutonium resulting from the reprocessing of such fuels is seriously degraded, with high ^{238}Pu and ^{242}Pu levels, and low ^{239}Pu and ^{241}Pu concentrations. A thorough study of the economic issues involved was published in 1989 by OECD/NEA and is still valid as general reference [13].

In a perspective of the use of advanced FBuRs (CAPRA) still higher plutonium concentrations are envisaged (up to 45%). The recycling of fuels containing high ^{238}Pu levels and limited amounts of MAs is still more difficult and requires the design and construction of remotely operated fuel fabrication plants.

For homogeneous recycling of MAs in FR-MOX, admixtures of 2.5% ^{237}Np and/or ^{241}Am are currently studied. The specific activity of ^{237}Np, an α-emitter, is low and there is no major handling problem involved but the admixture of ^{241}Am at the 2.5% level will induce a γ-field around the glove-boxes or hot cells. However, the major interfering nuclide is ^{238}Pu at the 3% level which is a heat and neutron source (5 kWth/tHM; 5.10×10^8 neutrons/s tHM).

The FR-MOX fuel fabrication with limited MA admixture will also be influenced by the degree of separation of the REs (strong γ emitters) and last but not least by ^{244}Cm which will accompany ^{241}Am and ^{243}Am when separated from HLLW. The presence of small amounts of ^{244}Cm will increase the neutron emission of the resulting fresh FR-MOX fuel.

The separation coefficients from REs and ^{244}Cm required in order to permit industrial fuel fabrication operations will greatly depend on the permissible RE concentration acceptable in fresh FR-MOX fuel and on the permissible ^{244}Cm concentration during the fuel fabrication process.

Heterogeneous recycling of MAs is a means to avoid the dilution of troublesome nuclides, e.g. ^{244}Cm, throughout the fuel fabrication step and carry out this operation in small, but dedicated and well shielded facilities.

3.2.3.8 Metal fuel fabrication for ALMRs and advanced fuels for burner reactors

In the framework of the Integral Fast Reactor (IFR) project a specific fuel fabrication technology has been developed and tested on cold (and hot) pilot scale. At the EBR-II facility metal fuel was recycled by casting a U-Pu-Zr alloy on laboratory and hot pilot scale. It is obvious that these processes are still in the exploratory stage and cannot be considered as proven technology but their potential should be investigated since metal fuel permits very high burn-ups and has good material and neutronic characteristics for transmutation of TRUs.

Very recently, attention was drawn on the potential of nitride and carbide fuels for FBuRs. Nitride TRU fuel containing macroscopic quantities of MAs can be produced by a combination of an internal gelation method and a carbothermic synthesis. These nitride fuels can be reprocessed by electro-refining methods similar to the technology developed for metal fuel.

3.2.3.9 Fast burner reactors (FBuR)

A large technological experience has been accumulated during thirty years of R&D all over the world for the fast breeder reactor (LMFBR). This experience can be transferred to FBuR technology. Details for the fuel can be found in Reference 11.

In France, the CAPRA research programme was launched in 1992 by CEA on enhanced plutonium burning in FBuRs, obtained by increasing the relative concentration of plutonium in the fuel. The SPIN programme was launched in parallel on MA incineration. In order to reduce as much as possible the TRU formation, U-free TRU fuel with inert matrices is receiving increasing attention.

In 1994 the Super-Phénix reactor (SPX) had been re-licensed to be progressively converted from a plutonium breeder to a burner, following the recommendation of a governmental commission [12]. To that aim, steel reflector assemblies have been fabricated to replace the radial, fertile blanket. Three test assemblies have also been manufactured: two CAPRA ones, differing in the origin of their plutonium, either from first or second generation, and one NACRE assembly containing 2% Np added to the usual MOX, in the line of the SPIN programme. After a successful power operation in 1996, Superphénix prepared for these core substitutions. However, the new French government decided in June 1997 and confirmed in February 1998, that SPX should be definitively shut down.

This decision will lead to marked reorganisations of the fast reactor programme. Concerning the investigations on enhanced TRU burning, a partial redeployment of the experiments from Superphénix to Phénix is under discussion, taking into account the lower power of Phénix and the limitation of its availability to 2004, as allowed by the safety authorities. To cope with this new context the most effective experiments will be selected so as to fulfil most requirements of the December 1991 French law on nuclear waste research.

In 1988, the Japanese government launched new transmutation projects in the framework of the OMEGA programme. These projects are being further developed but apart from FR-MOX fuel other types, e.g. nitrides and carbo-nitrides, are being investigated as possible alternatives.

The IFR project of ANL evolved into an industrial sodium cooled FR prototype design called the PRISM reactor which is supposed to use metallic fuel (U-Pu-Zr) with adjacent pyrochemical reprocessing facilities to recycle TRU fuel and discard the fission products. Core design studies and core performance analysis are underway to increase the TRU incineration.

3.2.3.10 FR-spent fuel reprocessing

The reduction of the radiotoxic inventory resulting from multiple recycling of spent FR-MOX or FR-Metal fuel is only possible if repeated reprocessing of FR-spent fuel is part of the AFC scenario. Irradiation of FR-MOX or FR-Metal fuel is carried out to very high burn-up levels.

Compared to LWR-MOX fuel, the target burn-ups of FR-fuel are 2 to 3 times higher, i.e. burn-ups ranging from 120 to 180 GWd/tHM are currently to be envisaged and even higher burn-up levels may be attained in a more distant future. But even these very high burn-up levels still correspond to a rather small transmutation or incineration yield.

After a prolonged irradiation campaign of e.g. 5 years and a burn-up of 150 GWd/tHM, the degree of depletion is only about 15 to 20% of the initial TRU inventory. In order to reach an overall TRU depletion yield of 90%, it is necessary to recycle the FR-spent fuel many times.

Any reprocessing campaign of spent FR-fuel based on the use of the PUREX process or on an equivalent aqueous extraction process cannot be conveniently carried out within a short-time interval after the discharge from the fast reactor because of the high decay heat. Cooling times between discharge and reprocessing are, as a result, increased from 7 to 10 or 12 years. The duration of the total AFC cycle becomes 15 to 17 years per cycle. The time interval to reach a global depletion of 90% is consequently ranging from 100 up to 250 years.

Pyrochemical reprocessing of spent FR-fuel was developed in the frame of the IFR project. In this case FR-Metal fuel irradiated at very high burn-up is transferred to a "on-site" pyrochemical hot-cell for reprocessing. The molten salt bath ($CdCl_2$, LiCl, KCl …) is much less affected by α-radiation damage and much shorter cooling times can be envisaged. However, this type of reprocessing technology is still in its infancy and will require extensive R&D before reaching industrial maturity in the civil nuclear sector.

The process involves electro-refining of spent metal fuel with a Cadmium anode, solid and liquid cathodes, and a molten salt electrolyte (LiCl+KCl) at 500°C. The heart of the process is a pyrochemical dissolution process of mechanically decladded spent fuel.

Pure uranium (free of plutonium) is electro-transported to the solid Cd cathode. The mixture of Pu+MAs with some uranium is electro-transported to a second but liquid Cd cathode and separated from the bulk of fission products. The Pu+MA fraction is recycled as metal and cast into the new fuel pins.

Similar pyrochemical processes are being developed in Japan by the CRIEPI and may, in a distant future, become an alternative to aqueous reprocessing with the PUREX process for highly irradiated FR-Metal/Nitride spent fuel.

The fundamental advantage of the pyrometalurgical reprocessing processes is their insensitivity to burn-up of the spent fuel and to criticality excursions during the operations. The possibility to reduce a single AFC fuel cycle to six or seven years reduces the time interval to get a given depletion yield in half the time period scheduled for an AFC with aqueous reprocessing. However, the material problems due to corrosion, the degree of separation of the TRUs and the waste handling are the most important issues which await fully convincing answers before this type of pyrochemical process will become a serious contender for the reprocessing of FR-Metal fuel.

3.2.3.11 Transmutation issues of long-lived fission products

Transmutation of long-lived FPs is a very difficult task because the capture cross-sections to transmute the radioactive nuclides into short-lived or stable nuclides are very small. In this way, very long irradiation periods are necessary to obtain a significant depletion. Dedicated reactors with large thermal neutron fluxes and/or dedicated accelerator-driven transmutation facilities are the only possible choices for carrying out this very expensive endeavour.

From reactor technology point of view, the transmutation of ^{99}Tc is the easiest approach since the metallic Tc target is transmuted into metallic ^{100}Ru, a stable ruthenium isotope. However, the thermal n-γ cross-section for thermal neutrons is only 20 barns (see 3.2.3.2) which is low for practical transmutation purposes. The transmutation "half-life", i.e. the time necessary to deplete the target with 50% varies from one thermal reactor to another but amounts to about 30 years in the best conditions.

The simultaneous production of ^{99}Tc by fission in the LWR-UO_2 driver fuel decreases the net transmutation rate and necessitates large Tc loadings. The reactor ought to be dedicated for that purpose. A dedicated reactor must be over-enriched to compensate the reactivity loss due to the negative reactivity of the Tc targets in the fuel assemblies. These long "half-life" times lead to the need for reprocessing of assemblies with irradiated LLFP, because of the irradiation damage (dpa) to the steel structures of the fuel assemblies.

Transmutation of ^{129}I is from pure neutronic point of view very similar to that of ^{99}Tc, but is from chemical-metallurgical standpoint a much more difficult process since the target product is chemically unstable and the neutron capture reaction product is a noble gas: ^{130}Xe which has to be vented from the irradiation capsule during its stay in the reactor. Any temperature excursion would result in a release of ^{129}I into the reactor off-gases.

ADS technologies with extreme high thermal neutron fluxes (10^{16} n/cm^2/s) should, in principle, be capable of reducing the transmutation half-life. This technology is presently in the conceptual phase. However, any type of thermal neutron transmutation will be energetically very expensive.

The radionuclides ^{93}Zr and ^{135}Cs cannot be considered for reactor transmutation unless they are separated from the other Zr and Cs isotopes before being submitted to irradiation.

The transmutation of ^{14}C has not yet been considered in the P&T context. Theoretically, the ^{14}C released from the spent fuel could partly (about 50%) be recovered from the reprocessing off-gases. There is, however, not enough knowledge about the chemistry of ^{14}C in dissolver conditions to improve this figure. Once transformed into a solid target e.g. barium carbonate ($BaCO_3$), it could be stored for an infinite period. The cross-section of ^{14}C for thermal neutrons is nearly zero. Transmutation by charged particles in high energy accelerators is a theoretical alternative in some cases but the practical feasibility and the economy of such approaches are very questionable.

3.2.3.12 Conclusions on the role of P&T in the AFC option

The P&T option within the AFC scenario, as described above, is the most comprehensive approach which can be reasonably proposed and constitutes a very important extension of the fuel cycle activities in comparison with the RFC and a fortiori with OTC scenario.

Partitioning operations could be envisaged as a series of stand alone operations, following conventional reprocessing, in order to decrease the radiotoxicity of the disposed waste and to allow improved conditioning techniques. However, within the context of this report, partitioning has mainly been described as a preparatory step to transmutation.

4. CRITICAL EVALUATION

- P&T may take its place in a future nuclear fuel cycle industry either as an additional activity or as a partial replacement of present fuel cycle activities.

- The P&T option can be chosen only if a conventional or advanced fuel cycle is operated. But during the R&D phase, all countries interested in a development of the fuel cycle or waste management can participate in an international effort.

- The P&T strategy should be oriented towards the gradual elimination of long-lived radionuclides from waste streams for which decisions in the field of waste disposal should be taken. But this approach will lead to a reduction of the radiotoxic inventory in the waste at the expense of a limited increase of radiotoxic inventory in the reactor cores and in the fuel cycle facilities unless a comprehensive actinide incineration route such as accelerator-driven transmutation is set up.

- In the course of a long-term nuclear programme, there might be a gradual shift from LWR-UO_2 fuel to LWR-MOX fuel to be followed by the introduction of FRs in the NPP park and by the use of accelerator-driven systems.

- Partitioning of minor actinides is an additional step to U and Pu recycling in MOX fuel. In order to carry out partitioning operations, a number of additional industrial facilities will have to be designed and constructed. But the present state of the art in partitioning is at the laboratory and hot-cell scale of development.

- Partitioning is a long-term venture which needs newly designed facilities for treatment of HLLW or an adapted PUREX extraction cycle with direct separation of all long-lived radionuclides.

- Partitioning of MAs from HLLW reduces the long-term radiotoxic inventory of vitrified HLW and contributes to improving the hazard perception of the HLW to be disposed of. However, it does not modify the technical aspects of waste. The fission products (^{137}Cs and ^{90}Sr) determine the heat output of HLW, while the leach rates of the TRUs, which determine the radiological impact in the long term, are controlled by their low solubilities.

- Partitioning of MAs (Np, Am, Cm) and selected fission product (Cs, Sr, Tc, etc.) creates more opportunities to improve the nuclide-specific conditioning of long-lived radionuclides. The increased thermodynamic stability of new waste forms offers, in principle, better perspectives for very long-term storage or disposal of TRUs in comparison with vitrification which has been designed especially for complex mixture of MAs and fission products. Further development of partitioning methods and technology is strongly recommended in order to broaden the technical basis for engineering assessment of partitioning.

- Transmutation is a general term which covers incineration, i.e. transformation into fission products, and transformation by neutron capture into another radionuclide or a stable isotope. The development of this technology may have important spin-offs for other nuclear programmes.

- Some transmutation reactions produce a majority of non-radioactive nuclides, some generate a variety of radionuclides with a range of half-lives. The produced nuclides should have a shorter half-life than the target, or a lower radiotoxicity, or lead in some cases to the production of another mother nuclides with less radiological impact.

- Transmutation of MAs is, in the short term, not always conducive to the reduction of short term radiotoxic inventory.

- Transmutation in thermal reactors proceeds in general by neutron capture. Recycling of industrial quantities of MAs requires an increased enrichment level with an accompanying increase in the nuclear power production cost. Yet, heterogeneous recycling of MAs in LWR-MOX fuelled reactors is possibly a viable route as long as surplus Pu is available to compensate for the negative reactivity induced by the MAs. However, on this transmutation route, the overall radiotoxicity of the fissile and fertile materials of the fuel cycle does not decrease significantly.

- Transmutation of MAs in fast neutron reactors reduces the radiotoxicity since all MAs are fissionable to a certain degree. The higher the mean neutron energy the higher the "incineration" yield. But for reasons of reactor safety the acceptable MA loading and the resulting fission and transmutation yields are limited. The theoretical energy potential of the MAs is about 10% of that of plutonium generated in UO_2 fuel. The fast neutron spectrum devices (FR, ADS) are more promising than LWRs for the transmutation of MAs.

- Some long-lived fission products have been considered as candidates for partitioning and transmutation. ^{129}I, ^{135}Cs, ^{99}Tc and ^{93}Zr are the most frequently mentioned.

- Partitioning of some FPs is already achieved industrially: iodine is removed from spent fuel solutions by sparging, scrubbing and separate treatment before disposal into the oceans (in some countries the discharge of iodine is not allowed). If the discharge of ^{129}I to the ocean should become difficult to continue over a long period of time, recovery, conditioning and perhaps transmutation ought to be re-examined. The transmutation of ^{135}Cs and ^{93}Zr is impossible without isotopic separation.

- The theoretical basis for Tc separation has been described but no practical implementation has been started. Transmutation of ^{99}Tc is possible in thermal reactors and in thermalized sections of fast reactors. However, the transmutation half-life in a thermal neutron spectrum device is fairly long (15 to 77 years). Unless neutrons will become available at marginal cost in future nuclear facilities (ADS) this option should not get first priority in R&D.

5. GENERAL CONCLUSIONS

- Fundamental R&D for the implementation of P&T needs long lead times and requires large investments in dedicated fast neutron spectrum devices (FRs, ADS which may complement or substitute for operating LWRs), extension of reprocessing plants and construction of remotely manipulated fuel and target fabrication plants.

- Partitioning facilities for MAs and some long-lived fission products could be designed and constructed as extensions to the existing reprocessing plants.

- Partitioning methods for long-lived radiotoxic elements have been developed on a laboratory scale but much work is still to be done on improving and scaling up the partitioning technology in order to make it compatible with industrial reprocessing practices.

- The short-term impact of partitioning would be to reduce long-term radiotoxic inventory of the resulting HLW at the expense of an increase of the operational requirements for the nuclear facilities concerned.

- Fast neutron-spectrum devices (dedicated FR or ADS facilities) are more efficient than current LWRs for recycling and transmuting long-lived radionuclides. The ADS might play an important role in "incinerating" the actinides and some FPs from the spent LWR-MOX or FR-MOX fuel in order to reduce the long-term radiotoxic inventory of the wastes.

- Recycling of Pu+MAs could stabilise the TRU inventory of a NPP park. Multiple recycling of TRUs is a long-term venture which may take decades to reach equilibrium TRU inventories.

- Conditioning of separated long-lived nuclides in appropriate matrices which are much less soluble than glass in geological media, or which could serve as irradiation matrix in a delayed transmutation option, is a possible outcome for the next decades.

- P&T will not replace the need for appropriate geological disposal of HLW, irradiated TRU concentrates and residual spent fuel loads from a composite reactor park.

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NOTE DE SYNTHÈSE ET PORTÉE DU RAPPORT

La séparation et la transmutation (P&T) sont des techniques destinées à réduire les inventaires d'actinides et de produits de fission à vie longue dans les déchets nucléaires. Si la mise en œuvre de ces technologies peut atténuer les dangers que présentent la gestion des déchets avant leur stockage et les perturbations physiques d'un dépôt de déchets, par exemple par intrusion humaine, elle n'est pas considérée par tous comme la seule solution susceptible de remplacer à court terme les stratégies actuellement en place pour l'aval du cycle du combustible, ni comme un moyen d'éviter la construction de dépôts en formations géologiques profondes pour y stocker les déchets à vie longue que l'on produit aujourd'hui.

Néanmoins, il est largement admis que les technologies permettant de réduire ces inventaires peuvent régler certains problèmes de stockage des déchets dans des cycles du combustible conçus spécifiquement pour les mettre en œuvre. Les études qui seront entreprises à ce sujet serviront au minimum à faire progresser la science nucléaire et à attirer de jeunes chercheurs compétents vers cette discipline en prévision d'un basculement de l'opinion publique mondiale en faveur de l'énergie nucléaire. Par ailleurs, les études consacrées à la technologie des lasers et des accélérateurs, par exemple, auront certainement des retombées sur d'autres domaines de la science et de la technologie.

La plupart des recherches sur ce terrain viennent tout juste de voir le jour, et l'on aurait beaucoup de mal à prévoir les résultats des travaux actuels de recherche et de développement, à fixer des échéances, ou encore à donner une indication des coûts du développement et de l'exploitation des technologies étudiées. Bien que le présent rapport contienne une présentation de la structure d'une étude systémique globale, avec les informations nécessaires pour la réaliser, la priorité est actuellement donnée à l'examen des technologies de séparation et de transmutation utilisables et à l'analyse de leur état d'avancement et des perspectives d'applications industrielles éventuelles.

Le but du rapport est de proposer une analyse faisant autorité des conséquences techniques, radiologiques et, dans la mesure du possible, économiques de la séparation et de la transmutation sur les options actuelles et futures pour le cycle du combustible. Le rapport comporte donc une partie générale destinée au non-spécialiste et une analyse systémique technique de tous les aspects de la séparation, de la transmutation et de la gestion à long terme des déchets.

Première partie : Présentation générale

- Présentation du programme international coordonné par l'AEN.
- Survol de l'état actuel et des perspectives des technologies de séparation et de transmutation et analyse comparée des problèmes posés par les différents cycles du combustible.

- Analyse critique afin de dégager les points forts et les faiblesses dans la stratégie de séparation et de transmutation.

- Conclusions générales et recommandations relatives aux priorités futures.

Deuxième partie : Analyse technique et étude systémique

- Analyse des perspectives des différentes méthodes de séparation envisagées soit comme des étapes supplémentaires au procédé PUREX soit comme des procédés autonomes de traitement pyrochimique du combustible.

- Analyse technique approfondie des solutions possibles pour transmuter les actinides mineurs et certains produits de fission dans des réacteurs à eau ordinaire (REO), des réacteurs rapides (RNR) et des systèmes de transmutation hybrides (réacteurs sous-critiques pilotés par des accélérateurs) comportant :
  - une étude de la faisabilité de la technologie et de la sûreté de la fabrication des combustibles et des cibles à base d'actinides mineurs ;
  - une étude approfondie des différents aspects de l'irradiation des combustibles et des cibles contenant des actinides mineurs dans les réacteurs à eau sous pression actuels (recyclage homogène et hétérogène) ;
  - un panorama complet des possibilités de transmutation des actinides mineurs dans des réacteurs rapides, sous forme de combustibles ou de cibles à base d'oxydes mixtes (MOX), de métaux et de matrices inertes ;
  - une première analyse des possibilités des systèmes de transmutation associant un réacteur sous-critique à un accélérateur, des problèmes que cette solution soulève et des projets actuels ;
  - une analyse comparée des possibilités de transmuter certains produits de fission à vie longue dans différents types de réacteurs et de systèmes hybrides.

- Description des principaux programmes de recherche et de développement entrepris au Japon et en France.

- Analyse approfondie des répercussions de la séparation et de la transmutation sur l'évaluation des risques et la gestion des déchets, et notamment de l'impact à long terme des méthodes proposées actuellement.

- Analyse des différents inventaires des éléments transuraniens en fonction des choix stratégiques de filières (réacteurs à eau ordinaire, réacteurs rapides, systèmes hybrides, etc.) qui seront opérés pour le futur réseau de production électronucléaire.

- Survol des concepts actuels de stockage et des répercussions éventuelles de la séparation et de la transmutation sur la structure et les caractéristiques des dépôts.

- Analyse de l'impact radiologique et radiotoxique à long terme des opérations de séparation et de transmutation.

Présentation générale

Cette partie contient une présentation générale de la situation actuelle du cycle du combustible et des pratiques de gestion des matières nucléaires. Un chapitre d'introduction est consacré aux nouvelles perspectives que pourraient offrir la séparation et la transmutation et à leurs limites. À titre d'entrée en matière pour les non-spécialistes, on y a décrit les opérations supplémentaires de recyclage des matières fissiles et fertiles (uranium et plutonium), des actinides mineurs (neptunium, américium, curium) et de certains produits de fission (technétium 99, césium 135, iode 129, etc.).

Suit une analyse détaillée de trois stratégies du cycle du combustible pour un parc électronucléaire de 100 GWe, par exemple. Il s'agit du cycle ouvert (CCO), du cycle fermé (CFR) avec retraitement de l'uranium et du plutonium et du cycle fermé et avancé (CCA) avec séparation et transmutation. Les questions dont l'issue dépend de décisions visant à réduire au minimum la radiotoxicité des déchets de la production électronucléaire sont développées.

Plusieurs tableaux récapitulatifs présentent des données de référence sur les combustibles UO_2 et à oxydes mixtes irradiés dans un éventail de conditions standard. L'annexe E de ce document contient des tableaux complets de données nucléaires correspondant à l'inventaire en radionucléides du combustible usé. Les données qui y figurent sont des valeurs moyennes tirées de codes de calcul et considérées par la communauté nucléaire comme des valeurs de référence pour le calcul de l'inventaire radiotoxique sur des périodes de temps prolongées.

Analyse critique et conclusions générales

La partie générale du rapport se termine sur une série de remarques critiques et de conclusions générales rédigées par le groupe de travail. Il s'agit essentiellement de mettre en évidence les points importants pour aider les décideurs à définir les priorités des futurs travaux de recherche et de développement, de souligner l'intérêt de la séparation et de la transmutation pour la réduction de déchets et de dessiner les contours de cette stratégie dans une politique nucléaire future.

Analyse technique et étude systémique

Séparation

Le retraitement du combustible usé des réacteurs à eau ordinaire est considéré comme un scénario de référence dans la première étape du cycle du combustible avancé, et est préalable aux séparations complémentaires des actinides mineurs et de certains produits de fission.

On commence par décrire brièvement des *techniques de séparation en milieu aqueux* compatibles avec le procédé bien connu PUREX en les illustrant par des schémas représentant les flux annuels. Suivent une courte présentation des améliorations possibles et souhaitables de l'étape d'extraction des éléments transuraniens dans le procédé PUREX afin de récupérer le neptunium 237 et une description, assortie de commentaires sur leurs avantages et limites, des procédés d'extraction des actinides mineurs « TRUEX », « DIDPA », « TRPO », « DIAMEX », « TPTZ », « SESAME » et des procédés les plus récents « CYANEX 301™ » de récupération de l'américium et du curium. L'impact de ces améliorations sur les procédés classiques de traitement de déchets, notamment sur la production de déchets secondaires, fait l'objet d'une courte analyse. L'annexe D contient une bibliographie complète sur la technologie de séparation.

Comme les fractions soluble et insoluble se dissocient dans le procédé PUREX, il faudra prévoir des opérations de tête supplémentaires pour pouvoir séparer certains produits de fission. La récupération de l'^{129}I et du ^{14}C dans les effluents gazeux du dissolveur est l'un des moyens de réduire l'impact du retraitement sur l'environnement. De même, en séparant deux autres radionucléides à vie longue, à savoir le ^{99}Tc et le ^{93}Zr, des flux de phosphate tributylique (TBP), on pourrait limiter les répercussions environnementales des déchets.

Au cours des vingt dernières années, l'extraction sélective des actinides mineurs et des produits de fission a fait des progrès considérables. Toutefois, comme on pourra le voir sur le tableau récapitulatif, les différentes méthodes utilisées n'ont pas atteint le même stade de maturité technique.

Plusieurs scénarios peuvent être envisagés pour la gestion des actinides mineurs séparés:

- Le neptunium issu directement du procédé de retraitement ou extrait des déchets de haute activité pourrait facilement être séparé et recyclé avec le combustible MOX ou encore conservé sous forme séparée le temps que les procédés avancés de transmutation-incinération (RNR, systèmes hybrides) aient atteint le stade industriel. Le neptunium ainsi séparé pourrait aussi être conditionné sous forme d'une phase thermodynamiquement stable en vue d'un stockage réversible à long terme. Dans ce cas, la matrice serait conçue pour servir de matrice d'irradiation ou de colis de déchets pour le stockage définitif.
- Que l'on prévoie une stratégie de transmutation ou un stockage, la fraction américium-curium séparée des déchets de haute activité pourrait être traitée comme une fraction unique. Ces deux éléments ont un comportement chimique si proche qu'il faut, pour les dissocier, recourir à des techniques radiochimiques et/ou électrochimiques très spécifiques. Au cas où l'on envisagerait de les garder ensemble, il serait possible soit de les stocker en ensemble soit de les conditionner dans une matrice de type céramique avant de les stocker dans des structures de stockage, le temps que les techniques de transmutation deviennent opérationels.

L'américium pourrait un jour être séparé seul et soit recyclé immédiatement dans des combustibles spéciaux REO-MOX ou RNR, où il serait transmuté en partie, soit conditionné sous une forme céramique avant de subir une transmutation-incinération prolongée par multirecyclage et passage dans des systèmes d'irradiation hybrides.

Jusqu'à présent, on n'a pas trouvé de solution convaincante pour la gestion du curium séparé, non seulement parce qu'il est difficile de le séparer de l'américium, mais aussi parce que ce nucléide, très radioactif (source de chaleur et de neutrons), ne peut pas être recyclé dans les combustibles REO-MOX ou RNR-MOX. Stocker ce nucléide séparé pendant un siècle atténuerait les problèmes de traitement et autoriserait son recyclage dans le cycle du combustible avancé.

Si l'on cherche à augmenter les taux de combustion et à raccourcir les temps de refroidissement, il devient intéressant de développer *les procédés non aqueux de séparation pyrochimique*. On a testé en laboratoire et en pilotes chauds des méthodes pyrochimiques utilisant des sels de chlorure fondus (LiCl-KCl ou NaCl-KCl) pour séparer l'uranium, le plutonium et les actinides mineurs des produits de fission. À l'avenir, on pourrait recourir à une solution alliant séparation électrolytique, précipitation chimique et extraction en milieu réducteur, soit comme étape préalable à la séparation par retraitement en milieu aqueux ou, éventuellement, comme procédé autonome de retraitement à sec du combustible irradié à des taux élevés, après des temps de refroidissement plus courts. L’application de cette technologie permettrait de raccourcir le délai avant son recyclage quantitatif.

Transmutation

Le chapitre consacré à la transmutation décrit plusieurs activités : la fabrication des combustibles et des cibles, la transmutation par capture neutronique et l’incinération par fission.

La transmutation peut s’effectuer dans des réacteurs thermiques, des réacteurs à neutrons rapides et des réacteurs sous-critiques pilotés par des accélérateurs. Le choix de ces installations de transmutation découle d’études stratégiques destinées à trouver la combinaison optimale de facteurs tels que la fabrication et la refabrication du combustible, sous tous leurs aspects, la disponibilité des différents types de réacteurs, les rendements de transmutation/incinération et surtout la réduction significative de l’inventaire radiotoxique global des déchets nucléaires. Les pays de l’Union européenne et le Japon ont réalisé des études approfondies afin de dégager une première approche stratégique.

Toutefois, ces études sont subordonnées à des décisions politiques et des réalités économiques que l’on ne saurait prévoir sur les dizaines ou même centaines d’années nécessaires pour parvenir à un équilibre entre la consommation et la production de matières nucléaires fissiles et fertiles.

Fabrication des combustibles et des cibles

On trouvera dans le rapport une description concrète des problèmes que pose l’introduction des actinides mineurs dans le cycle du combustible nucléaire. La fabrication du combustible à oxydes mixtes à faible teneur en neptunium et américium (< 2.5 pour cent) est une solution qui n’exige pas d’investissements lourds dans les usines de fabrication d’oxydes mixtes pour les réacteurs à eau ordinaire (REO-MOX). Cependant, l’introduction de ces actinides mineurs rend plus difficile le retraitement et la refabrication ultérieure du combustible en raison de la présence de grandes quantités de ^{238}Pu dans le flux de plutonium recyclé.

Il serait nettement préférable de recourir au recyclage hétérogène des actinides mineurs dans des réacteurs à eau ordinaire, dans la mesure où cette solution permet de gérer les cibles indépendamment du combustible REO-MOX et de les recycler dans des installations de petite taille dédiées au traitement des cibles irradiées. Le principal problème vient du recyclage du ^{244}Cm, qui est une forte source de chaleur et de neutrons et ne peut pas être traité dans des usines de fabrication du combustible standard. Aucune solution satisfaisante n’a encore été trouvée. Par conséquent, il convient de poursuivre les recherches sur la séparation afin de mettre au point une technologie viable.

On produit déjà depuis plusieurs dizaines d’années du combustible pour les réacteurs rapides, et on a fabriqué des combustibles expérimentaux à forte teneur en américium et en neptunium dans le cadre du programme « SUPERFACT ». Pour l’irradiation des actinides mineurs dans des flux neutroniques rapides, il faudrait privilégier les matrices inertes (oxyde d’aluminium, oxyde de magnésium, etc.).

De nouvelles formes de combustibles adaptés à des taux de combustion et des rendements de transmutation supérieurs sont à l’étude, notamment des combustibles à base d’alliages métalliques (d’actinides mineurs, d’uranium, de plutonium et de zirconium) et de nitrures. L’intérêt des combustibles métalliques tient à la possibilité d’établir un cycle du combustible « compact » fondé sur les techniques pyrochimiques, mêmes si d'importants travaux de recherche et de développement seront nécessaires avant sa réalisation.

La faisabilité du combustible à base de nitrure dépend de la disponibilité d'usines d'enrichissement en ^{15}N et du développement des techniques de récupération de l'^{15}N au cours des procédés de recyclage. Le recyclage de cet élément serait également facilité si l'on utilisait des procédés pyrochimiques pour traiter les combustibles nitrures.

Transmutation des actinides mineurs

La physique de la transmutation/incinération des actinides mineurs montre que les neutrons rapides, dans des réacteurs rapides ou dans des systèmes hybrides, sont mieux adaptés que les neutrons thermiques en raison du rapport entre les sections efficaces de fission et de capture. En outre, tous les actinides mineurs sont, dans une certaine mesure, fissiles dans un spectre neutronique rapide et, de plus, contribuent à la production d'énergie globale pendant l'irradiation.

On trouvera dans ce rapport des tableaux comparatifs indiquant les rendements calculés de la transmutation/incinération d'actinides mineurs recyclés en mode homogène ou hétérogène dans des réacteurs à eau ordinaire brûlant des oxydes mixtes ainsi que dans des réacteurs incinérateurs rapides et des réacteurs avancés à métal liquide. La plupart des concepts de réacteurs avancés n'ont pas dépassé le stade de la recherche théorique, de sorte que les rendements calculés doivent encore être vérifiés dans des conditions représentatives.

La transmutation des actinides mineurs dans des cœurs de réacteurs rapides (réacteurs surgénérateurs rapides à métal liquide, réacteurs à métal liquide avancé) ne présente pas d'inconvénient majeur en termes de performances du cœur, à condition que le rapport entre les actinides mineurs et le combustibles reste petit.

La conception des réacteurs-incinérateurs rapides et des réacteurs-incinérateurs d'actinides mineurs nécessite encore des travaux de recherche et de développement, car il n'est pas évident que des concentrations élevées en actinides mineurs puissent être chargés dans le cœur de ces réacteurs parce qu'il engendrent des effets négatifs sur les coefficients de réactivité et les paramètres cinétiques.

Ces derniers temps, la communauté internationale s'est intéressée de près aux systèmes hybrides combinant un réacteur sous-critique à un accélérateur pour remplacer les réacteurs nucléaires de conception classique. Les accélérateurs, qu'il s'agisse d'accélérateurs linéaires ou de cyclotrons, produisent un flux de protons qui, s'il entre en collision avec une cible en métal lourd, provoque la spallation de la cible. Les neutrons de spallation percutent à leur tour les matières nucléaires fissiles et fertiles, par exemple des actinides mineurs, qui subissent des réactions de fission ou de capture.

La transmutation des actinides mineurs ne peut pas être dissociée de celle du plutonium, et toute stratégie l'intégrant doit prendre en compte la consommation des actinides mineurs dans les cibles ou les combustibles et la production simultanée d'actinides mineurs au départ du plutonium.

Transmutation des produits de fission à vie longue

Les études réalisées sur la transmutation des produits de fission à vie longue (^{99}Tc, ^{129}I) en nucléides à vie courte ou en nucléides stables ont montré que cette approche est théoriquement réalisable, à condition d'augmenter le taux d'enrichissement du combustible si l'on envisage d'utiliser des réacteurs à eau ordinaire. La période de transmutation varie de 15 à 18 ans, dans les réacteurs rapides, à 40-77 ans, dans des réacteurs à eau ordinaire. Irradier certains produits de fission dans des systèmes hybrides ou dans des réacteurs rapides optimisés permettrait d'écourter la période de transmutation. Il faudrait, dans ce cas, entreprendre des travaux très importants afin de mettre au point les matériaux des assemblages combustibles et des cibles (comme dans le cas de l'iode) et, pour certains éléments, procéder à une séparation isotopique très fine (par exemple, Se , Zr, Cs, Sn …).

Tendances actuelles de la recherche sur la séparation et la transmutation

Étant donné l'état d'avancement des programmes de recherche et de développement au Japon et en France, une section spéciale ainsi que les annexes B et C sont consacrées aux travaux, en cours dans ces pays, sur la séparation et la transmutation. Ces programmes, auxquels participent directement ou indirectement de nombreux pays, sont incontestablement les plus importants dans le monde.

Conséquences de la séparation et de la transmutation sur les évaluations des risques et la gestion des déchets

Afin d'exposer de la façon la plus claire possible les conséquences que peuvent avoir la séparation et la transmutation sur la radiotoxicité à long terme, nous avons souhaité définir ce qu'est l'inventaire radiotoxique et en montrer la signification réelle dans une section spéciale de ce rapport.

Cet inventaire radiotoxique dépend de l'inventaire physique des radionucléides dans les divers cycles du combustible ainsi que des coefficients de dose efficace de chaque radionucléide. Des schémas illustrent l'évolution naturelle des inventaires radiotoxiques dans le cycle ouvert, le cycle fermé avec retraitement et le cycle fermé avancé.

La place qu'occuperont la séparation et la transmutation dans la gestion des déchets dépendra essentiellement des inventaires d'actinides mineurs à traiter dans les installations du cycle du combustible avancé. On évoque dans cette section la stratégie d'utilisation des réacteurs qu'il faudra mettre en œuvre pour parvenir à une réduction significative de l'inventaire radiotoxique. Trois cas représentatifs seront considérés qui correspondent à diverses combinaisons de réacteurs à eau ordinaire chargés en combustible au dioxyde d'uranium ou en combustible à oxydes mixtes et de réacteurs rapides brûlant du combustible à oxydes mixtes. Il apparaît qu'on aura besoin d'une proportion importante de réacteurs rapides. Dans chacun des cas examinés, l'inventaire radiotoxique des flux de déchets diminue d'un facteur proche de 10. Cependant, cette réduction de la radiotoxicité des déchets s'accompagne d'une progression régulière de l'inventaire radiotoxique des réacteurs et des installations du cycle du combustible et exige, de surcroît, que l'on fasse fonctionner ces installations durant une longue période.

Si l'on recycle tous les actinides mineurs, y compris la fraction curium, le facteur de réduction de l'inventaire radiotoxique, par rapport au cycle ouvert, se situe entre 77 et 100 après 10 000 ans et, à très long terme (10^5 et 10^6 années), entre 80 et 150.

Si l'on ne recycle pas la fraction curium, cette réduction de l'inventaire radiotoxique n'atteint que 7 à 10 au bout de 10 000 ans en raison de la décroissance du ^{244}Cu en ^{240}Pu et du ^{243}Cu en ^{239}Pu.

Le bilan massique global des actinides mineurs produits dans un programme de recyclages multiples à long terme révèle que l'inventaire radiotoxique des matières nucléaires accumulées dans les réacteurs et les installations du cycle du combustible atteint des valeurs très élevées par rapport aux déchets rejetés chaque année. Dans divers cas étudiés, les inventaires à l'équilibre représentent plusieurs centaines de tonnes d'éléments transuraniens.

Concepts de gestion des déchets

Toute estimation des avantages radiologiques de l'option séparation/transmutation doit tenir compte de la mobilité des éléments dans la géosphère. Ce facteur peut en effet considérablement modifier le tableau assez simple que donnerait une étude limitée à la réduction de l'inventaire radiotoxique. D'un autre côté, la perspective de trouver des conditionnements et des sites particulièrement sûrs pour les radionucléides à vie longue présente d'autres avantages, notamment en ce qui concerne l'effet d'une intrusion humaine.

Pour replacer la séparation et la transmutation dans le contexte des stratégies actuelles de gestion des déchets, nous décrivons brièvement les concepts de stockage direct en formation granitique adoptés par la Suède et l'Espagne. Une section est consacrée à la sûreté en matière de criticité du stockage du combustible usé.

Pour le cycle du combustible avec retraitement, nous analyserons, à titre d'illustration, l'impact du stockage des déchets de haute et moyenne activité tel que l'ont prévu l'Allemagne, la Suisse et la Belgique dans des formations salifères, granitiques et argileuses. Plusieurs options sont actuellement à l'étude pour le stockage des colis de déchets produits par le cycle du combustible avancé avec séparation et transmutation. Néanmoins, le rapport apporte quelques précisions sur les aspects techniques et opérationnels de cette stratégie, notamment en ce qui concerne l'entreposage et le devenir des cibles irradiées et des cœurs de réacteurs à l'issue d'une campagne prolongée de production d'énergie.

Outre les matières hautement radioactives produites par le cycle du combustible avancé, il convient de comptabiliser les quantités importantes d'uranium appauvri issues du procédé d'enrichissement. À court terme, ces stocks d'uranium appauvri peuvent être considérés comme une matière stratégique que l'on réutilisera si les RNR contribuent un jour, dans des proportions significatives, à la production d'électricité.

En revanche, si les RNR ne font pas une percée dans la production d'électricité, cet uranium appauvri deviendra un déchet dont la radiotoxicité atteindra finalement celle du minerai d'uranium.

PREMIÈRE PARTIE : PRÉSENTATION GÉNÉRALE

1. INTRODUCTION

1.1 Activités de l’Agence de l’OCDE pour l’énergie nucléaire

La décision du gouvernement japonais, en 1988, de lancer un programme de recherche et développement à long terme sur le recyclage et la transmutation des actinides et des produits de fission à vie longue a avivé l’intérêt de certains pays Membres de l’OCDE pour cette technologie. Le programme japonais, du nom d’OMEGA (Options for Making Extra Gains from Actinides and fission products), a été entrepris à l’initiative de l’Atomic Energy Bureau de la Japanese Science and Technology Agency (STA) afin d’améliorer l’efficacité du stockage des déchets de haute activité et d’exploiter au mieux les ressources du combustible usé. Ce programme était conçu à la fois comme une recherche fondamentale à long terme dont bénéficieraient les générations futures et comme un moyen d’inciter les jeunes chercheurs à mettre leurs compétences au service de l’étude des aspects scientifiques de la gestion des déchets nucléaires.

Le gouvernement japonais a invité l’OCDE-AEN à organiser un projet international consacré à la séparation et à l’utilisation des actinides. Après quelques discussions, il a été décidé de lancer un programme d’échange d’informations techniques qui constituerait le pivot des recherches multilatérales réalisées par les pays de l’OCDE-AEN dans le domaine de la séparation et de la transmutation. Dès le lancement de cette activité, il était clair que ce programme n’aurait aucune répercussion sur les politiques nationales arretées pour l’aval du cycle du combustible avec ou sans retraitement, vitrification des déchets de haute activité et stockage des déchets ou du combustible usé dans des formations géologiques.

La première réunion internationale d’échange d’informations a été organisée à Mito (Japon) en novembre 1990. Elle a été l’occasion de prendre connaissance de communications très stimulantes consacrées tant aux orientations des politiques dans ce domaine qu’aux aspects scientifiques. Les discussions ont révélé une grande disparité dans les approches adoptées et qui recouvraient une diversité de procédés chimiques aqueux et non aqueux et plusieurs de systèmes de transmutation en réacteur ou pilotés par des accélérateurs [1].

La deuxième réunion internationale d’échange d’informations, qui a eu lieu en novembre 1992 dans les locaux de l’Argonne National Laboratory (Illinois, États-Unis), a révélé la nécessité de cerner les besoins et les priorités de la recherche. Cette réunion a permis d’identifier plusieurs thèmes nouveaux d’importance, notamment le contexte juridique ainsi que les motivations et les conséquences de la voie de la séparation/transmutation sur l’ensemble du cycle du combustible suivant les pays et les politiques nucléaires. C’est pourquoi, il a été décidé d’articuler les travaux sur la séparation et la transmutation entrepris par le Comité chargé des études techniques et économiques sur le développement de l’énergie nucléaire et le cycle du combustible de l’OCDE-AEN autour d’une comparaison des études systémiques déjà entreprises dans ce domaine [2].

La troisième réunion internationale d'échange d'informations s'est tenue en décembre 1994 au Centre d'études du CEA à Cadarache (France). Cette manifestation, qui a réuni plus de 80 participants de 11 pays auxquels s'étaient joints la Russie et l'AIEA, témoignait du formidable intérêt manifesté par la communauté internationale pour ce sujet et de la prise de conscience par les pays de l'OCDE de la véritable portée de la séparation et de la transmutation. Elle a permis de dégager les éléments de base indispensables à la poursuite des études comparatives consacrées au bilan global des coûts et avantages de l'introduction de la séparation et de la transmutation dans le cycle du combustible nucléaire [3].

La quatrième réunion internationale d'échange d'informations a été organisée à Mito, au Japon, en septembre 1996 à l'invitation de la Science and Technology Agency, de JAERI et de PNC. Près de 100 spécialistes de 11 pays de l'AEN et de Russie ainsi que de l'AIEA et de la Commission des Communautés européennes ont assisté à cette réunion qui, comme les précédentes, était consacrée aux études systémiques de la séparation et de la transmutation. Quarante communications y ont été présentées au total [4]. Un large consensus s'y est dégagé quant aux voies à suivre. Plusieurs délégations ont souligné la nécessité d'établir un rapport d'analyse systémique et ont proposé de participer à sa rédaction.

La cinquième réunion internationale d'échange d'informations a été organisée à Mol, en Belgique, en Novembre 1998 à l'invitation du Centre de Recherches Nucléaires SCK•CEN. Près de 130 participants ont assisté à cette réunion. Quarante trois communications et 13 posters y ont été présentées. L'essence même de cette réunion peut plus particulièrement se définir par rapport à la très remarquée avancée technologique dans la technique de séparation résultant de celle de Ln/An et de l'isolement de l'Am en laboratoires. La transmutation quant à elle fut discutée de manière plus générale, s'intéressant à la transmutation dans des réacteurs à eau légère (cible Am), dans des RNRs et des ADS. La fiabilité et les principaux défis technologiques de ces ADS furent soulignés.

1.2 Pourquoi la séparation et la transmutation ?

L'exploitation de l'énergie nucléaire produit régulièrement une masse de combustible usé qui contient, à côté des produits de fission à vie courte, une grande quantité d'actinides et de produits de fission de toxicité très élevée et de périodes radioactives très longues. Ces nucléides représentent l'inventaire radiotoxique[a)] à long terme[b)] qui demeure dangereux pendant des périodes dépassant l'imagination humaine.

Les processus classiques de retraitement permettent de recycler le gros des actinides majeurs (U et Pu), tandis que les actinides mineurs AM [essentiellement le neptunium (Np), l'américium (Am) et le curium (Cm)], dont les périodes radioactives peuvent atteindre 2 millions d'années, restent auprès des produits de fission et sont vitrifiés avant d'être enfouis dans des dépôts profonds. La séparation des actinides mineurs (Np, Am, et Cm) et de certains produits de fission est une méthode qui permettrait de réduire la radiotoxicité à long terme des déchets résiduels d'un facteur proportionnel au rendement de séparation. Les actinides mineurs ainsi récupérés pourraient être recyclés dans le cycle du combustible et réintégrés à l'inventaire fissile et fertile des réacteurs pour y être transmutés en isotopes à vie courte. C'est ainsi que, progressivement, les actinides mineurs et certains produits de fission à vie longue

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a) *Voir à la Partie II, section 4.1*, la définition de l'inventaire radiotoxique

b) Dans ce rapport, le court terme désigne les 100 premières années, le moyen terme la période comprise entre 100 et 1000 ans et le long terme la période au-delà de 10 000 ans.

pourraient être incinérés. En principe, cette technique permettrait d'atténuer le risque de contamination à long terme que présentent les déchets de haute activité et de raccourcir le temps de séjour nécessaire des déchets contenant des actinides dans les dépôts de déchets en formation géologique. La séparation et la transmutation sont donc a priori capables d'écourter la période de radiotoxicité, bien qu'il reste un certain nombre de difficultés pratiques à surmonter avant d'y parvenir.

1.3 Pourquoi une étude systémique de la séparation et de la transmutation ?

Les répercussions de la mise en œuvre des procédés de séparation sur le cycle du combustible avec retraitement varient largement en fonction des effluents ou flux de produits dans lesquels ces nucléides sont entraînés. Toute modification du procédé de séparation a d'importantes conséquences sur la qualité et la nature des produits nucléaires séparés. C'est pourquoi, une analyse des conséquences de la séparation des actinides mineurs et des produits de fission qui se veut complète doit reposer sur une étude systémique de l'impact des produits recyclés sur les déchets et sur la sûreté des différentes options.

On peut se demander que faire des concentrés une fois que les actinides mineurs et les produits de fission à vie longue ont été isolés. La première solution est le conditionnement associé au stockage à long terme réversible où la stabilité de la matrice et la décroissance naturelle jouent un rôle important. La deuxième solution consiste à les transformer en cibles pour les irradier ensuite. L'irradiation des cibles peut engendrer des produits de transmutation stables, mais aussi des nucléides fortement radiotoxiques de périodes moyennes.

On trouvera dans ce rapport une étude des différentes solutions permettant d'atténuer la radiotoxicité du produit final, assortie d'une analyse systémique limitée des principales options qui se veut une tentative pour éclairer les choix parmi le dédale de solutions possibles. L'analyse systémique préliminaire part de la situation actuelle des techniques employées dans le cycle du combustible et met en évidence quelques avancées des technologies de séparation et de transmutation qui pourraient donner naissance à un cycle du combustible avancé permettant une réduction globale de l'inventaire radiotoxique et de son impact sur la biosphère.

1.4 Groupe d'experts

Un groupe d'experts sur l'analyse systémique, auquel étaient représentés 12 pays et deux organisations internationales, a été constitué en 1996. Il a été demandé à ce groupe de préparer un rapport, le plus complet possible, sur l'analyse systémique de la séparation et la transmutation afin de proposer aux décideurs un document transparent, faisant autorité, où seraient décrites les répercussions de la séparation et de la transmutation sur les politiques adoptées en matière du cycle du combustible et de gestion des déchets. Il s'agissait notamment de bien montrer les conséquences de toute décision concernant la séparation et la transmutation sur la technologie, la sûreté à long terme, et les aspects économiques du cycle du combustible.

Le groupe de travail (voir Annexe A) s'est inspiré des différents rapports d'analyse nationaux et internationaux [5-10] publiés pour en tirer des conclusions quant au rôle que pourraient jouer la séparation et la transmutation dans le cycle du combustible et à l'intérêt respectif des différentes options prises en matière de recherche et de développement. La qualité des comparaisons entre scénarios et options dépend de l'harmonisation des critères et des contraintes utilisées.

1.5 Objectifs du rapport

Dans l’ensemble, ce rapport se veut une analyse scientifique et technique de l’impact de la séparation et de la transmutation sur l’aval du cycle du combustible. Les chapitres d’introduction, que l’on trouvera dans la Partie I : Présentation Générale, sont destinés à éclairer les responsables politiques et industriels sur les enjeux de la séparation et de la transmutation sans entrer dans les détails. Les chapitres techniques de la Partie II : Analyse Technique et Étude Systémique, sont consacrés à une analyse approfondie de l’introduction de la séparation et de la transmutation dans un scénario du cycle du combustible plus évolué et des conséquences d’une série de décisions. Ces chapitres techniques s’intitulent : *Séparation, transmutation, analyse des risques et gestion des déchets* et s’accompagnent d’une description des principaux programmes de recherche et de développement au Japon et en France. Le rapport fait une large place aux aspects de la sûreté à long terme sur lesquels la séparation et la transmutation pourrait avoir un impact dans l’hypothèse de la mise en place d’une industrie du cycle du combustible plus évoluée et de la poursuite du développement de l’électronucléaire. La séparation, en tant que technologie isolée, serait intéressante pour la gestion des déchets de haute activité, car elle permettrait de mieux stabiliser les nucléides à vie longue que la vitrification, de réduire le volume de déchets de haute activité et, éventuellement, de récupérer certains éléments précieux et rares. Cependant, dans ce rapport, la séparation est analysée essentiellement en tant qu’étape préliminaire à la transmutation.

Ce document doit être considéré comme un rapport d’évaluation décrivant l’état actuel de la séparation et de la transmutation assorti d’une analyse systémique préliminaire des conséquences de ces techniques sur le cycle du combustible et la gestion des déchets.

2. ÉTAT ACTUEL ET PERSPECTIVES DES TECHNOLOGIES DE SÉPARATION ET DE TRANSMUTATION

La séparation des actinides mineurs et des produits de fission à vie longue du combustible usé et la transmutation, c'est-à-dire la transformation, par des réactions nucléaires, de radionucléides à vie longue en éléments à vie courte ou stables sont deux techniques qui doivent être considérées comme des compléments ou des améliorations du cycle avec retraitement. Au cours de ce cycle, le plutonium et l'uranium sont séparés du combustible nucléaire et recyclés dans le cycle comme le montre la Figure I.1. On notera sur cette figure que le cycle du combustible se divise en une partie « amont », qui recouvre toutes les opérations effectuées avant que le combustible ne soit chargé dans le réacteur, et une partie « aval » englobant tous les traitements effectués après l'utilisation du combustible en réacteur. La séparation et la transmutation sont, par essence, des procédés intervenant en aval du cycle puisqu'elles sont conçues comme des moyens d'améliorer les options de stockage des déchets. Idéalement, la mise en œuvre de la séparation et de la transmutation pourrait déboucher sur un cycle du combustible avancé, faisant appel à une technologie spécifique pour optimiser les résultats (voir Figure I.2). Cependant, comme le recyclage dans les réacteurs de conception actuelle doit également être pris en compte, la séparation et la transmutation peuvent aussi avoir des répercussions sur l'étape de fabrication du combustible des cycles avec retraitement.

Nous décrivons dans ce chapitre l'état actuel et les perspectives de la technologie de la séparation et de la transmutation à travers l'examen des stratégies de recyclage, des solutions possibles pour le retraitement (la séparation) et de la technologie de fabrication des combustibles et des cibles. Nous ne nous limitons pas aux actinides mineurs et aux produits de fission à vie longue mais décrivons également le recyclage du plutonium et de l'uranium, étant donné que l'intérêt de la séparation et de la transmutation ne peut s'apprécier que par comparaison avec le recyclage du plutonium.

2.1 Stratégies

2.1.1 Plutonium et uranium

La séparation du plutonium du combustible usé permet d'abaisser la radiotoxicité à long terme des déchets résiduels d'un facteur qui, théoriquement, peut atteindre 10. Ce chiffre découle simplement du fait que la quantité de plutonium présente dans le combustible UO_2 usé est à peu près dix fois supérieure à celle des actinides mineurs dont la radiotoxicité est plus ou moins équivalente à celle des isotopes du plutonium. L'uranium séparé de ses produits de filiation contribue relativement peu à la radiotoxicité. Pourtant, en recyclant le plutonium sous forme de MOX dans les REO (REO-MOX) on ne divise la radiotoxicité que par trois, si l'on considère le combustible MOX usé comme un déchet.

Figure I.1 **Schéma de principe du cycle avec retraitement du combustible des réacteurs à eau ordinaire**

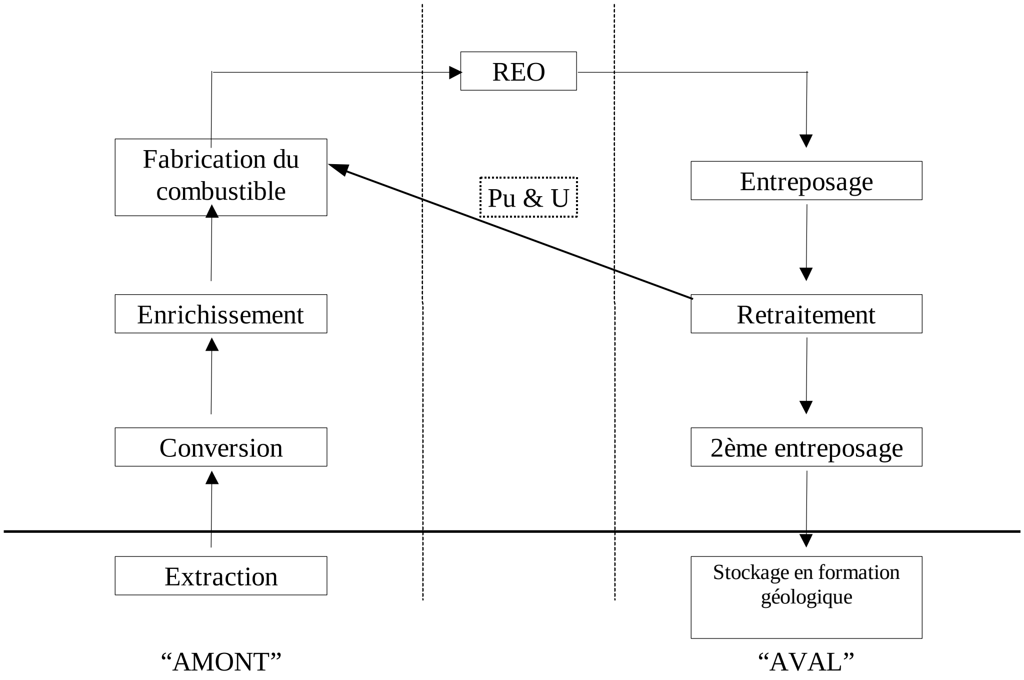
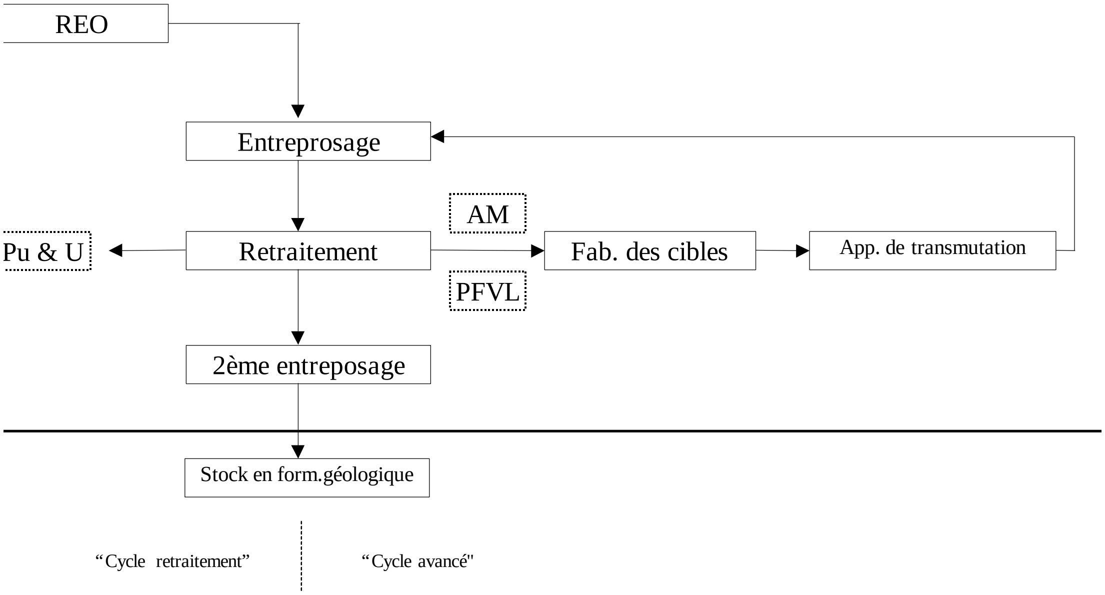


Figure I.2 **Représentation schématique de l'aval d'un cycle du combustible avancé avec recyclage des actinides mineurs**



Pour recycler le plutonium, la méthode la plus simple consiste à charger les REO en assemblages à oxydes mixtes (MOX). Un assemblage au MOX contient 35 kg de plutonium avant son irradiation et 25 kg après, ce qui représente une consommation de 10 kg par assemblage combustible, à comparer aux 5 kg de plutonium qui sont produits dans un assemblage standard à oxyde d'uranium. À l’heure actuelle, des réacteurs sont exploités en France, en Allemagne, en Belgique et en Suisse, avec des cœurs comportant jusqu’à 30 pour cent d’assemblages combustibles MOX. Dans ce cas, on produit $[(2x5) - (1x10)] \cong 0$ kg de plutonium, soit un bilan presque nul. Par conséquent, augmenter le nombre d’assemblages MOX dans un cœur de réacteur (jusqu’à 100 pour cent) permettra une consommation nette de plutonium, mais cette solution nécessite que l’on modifie le rapport de modération. Il est possible d’obtenir des taux de consommation du plutonium plus élevés en mélangeant le plutonium avec un matériau inerte sur le plan neutronique que l’on appelle matrice inerte, puisque dans ce cas la source de production de plutonium (^{238}U) n’existe plus. Cependant, la présence d’une matrice inerte modifiera le coefficient Doppler du combustible, en raison de l’absence de résonance de l’^{238}U. Une autre possibilité, à l’étude actuellement, consiste à mélanger le plutonium avec du thorium.

Le nombre d’étapes de recyclage dans un REO est limité par l’accumulation d’isotopes du plutonium non-fissiles dans un spectre de neutrons thermiques. C’est pourquoi, les systèmes qui produisent des flux importants de neutrons rapides (réacteurs rapides, systèmes sous-critiques pilotés par des accélérateurs) sont toujours nécessaires pour incinérer le plutonium dégradé provenant du recyclage du plutonium dans des REO. Les calculs effectués pour la gestion du combustible ont montré que ce type d’appareil peut être exploité en symbiose avec un parc de REO. En Europe, les spécifications du projet CAPRA ont été définies pour la conception d’un réacteur rapide qui consommera le plus possible de plutonium. Dans la conception de référence actuelle de CAPRA, il est prévu d’utiliser un cœur à oxydes mixtes dont le combustible MOX contiendrait jusqu’à 45 pour cent d’oxyde de plutonium. Cependant, comme les solutions d’acide nitrique actuellement employées dans le procédé PUREX ne sont pas capables de dissoudre ce combustible, on envisage de recourir à d’autres combustibles. Le Japon, mais aussi certains pays d’Europe, se tournent actuellement vers des combustibles sous forme de nitrure, puisque le PuN se dissout facilement dans le HNO_3, qui est le milieu aqueux utilisé dans le procédé PUREX. Mais cette solution exige que l’on utilise des réactifs fortement enrichis en ^{15}N pour produire le PuN si l’on veut éviter la formation de ^{14}C dans le combustible usé.

À l’heure actuelle, on recycle peu l’uranium de retraitement (URT), parce que, d’une part, l’uranium naturel n’est pas cher et, d’autre part, l’URT contient un peu de ^{236}U qui est un poison neutronique diminuant la réactivité de l’uranium de retraitement. Si l’on tarde à recycler l’URT dans les réacteurs, voire si l’on abandonne cette solution, on risque de voir s’accumuler les produits très radiotoxiques de la décroissance de l’^{232}U et de l’^{234}U, notamment le ^{228}Th et le ^{208}Tl. Il conviendra donc de prendre en compte les stocks d’URT et d’uranium appauvri issu du retraitement dans les stratégies globales de gestion des déchets radioactifs. À très long terme, la radioactivité totale de l’uranium appauvri, s’il est considéré comme un déchet, dépasse celle du neptunium.

2.1.2 Actinides mineurs

Si le recyclage du plutonium peut être efficacement réalisé à l’échelle industrielle, le recyclage de l’américium doit être envisagé ensuite, et cela pour les raisons suivantes :

- l’américium se place en deuxième position par sa contribution à la radiotoxicité du combustible usé ;
- dans les évaluations des performances des dépôts souterrains, la contribution de l’américium à la radiotoxicité est prépondérante au cours des 1 000 premières années ;

- l'^{241}Am est le précurseur du ^{237}Np à vie longue qui, en général, domine les scénarios d'évolution normale dans les évaluations des performances en raison de sa longue période radioactive ;
- le recyclage du plutonium a pour effet d'accroître la quantité d'américium produit.

Comme pour le plutonium et l'uranium, la réaction de transmutation la plus intéressante pour les actinides mineurs est la fission, étant donné que la capture ou les réactions (n, 2n) produisent en général d'autres actinides à vie longue. La solution la plus efficace consiste alors à recourir à un flux de neutrons rapides dans lequel la plupart des isotopes d'actinides peuvent être fissionnés. Toutefois, même dans ce cas, la capture suivie d'une fission n'est pas à négliger. Dans les spectres thermiques, on a besoin de neutrons supplémentaires pour convertir les isotopes non fissiles en isotopes fissiles (par exemple, l'^{241}Am en ^{242m}Am), d'où la nécessité d'un enrichissement supplémentaire du combustible utilisé.

Les stratégies de transmutation des actinides mineurs dans des systèmes à flux rapides ressemblent aux voies choisies pour la transmutation du plutonium, et cela n'a rien de surprenant. Ces actinides peuvent être soit mélangés de façon homogène au combustible MOX, appelé combustible MINOX, ou incorporés à des assemblages combustibles spéciaux dans des matrices inertes à base d'oxydes ou éventuellement de nitrures et de carbures. Du point de vue de la physique des réacteurs, ces deux solutions sont réalisables ; les seules contraintes proviennent de la fabrication des combustibles.

À la place du recyclage, il est possible de recourir à une stratégie consistant à incinérer le plus possible d'actinides mineurs lors d'une irradiation unique prolongée suivie du stockage définitif, solution appelée encore « cycle ouvert ». On peut, pour ce faire, utiliser une installation d'irradiation à haut flux thermique ou intégrer les actinides à un sous-assemblage sous-critique d'un réacteur rapide. En sélectionnant avec soin la matrice inerte dans laquelle le déchet résiduel formé sera immobilisé, il est possible d'atténuer encore les effets radiologiques du stockage puisque l'on aura ajouté une barrière supplémentaire. Des minéraux géologiquement stables, comme le zircon ($ZrSio_4$) ou la monazite ($CePO_4$) comptent parmi les candidats envisagés pour cette application.

2.1.3 Produits de fission

Du point de vue de la radiotoxicité des déchets, la transmutation des produits de fission présente peu d'intérêt. Après 250 ans, la majorité d'entre eux a disparu par décroissance radioactive, et leur contribution à la radiotoxicité du combustible usé, d'abord très forte au cours des cent premières années de stockage, est devenue faible. Cependant, il existe des produits de fission qui sont très mobiles dans certains environnements géologiques et sont responsables d'une fraction importante des effets radiologiques du stockage souterrain. Par ailleurs, il faut comptabiliser dans les effets radiologiques à long terme de la production électronucléaire les effluents gazeux et liquides produits lors du traitement des combustibles usés. La séparation et/ou le piégeage de ces produits dans les flux de déchets, suivis par leur transmutation permettraient d'atténuer les risques radiologiques à long terme. De ce point de vue, les produits de fission à considérer en priorité sont le technétium (Tc), le césium (Cs) et l'iode (I), car le ^{99}Tc et le ^{135}Cs sont les isotopes prépondérants dans les analyses de risques relatives au stockage du combustible usé, et l'^{129}I, qui n'est pas incorporé aux déchets de haute activité vitrifiés, est l'isotope qui a les effets radiologiques les plus importants dans les effluents issus du retraitement ou même dans le combustible usé placé dans certaines formations géologiques.

Le technétium présent sous une seule forme isotopique, le ^{99}Tc, peut être transmuté par capture neutronique en ruthénium (^{100}Ru) qui est un métal noble stable. Cependant, la transmutation du ^{99}Tc dans des réacteurs thermiques tels que les REO de conception actuelle, sera difficile parce qu'elle nécessitera de très longues périodes de transmutation et des inventaires considérables. On obtiendrait de meilleurs résultats dans les réacteurs à eau lourde, des sous-assemblages modérés de réacteurs rapides et, éventuellement, dans des réacteurs à hauts flux hybrides. Dans tous les cas, il faudra surenrichir le combustible, sauf si on pouvait appliquer des techniques de séparation isotopique.

Contrairement au technétium, le césium séparé du combustible usé ne se présente pas sous la forme d'un seul isotope mais d'un mélange à quantités à peu près égales de ^{135}Cs à vie longue, de ^{137}Cs à vie courte et de l'isotope stable, le ^{133}Cs. Une capture neutronique parasite se produira par conséquent pendant l'irradiation, surtout dans le ^{133}Cs. Par ailleurs, étant donné que le ^{135}Cs a une section efficace d'absorption neutronique assez faible, la transmutation du césium ne peut pas être considérée comme réalisable.

Bien que l'iode séparé du combustible usé se présente sous la forme d'un mélange d'^{129}I et d'^{127}I, la proportion du deuxième isotope peut être jugée acceptable (16 pour cent). Il serait donc théoriquement possible de transmuter par une capture neutronique l'^{129}I en xénon (^{130}Xe) qui est un gaz rare. Cette transmutation de l'iode dans des réacteurs thermiques exigerait également des périodes de transmutation très longues et de forts chargements du réacteur.

2.2. Retraitement

2.2.1 Plutonium et uranium

Plusieurs pays pratiquent le retraitement du combustible usé provenant des REO à l'échelle industrielle (France, Royaume-Uni, Inde et Russie). Dans ces usines, les éléments combustibles des réacteurs à eau légère subissent un traitement destiné à récupérer l'uranium et le plutonium. On peut aussi y traiter des éléments combustibles MOX à condition de les diluer avec des éléments combustibles standard à l'UO_2 afin de respecter les limites fixées pour les doses et la quantité de plutonium. Il est impossible de retraiter, dans les usines de retraitement actuelles, le combustible des réacteurs rapides à forte teneur en Pu, comme celui des assemblages CAPRA actuels, car il ne se dissout pas entièrement dans la solution d'acide nitrique standard.

Plusieurs pays Membres de l'OCDE, mais aussi l'Inde et la Russie, ont choisi de retraiter leur combustible usé. Parmi les exceptions notables, il convient de citer le Canada, l'Espagne, les États-Unis et la Suède qui envisagent de stocker directement leur combustible irradié. Toutes les usines de retraitement emploient, pour extraire le plutonium et l'uranium, le procédé PUREX, un procédé d'extraction chimique par voie humide comportant la dissolution du combustible dans de l'acide nitrique et l'extraction de l'U et du Pu à l'aide de phosphate tributylique (TBP). Dans l'état actuel de la technique, ce retraitement permet de séparer 99.88 pour cent de l'uranium et du plutonium contenus dans les combustibles de REO. Les 0.12 pour cent restants sont incorporés aux déchets vitrifiés ou enrobés, avec les coques et les embouts, dans du béton.

Comme le combustible reste la propriété de la compagnie d'électricité, tous les déchets issus du retraitement (vitrifiés ou en matrices de béton) sont renvoyés dans leur pays d'origine. Mille kilos de combustible irradié à raison de 33 GWj donnent, après retraitement, 955 kg d'uranium, 10 kg de plutonium et environ 2 m^3 de déchets vitrifiés ou en matrices de béton. L'évolution actuelle laisse prévoir une nouvelle réduction du volume de déchets qui résultera de l'optimisation du procédé de retraitement et de la gestion des déchets de haute activité.

Le coût du retraitement dépend du moment où il intervient, du taux d'irradiation du combustible, etc. Néanmoins, des estimations raisonnables effectuées par l'AEN donnent un ordre de grandeur de 720 ECU/kg d'U. Mais il convient de comptabiliser ce coût avec celui des autres étapes de l'aval du cycle du combustible et de le comparer à ceux du stockage direct. Cette étape permet en effet d'économiser sur l'extraction et le traitement d'uranium neuf et sur les services d'enrichissement. En tout état de cause, les économies réalisées sur la demande d'uranium naturel et de services d'enrichissement du combustible neuf sont la principale justification de l'utilisation d'éléments combustibles MOX dans les REO et du recyclage du plutonium après retraitement.

Certains pays étudient également les possibilités de retraiter le combustible usé par des procédés pyrochimiques utilisant des sels fondus. L'intérêt principal de cette technologie tient à l'excellente tenue au rayonnement des sels fondus qui permet de raccourcir les temps de refroidissement. Cette technologie a été mise au point pour le projet Integral Fast Reactor/ALMR aux États-Unis, et continue d'être étudiée par les compagnies d'électricité japonaises (CRIEPI).

2.2.2 Actinides mineurs

À l'heure actuelle, les actinides mineurs que sont le neptunium (Np), l'américium (Am) et le curium (Cm) ne sont pas séparés dans des usines de retraitement de taille industrielle. Ces éléments sont incorporés aux déchets de haute activité vitrifiés avec les produits de fission. Deux facteurs expliquent cette situation : d'une part, on avait jusqu'à présent peu de raisons de séparer ces éléments, et d'autre part, il était impossible, avec le procédé PUREX, de séparer les actinides trivalents (Am, Cm). On peut, en revanche, récupérer le neptunium en adaptant le procédé PUREX : il suffit de modifier le degré d'oxydation du Np pour l'extraire avec l'uranium et le plutonium et ensuite le séparer du flux de métaux lourds soit seul, soit par coextraction avec l'uranium, soit encore en le dirigeant vers le flux de plutonium.

De nombreuses recherches ont été engagées afin de mettre au point des méthodes efficaces pour séparer les actinides trivalents des flux de déchets du procédé PUREX. Ces dernières années, plusieurs procédés ont été mis au point : DIAMEX en France, DIDPA au Japon, TRPO en Chine et TRUEX aux États-Unis et au Japon. La difficulté principale à résoudre consiste à séparer les actinides trivalents (Ans) des lanthanides trivalents (Lns). Étant donné que le rapport entre les lanthanides et les actinides peut varier de 10 à 20 suivant le taux d'irradiation du combustible, la purification des actinides mineurs trivalents (fraction Am-Cm) exige des taux de séparation élevés. La recherche dans cette direction est l'une des facettes des travaux sur les procédés DIDPA et TRUEX. Cependant, on étudie également d'autres procédés faisant appel à de nouvelles molécules extractantes. D'importants travaux de recherche fondamentale ont été entrepris sur des extractants et systèmes complexes capables de dissocier les actinides des lanthanides. Les progrès récents réalisés en Chine, en Allemagne, en France et au Japon sur les composés « CYANEX 301 » paraissent très prometteurs.

Pour remplacer les méthodes de traitement en milieu aqueux, les États-Unis (ANL) et le Japon (CRIEPI) travaillent activement au développement de méthodes de séparation pyrochimiques. Ces techniques de retraitement par voie sèche sont adaptées aux combustibles usés à haut taux de combustion, refroidis peu de temps, car elles ne sont pas sensibles au dommage d'irradiation et sont moins contraignantes vis à vis du risque de criticité. On peut ainsi traiter directement les combustibles en alliages métalliques à base de transuraniens par électrolyse combinée à une extraction réductrice dans un bain de sels et de métaux fondus (Cd). Cette technologie fait l'objet d'études théoriques et d'expériences en laboratoire. Les températures élevées auxquelles les procédés sont mis en œuvre et l'utilisation de réactifs corrosifs compliquent l'application industrielle à grande échelle de cette méthode. Toutefois, comme le recyclage multiple de combustibles de réacteurs rapides à haut taux de combustion devrait se révéler très difficile avec des méthodes aqueuses, les techniques pyrochimiques demeurent une solution alternative valable pour l'avenir.

2.2.3 Produits de fission

Dans le procédé PUREX, les produits de fission se répartissent entre trois flux de déchets différents :

- les effluents gazeux rejetés dans l'air qui contiennent essentiellement un gaz rare, le ^{85}Kr ;
- les effluents liquides de faible activité rejetés dans la mer ou les océans qui contiennent majoritairement de l'^{129}I. ;
- les déchets liquides de haute activité (DHA) où sont concentrés tous les autres produits de fission.

Au cours de l'étape de dissolution du procédé PUREX, l'iode contenu dans le combustible s'oxyde en iode élémentaire. Suivant les conditions dans lesquelles se déroule le procédé, cet iode peut être rejeté avec les effluents gazeux ou concentré dans la solution. La première méthode permet une séparation très efficace, mais l'iode doit ensuite être piégé dans les effluents gazeux. Pour ce faire, on utilise normalement un système de lavage à contre-courant ou des tamis moléculaires imprégnés d'argent permettant d'atteindre des facteurs de décontamination d'environ 10^4.

Le technétium se présente après le retraitement en partie sous une forme insoluble avec les autres métaux nobles (Ru, Rh, Pd) et en partie sous forme de l'espèce soluble $^{99}TcO^-_4$. Il est techniquement possible de séparer le technétium soluble moyennant quelques adaptations du procédé PUREX. Il peut être séparé sous forme d'acide pertechnétique ($HTcO_4$) par des molécules extractantes de type monoamide ou par une méthode d'absorption sur du charbon actif. On parvient également à séparer par précipitation le technétium des déchets de haute activité liquides en réduisant la concentration d'acide nitrique. Avant toute transmutation, les deux fractions doivent être recombinées.

Au cours du procédé PUREX, le césium reste dans les déchets de haute activité liquides. Une méthode d'absorption à base d'échangeurs d'ions inorganiques a été mise au point pour la séparation du Cs et a fait ses preuves avec des déchets de haute activité réels. Récemment, on a développé à l'échelle du laboratoire une nouvelle méthode permettant de séparer le césium de cette solution à l'aide de molécules macrocycliques fonctionnalisées (calixarènes) dans des solutions dont on imprègne des membranes liquides. Il est possible d'extraire ainsi des solutions acides 99.8 pour cent du césium lorsqu'il traverse la membrane.

2.3 Technologie de fabrication des combustibles et des cibles

2.3.1 Plutonium et uranium

La fabrication des assemblages au MOX fait appel à une technologie courante utilisée à l'échelle industrielle. La capacité annuelle des usines de fabrication en Belgique, en France, au Royaume-Uni et au Japon avoisine 200 t de ML, mais elle devrait être portée à 300 t de ML en 2000. La fabrication du MOX revient à peu près quatre fois plus cher que la fabrication du combustible standard à l'oxyde d'uranium (275 à 300 dollars/kg d'U) en raison du systèmes des garanties des matières fissiles et de la sévérité des normes de sûreté pour l'essentiel destinées à assurer la protection radiologique des travailleurs par l'emploi de boîtes à gants. En revanche, le recyclage du plutonium permettra d'économiser sur des opérations relevant de l'amont du cycle du combustible, notamment les activités minières et l'enrichissement, qui contribuent pour une bonne part au débit de dose reçu par les travailleurs et aux coûts de la partie amont du cycle du combustible.

Les combustibles à matrices inertes sont le fruit de travaux récents effectués pour optimiser les taux d'incinération. Dans ces combustibles, on mélange les composés à transmuter avec une substance inerte sur le plan neutronique. On peut utiliser, pour ce faire, une dispersion (échelle macroscopique) ou une solution solide (échelle microscopique). Parmi les matériaux envisagés comme matrices inertes, on peut citer des oxydes tels que la spinelle ($MgAl_2O_4$), l'oxyde d'yttrium (Y_2O_3), un oxyde mixte d'yttrium et d'aluminium ($Y_3Al_5O_{12}$), des métaux comme le tungstène ou le vanadium, ou encore le carbure de silicium (SiC). Toutefois, la fabrication et la caractérisation de ces combustibles méritent des recherches supplémentaires, notamment sur leur comportement sous irradiation et leur compatibilité avec les techniques de retraitement actuelles.

Outre les combustibles oxydes, on envisage d'utiliser, pour l'incinération du plutonium, des combustibles nitrures et métalliques. Ces deux nouvelles formes de combustible sont compatibles avec le sodium liquide et pourraient donc être utilisées dans des réacteurs incinérateurs rapides. Les combustibles nitrures proposés pour des cœurs de réacteurs incinérateurs de type CAPRA ont été produits à l'échelle du laboratoire dans les années 60. Comme nous l'avons vu plus haut, l'intérêt actuel pour les combustibles nitrures tient en partie au fait que les combustibles oxydes à forte teneur en plutonium ne se dissolvent pas dans l'acide nitrique, contrairement aux combustibles nitrures. Ces derniers combustibles présentent, par ailleurs, une conductivité thermique nettement supérieure, ce qui permettra d'abaisser la température au centre de l'élément combustible et ainsi d'accroître les marges de sûreté. Le principal inconvénient de ces combustibles est qu'ils produisent du ^{14}C au cours de la réaction (n, p) de l'^{14}N. L'enrichissement en ^{15}N de l'azote utilisé pour fabriquer le combustible est donc impératif. De plus, les procédés pyrochimiques étudiés au Japon faciliteraient le recyclage de l'azote enrichi.

Dans le cadre de recherches du projet IFR, les États-Unis ont étudié des combustibles métalliques formés d'un alliage d'uranium, de plutonium et de zirconium (Zr) et en ont produit à l'échelle du pilote. Comme les combustibles nitrures, les combustibles métalliques ont une meilleure conductivité thermique que les combustible oxydes et, de plus, présentent une bonne stabilité aux rayonnements, autorisant des taux de combustion élevés. Dans le concept de l'IFR/ALMR, le combustible métallique serait retraité par des techniques pyrochimiques faisant appel à l'emploi de sels fondus.

2.3.2 Actinides mineurs

Le recyclage du neptunium dans le combustible MINOX présenterait l'avantage de pouvoir s'effectuer dans les usines existantes de fabrication de MOX moyennant quelques adaptations mineures. Il s'agirait essentiellement d'améliorer le blindage biologique et d'automatiser davantage le procédé afin d'améliorer la protection contre les niveaux de rayonnements plus élevés. On prévoit de ce fait une hausse des coûts de fabrication d'environ 20 pour cent. L'Institut des transuraniens (ITU) de Karlsruhe a réussi à fabriquer des combustibles de composition $(U_{0.55}Pu_{0.40}Np_{0.05})O_2$ destinés à une campagne d'irradiation dans le cadre du projet CAPRA.

À cause des limites de doses à respecter lors la fabrication, on ne peut ajouter l'américium au combustible UO_2 qu'en petite quantité. La dose élevée de rayonnement γ n'est pas due seulement à l'américium mais à certains lanthanides qui sont présents sous forme d'impuretés. C'est pourquoi, la solution la plus efficace pour incinérer l'américium à grande échelle consistera à l'intégrer à un combustible placé dans une matrice inerte qu'il faudra préparer dans des installations spécialement conçues et équipées de blindages extrêmement efficaces et de systèmes de télémanipulation. La matrice inerte qui sera utilisée pour incinérer l'américium serait constituée de matériaux proches de ceux envisagés pour l'incinération du plutonium (voir plus haut). Cependant, les propriétés de l'oxyde d'américium (AmO_2) ne sont pas très encourageantes : il a une mauvaise conductivité thermique, une forte affinité avec l'oxygène et, de surcroît, réagit avec le sodium liquide.

Pour incinérer l'américium et, éventuellement, d'autres actinides mineurs, il serait par conséquent plus intéressant de les recycler sous formes métalliques ou nitrures, avec ou sans matrice inerte. Le JAERI étudie actuellement cette solution.

Il semble impossible à l'heure actuelle de recycler le curium pur dans un système spécialement dédié à cette opération en raison des rayonnements α, γ et neutroniques très élevés que produisent sa décroissance et la fission spontanée. Une solution consisterait donc à entreposer le curium durant environ 100 ans, le temps nécessaire pour que les isotopes du curium à vie relativement courte (^{242}Cm, ^{243}Cm et ^{244}Cm) se transforment en isotopes de plutonium qui pourraient alors être recyclés comme nous l'avons décrit ci-dessus. Dans certains scénarios, on envisage encore de recycler un mélange d'américium et de curium. Néanmoins, il faut aussi prévoir une méthode efficace pour séparer au préalable l'américium du curium. .

2.3.3 Choix des cibles pour les produits de fission

Au cas où l'on opterait pour la transmutation des produits de fission, à savoir le technétium et l'iode, l'irradiation serait vraisemblablement réalisée dans des cibles spéciales. La recherche de matériaux adaptés ainsi que des expériences d'irradiation pilotes ont été entreprises dans le cadre de la collaboration européenne EFTTRA. Les premiers résultats de ces études ont montré que l'on peut utiliser comme matériau de cible du technétium métallique. On a développé un procédé de fabrication permettant de fondre le métal en aiguilles, et les expériences d'irradiation effectuées dans un spectre thermique (jusqu'à un taux de combustion d'environ 6 pour cent) n'ont pas mis en évidence des phénomènes de gonflement ou de désintégration de ces aiguilles.

En raison de sa volatilité et de sa réactivité chimique, l'iode ne peut pas être transmuté sous sa forme moléculaire. Il faudra donc utiliser des iodures métalliques. Les expériences réalisées dans le cadre de la coopération EFTTRA ont montré que, pour la transmutation de l'iode, l'iodure de sodium (NaI) serait le meilleur choix.

3. DESCRIPTION DES CYCLES DU COMBUSTIBLE

Définition de la séparation et de la transmutation

On appelle *séparation* un ensemble complexe de traitements chimiques et/ou métallurgiques destinés à séparer des nucléides, ou groupes de nucléides radiotoxiques spécifiques, de l'ensemble des radionucléides que contiennent les éléments combustibles usés et périodiquement déchargés des réacteurs nucléaires. Les nucléides, ou groupes de nucléides, que l'on a ainsi séparés peuvent être stockés tels quels ou transformés en nouveaux éléments combustibles ou cibles d'irradiation.

La *transmutation* est un terme général qui recouvre tant les transformations nucléaires élémentaires par capture d'un neutron que la fission de nucléides lourds, la spallation et d'autres réactions nucléaires faisant intervenir des neutrons. Dans le cadre de cette étude, la transmutation a pour but de réduire l'inventaire à long terme des nucléides radiotoxiques en convertissant les nucléides d'origine en radionucléides à vie courte ou en nucléides stables.

3.1 Les cycles du combustible nucléaire

L'expression cycle du combustible nucléaire recouvre toutes les opérations nécessaires pour alimenter les centrales en combustibles neufs et gérer les combustibles usés déchargés des réacteurs. On trouve sur la Figure I.3 les principales étapes d'un cycle du combustible avec retraitement qui, pour les réacteurs à eau ordinaire, sont les suivantes :

- *l'amont du cycle du combustible* englobe toutes les opérations depuis les activités de prospection du minerai d'uranium jusqu'au transport des éléments combustibles avant leur rechargement dans les réacteurs nucléaires, soit l'extraction et le traitement du minerai afin d'obtenir un concentré d'uranium, le conversion de ce concentré en hexafluorure pour l'expédier dans les usines d'enrichissement d'uranium où l'on fait passer la teneur en ^{235}U de son niveau naturel (0.72 pour cent) à 3 à 5 pour cent, et enfin, la fabrication des éléments combustibles.
- *l'aval du cycle du combustible* comprend à l'heure actuelle deux approches alternatives :
    - le cycle fermé, fondé sur le retraitement du combustible usé, opération qui permet de séparer et de recycler les matières énergétiques (essentiellement le plutonium) que ce combustible contient et d'optimiser ainsi la gestion des déchets qui seront ensuite evacués dans des dépôts en formation geologique ;
    - le cycle ouvert, qui consiste à enfouir directement les combustibles irradiés dans un dépôt en formation géologique après une période d'entreposage de durée variable.

On peut voir sur la Figure I.4 les flux annuels de matières dans le cycle du combustible d'un parc mixte de réacteurs comportant des REP et des RNR et, sur la Figure I.5, ceux d'un parc utilisant exclusivement des RNR pour la production d'énergie.

Nous examinerons dans ce rapport quatre modes de gestion du combustible :

3.1.1 Le cycle ouvert (CCO)

Ce cycle correspond au scénario dans lequel tout le combustible usé déchargé d'un réacteur nucléaire est stocké dans des ouvrages, le temps qu'il refroidisse suffisamment pour pouvoir être transporté dans un dépôt permanent aménagé dans des formations géologiques profondes.

3.1.2 Le cycle avec retraitement (CCF)

Ce scénario comporte une étape de « retraitement » destinée à récupérer l'uranium et le plutonium contenus dans les éléments combustibles dissous et à faire passer les produits de fission et actinides mineurs (AM = Np, Am, Cm) dans les déchets liquides de haute activité que l'on entreposera plusieurs années avant de les vitrifier. Les matrices vitreuses ainsi obtenues sont ensuite stockées dans des installations ouvragées de surface avant d'être enfouies en tant que déchets de haute activité dans des dépôts en formations géologiques.

3.1.3 Cycle du combustible avancé avec recyclage des transuraniens et de quelques produits de fission (CCA)

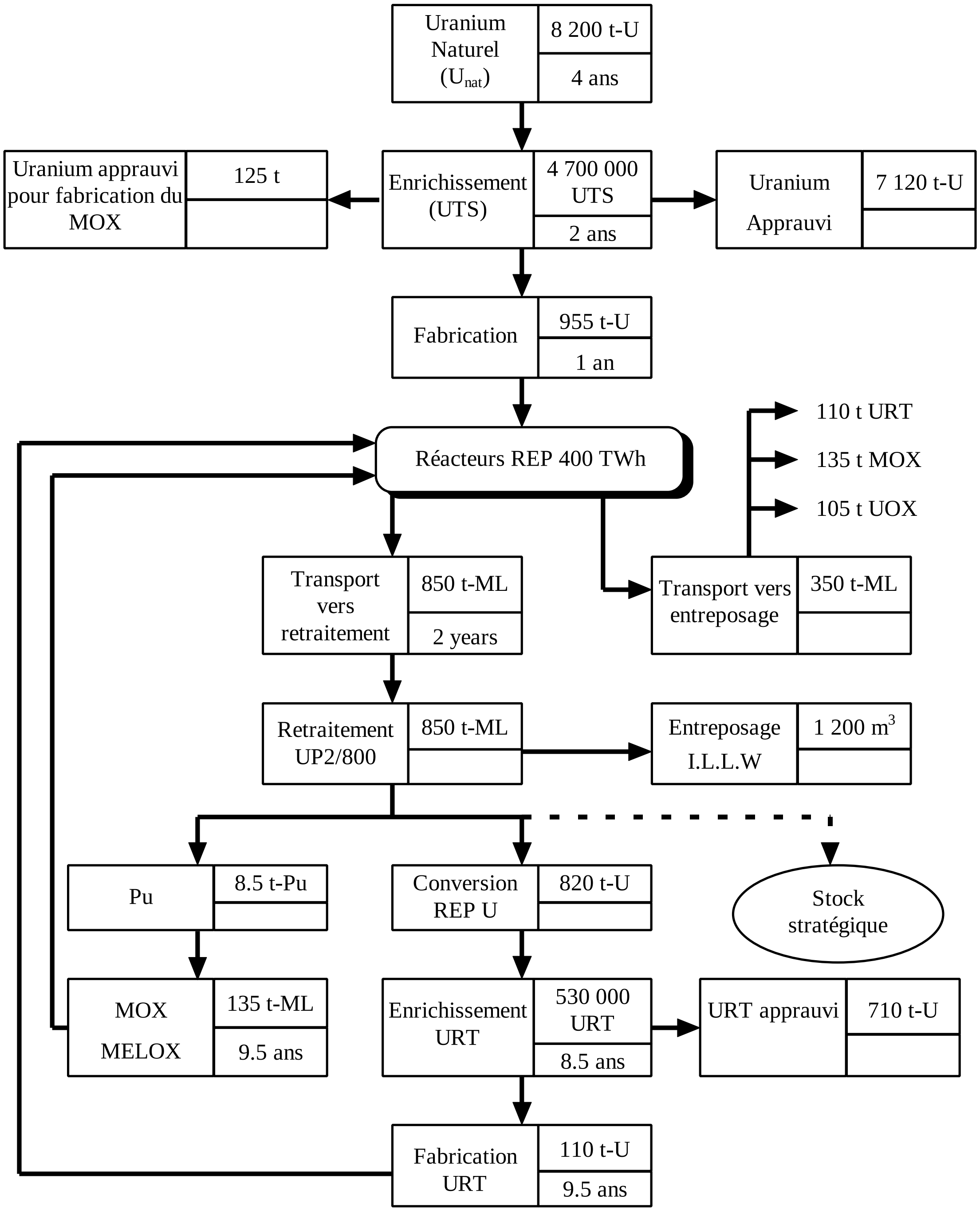
Ce cycle comporte une série de traitements chimiques et métallurgiques et de réactions nucléaires, dont l'objectif est de séparer tous les actinides (Pu, Am, Cm, Np) et certains produits de fission(^{99}Tc, ^{129}I) du flux principal de déchets et de les recycler comme cibles dans les centrales nucléaires ou dans des réacteurs nucléaires dédiés et/ou dans des systèmes hybrides afin d'en réduire de façon substantielle l'inventaire radiotoxique.

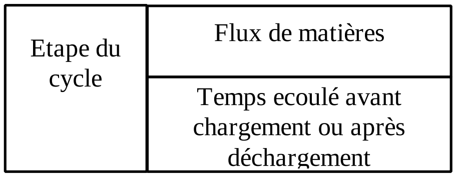
3.1.4 Stratégie à double strate

Il s'agit d'une combinaison du cycle avec retraitement (CCF) et du cycle avancé (CCA) aboutissant à la constitution d'un parc mixte de réacteurs consacrés à la production d'énergie et à la destruction des déchets. Dans ce système, les actinides mineurs et certains produits de fission rejetés comme déchets par les usines de retraitement classiques seraient recyclés à l'aide d'installations spécialisés de retraitement et de fabrication du combustible dans un parc de réacteurs mixte composé de réacteurs à eau ordinaire à combustible UO_2 (REO-UO_2), de réacteurs à eau ordinaire chargés en MOX (REO-MOX), de réacteurs rapides à combustible MOX (RNR-MOX) et de réacteurs rapides à combustibles métalliques ou nitrures, mais pourraient aussi être expédiés dans des réacteurs hybrides dédiés pour y subir une transmutation ou une incinération partielle.

On trouvera à l'annexe E les quantités et les caractéristiques des matières composant le combustible nucléaire et les assemblages combustibles déchargés. Pour faciliter la compréhension, ces données ont été établies seulement pour l'irradiation du combustible en REP. Les chiffres ne sont pas les mêmes pour les REB, les CANDU ou d'autres types de réacteurs, mais nous n'avons pas voulu les présenter ici pour éviter la multiplication des tableaux.

Figure I.3 **Schéma du cycle du combustible avec retraitement pour un parc de REO de 400 TWh (étude de cas française)**





Ce schéma repose sur les hypothèses suivantes:

- Les quantités retraitées de combustible UO_2 sont adaptées aux possibilités de recyclage du Pu dans le combustible MOX
- La capacité de l'usine UP2 de La Hague (850 t/an) pour le retraitement et de l'usine MELOX (135 t/an) pour la fabrication du MOX est utilisée à plein

Figure I.4 **Flux de matières dans un parc électronucléaire mixte de 700 TWh composé de REP et de RNR (étude de cas japonaise)**

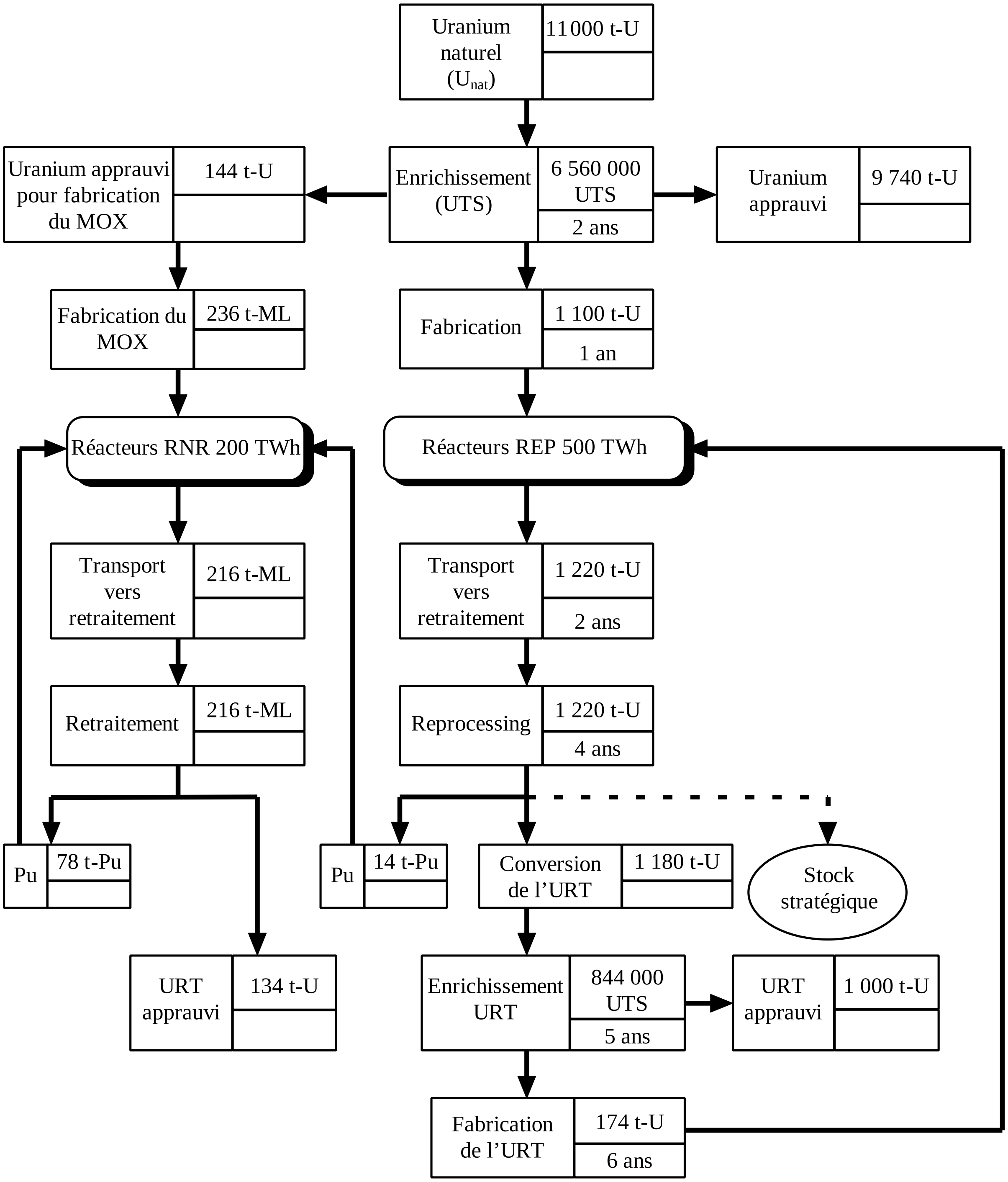
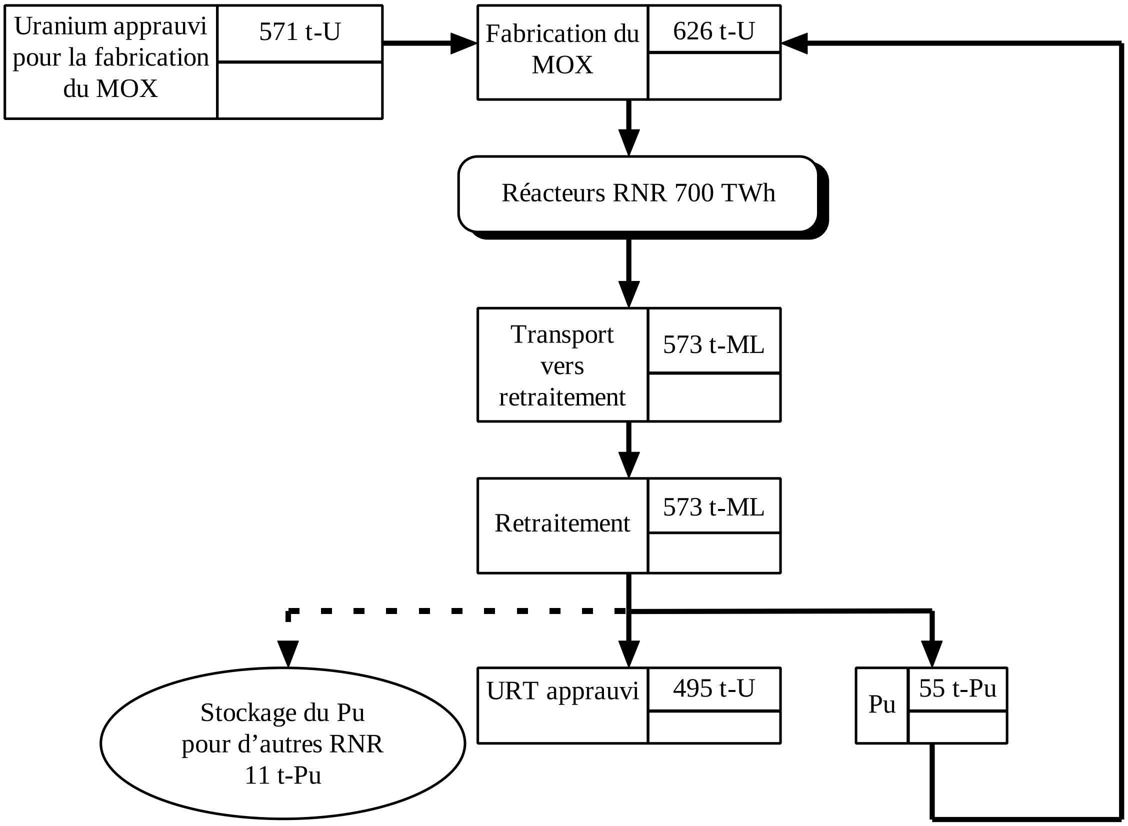


Figure I.5 **Flux de matières théorique pour un parc de RNR de 700 TWh (étude de cas japonaise)**



3.2 Aperçu des cycles du combustible et questions annexes

3.2.1 Cycle ouvert

Le cycle ouvert est la solution prioritaire qu'ont choisie le Canada, l'Espagne, les États-Unis, la Suède et quelques autres pays. Comme les prix de l'uranium sont actuellement assez bas, cette solution produit l'énergie nucléaire la moins chère. Cependant, elle suppose que l'on ne récupère pas les matières fissiles résiduelles (1 pour cent de Pu et 0.8 pour cent d'^{235}U) ni les matières fertiles (^{238}U) qui subsistent dans le combustible usé.

La radiotoxicité potentielle à long terme du combustible nucléaire usé provient essentiellement des actinides qu'il renferme, notamment des nucléides transuraniens (TRU = Pu, Np, Am, Cm…). Ces éléments constituent un important terme source radiologique dans un dépôt de combustible usé, et cela sur de très longues périodes (des centaines de milliers d'années). Cependant, en raison de l'insolubilité intrinsèque des actinides dans les formations géologiques profondes, les transuraniens peuvent être efficacement confinés.

À court terme, les produits de fission constituent le facteur le plus contraignant pour la conception de dépôts parce que le rayonnement γ et la chaleur résiduelle qu'ils produisent augmentent proportionnellement avec le taux de combustion. Après 300 à 500 ans, la plupart des produits de fission se sont désintégrés à l'exception de quelques nucléides à vie longue (^{135}Cs, ^{99}Tc, ^{129}I, ^{93}Zr…). Certains d'entre eux sont assez mobiles dans la géosphère et peuvent contribuer à l'exposition de l'homme.

On peut limiter l'impact radiologique à long terme du cycle ouvert en prévoyant un système de barrières ouvragées et de barrières naturelles qui devraient protéger l'homme des déchets le temps que durera le terme source radiologique relatif à ces déchets. En raison des périodes très longues que cela représente, il convient d'analyser soigneusement les processus en jeu ainsi que les conséquences à long terme de tous les scénarios concevables.

À l'heure actuelle, il n'existe pas de consensus quant à la durée de confinement des déchets de haute activité dans des dépôts en formations géologiques. On s'est fixé comme objectif des périodes de 1 000, 10 000, 100 000 années, voire des durées plus longues, mais la communauté internationale n'a pu jusqu'à présent s'entendre sur une période de confinement donnée.

Le cycle ouvert comporte quelques spécificités du point de vue de la réglementation et de la sûreté :

- Le risque de criticité doit être pris en compte dans la procédure d'autorisation d'un dépôt de combustible usé en raison des quantités importantes de matières fissiles que l'on prévoit d'y stocker.

- La chaleur que dégage le combustible usé au-delà de 500 ans du fait de la présence des actinides est nettement supérieure à celle des seuls produits de fission et, de ce fait, doit être prise en compte dans la conception du dépôt. Cette producteur de chaleur à long terme est toutefois, en valeur absolue, nettement inférieure à celle des produits de fission chargés dans le réacteur.

- Aux États-Unis, la quantité maximale de combustible usé admise dans un dépôt de stockage est limitée à 70 000 tonnes de métal lourd. De ce fait, un grand pays disposant d'un parc nucléaire important, comme les États-Unis, aura besoin de créer un nouveau dépôt à peu près tous les 30 ans.

3.2.2 Cycle du combustible avec retraitement (CCF) et recyclage de l'uranium et du plutonium

Étant donné que l'uranium naturel ne contient que 0.72 pour cent de l'isotope fissile ^{235}U, le recyclage de l'uranium et du plutonium contenus dans le combustible usé a été considéré depuis l'aube de l'ère nucléaire comme la solution de référence. Ces dernières années, de nombreux pays de l'OCDE ont manifesté davantage de réticence à son égard pour des raisons économiques mais aussi par crainte de prolifération.

Ce cycle CCR permet d'extraire un très forte proportion (99.9 pour cent) des flux d'uranium et de plutonium et de ne rejeter qu'un pourcentage minime des actinides majeurs dans les déchets de haute activité liquides (et par conséquent, les déchets de haute activité) et, pour finir, dans le dépôt en formation géologique.

La séparation des actinides mineurs des déchets de haute activité liquides, envisagée comme un moyen de réduire encore la radiotoxicité résiduelle des déchets de haute activité, fait l'objet d'études depuis les années 70. À l'origine, l'objectif des travaux de recherche et de développement était d'éliminer totalement les actinides mineurs pour éviter d'avoir à stocker les déchets à long terme et à les enfouir dans des formations géologiques profondes. Cette option, irréalisable, a été abandonnée.

Cependant, au cas où le public ou les instances politiques refuseraient la solution du stockage à très long terme des déchets de haute activité, l'élimination des actinides mineurs des déchets de haute activité liquides permettrait d'atténuer la radiotoxicité résiduelle des déchets de haute activité. Il faut savoir néanmoins que l'augmentation du taux de combustion s'accompagne d'une production plus importante d'actinides mineurs. Dans ce cas, la solution qui s'impose de prime abord consisterait à ajouter un module de séparation des actinides mineurs aux usines de retraitement classiques. Les pays qui disposent actuellement d'une infrastructure de retraitement (France, Royaume-Uni, Japon, Inde, Russie et Chine) ainsi que leurs partenaires pourraient, à moyen terme, adopter un procédé de séparation partiel permettant d'eliminer des déchets de haute activité de la quasi-totalité des transuraniens à vie longue.

Se pose alors néanmoins la question de savoir quoi faire de l'uranium, du plutonium et des actinides mineurs ainsi récupérés. Les pays qui ont choisi de retraiter le combustible usé étaient guidés par le souci de récupérer les actinides mineurs (uranium et plutonium) afin de réduire leurs achats d'uranium neuf (de 20 pour cent) et d'utiliser les composants fissiles résiduels que contient le combustible usé (environ 1 pour cent d'^{235}U et 1 pour cent de Pu) et qui représentent environ 25 pour cent des coût standard d'UTS pour l'étape de l'enrichissement de l'uranium.

Pendant quelques décennies, on a envisagé de recycler le plutonium dans des réacteurs rapides, mais, pour des raisons tant économiques que politiques, la mise en place de cette option à long terme a pris du retard ou même été abandonnée. En effet, avec la chute des prix de l'uranium, le stock de plutonium constitué dans les usines de retraitement et qui devait initialement être utilisé dans des surgénérateurs rapides à métal liquide est devenu superflu.

Le recyclage dans des réacteurs thermiques du plutonium issu des opérations de retraitement a pris une dimension industrielle il y a plusieurs années. En Europe occidentale, certains pays comme la France, l'Allemagne, la Suisse et la Belgique ont en effet introduit le combustible REO-MOX dans le cycle du combustible avec retraitement et transforment des quantités industrielles de PuO_2 en combustible MOX pour REO qui est ensuite irradié dans des réacteurs bénéficiant d'une autorisation spéciale. Dans une certaine mesure, le recyclage du plutonium est une première étape sur la voie d'un scénario de séparation-transmutation s'inscrivant dans une perspective plus vaste de recyclage des ressources et de réduction des déchets à vie longue produits par les programmes nucléaires.

Ce recyclage du plutonium sous forme de MOX dans les REO est a posteriori une solution qui permet de compenser la dépense que représente le retraitement en produisant de l'électricité dans un deuxième cœur de réacteur. Du point de vue de la radiotoxicité des déchets, le gain réalisé est plutôt maigre étant donné que seulement 25 pour cent du plutonium recyclé seulement est fissionné et que près de 10 pour cent de ce plutonium se transforment en actinides mineurs radiotoxiques à long terme. À l'heure actuelle, le recyclage du combustible usé des REO sous forme de MOX permet une réduction globale en masse (d'un facteur 5 environ) de ce terme source radiotoxique, mais il n'a pas d'effet significatif sur l'inventaire radiotoxique total. Il est théoriquement possible de procéder à un recyclage multiple du MOX dans les REO, mais, dans ce cas, la radiotoxicité résultante augmente à chaque nouvelle campagne de recyclage.

Par conséquent, au cas où l'on serait obligé de réduire davantage l'inventaire radiotoxique global, il faudrait faire passer la fraction de transuraniens que contient le combustible REO-MOX dans un cycle du combustible de la génération suivante à base de RNR-MOX, à savoir le cycle avancé. D'ici là, la solution la plus efficace consisterait donc à recourir au stockage réversible du combustible REO-MOX le temps que la technologie d'incinération en réacteurs à neutrons rapides soit utilisable à l'échelle industrielle. Dans les usines de retraitement actuelles, il est théoriquement possible de retraiter le combustible REO-MOX usé en prévision du recyclage des transuraniens dans des RNR électrogènes.

Pour abaisser dans de fortes proportions la radiotoxicité des déchets liquides de haute activité issus du retraitement du combustible REO-MOX par rapport à celle du même combustible usé mais non retraité, on aura besoin d'un module supplémentaire de séparation des transuraniens. La technologie proposée pour traiter les déchets de haute activité liquides issus du retraitement du combustible REO-UO_2 serait appropriée.

Dès le lancement des programmes nucléaires, il était clair que l'on chercherait à récupérer l'uranium dans le combustible usé, ce qu'a permis le procédé PUREX à base de TBP. Pendant de nombreuses années, on a ainsi récupéré l'uranium au cours du retraitement, mais une très petite proportion du stock ainsi constitué a été recyclée dans des réacteurs. L'uranium de retraitement contient en effet quelques radioisotopes gênants comme l'^{232}U, dont la décroissance naturelle donne des produits radiotoxiques, ou l'^{236}U qui, lorsqu'il est irradié, forme du ^{237}Np.

3.2.3 Cycle du combustible avancé avec recyclage des transuraniens

Tout cycle du combustible avancé exige en première étape le retraitement du combustible usé.

On peut décomposer ainsi le cycle du combustible avancé avec séparation et transmutation de la façon suivante :

- Retraitement du combustible REO-UO_2.
- Séparation des actinides mineurs des déchets de haute activité liquides produits lors du retraitement du REO-UO_2.
- Fabrication des cibles à base d'actinides mineurs en prévision de leur irradiation hétérogène dans des REO.
- Recyclage quantitatif de l'uranium et du plutonium dans le combustible REO-MOX (monorecyclage ou multirecyclage).
- Retraitement du combustible REO-MOX usé dans des installations adaptées (inventaire en plutonium plus important).
- Séparation des actinides mineurs des déchets de haute activité liquides et conditionnement des différents éléments (Np, Am, Cm).
- Fabrication de combustible pour RNR (MOX, métal ou nitrure) à faible teneur en actinides mineurs.
- Irradiation du combustible RNR dans des réacteurs incinérateurs rapides ou dans des installations hybrides conçues et dédiées à ce but (très haut taux de combustion).

- Retraitement du combustible RNR usé dans des installations spécialement conçues et bénéficiant d'autorisations adéquates (procédés aqueux et non aqueux).
- Séparation quantitative de tous les transuraniens issus du retraitement du combustible RNR usé lors des recyclages multiples.
- Multirecyclage du combustible RNR-MOX à forte teneur en transuraniens jusqu'à ce que la proportion consommée soit suffisante.
- Séparation de certains produits de fission de périodes longues, si nécessaire pour le stockage.
- Révision de la stratégie de gestion des produits de fission : séparation du ^{99}Tc (opérations de tête supplémentaires, déchets de haute activité liquides).
- Séparation et récupération des métaux nobles (platinoides).

Nous allons commenter ci-dessous les étapes les plus importantes.

Dans un cycle du combustible avancé dont la finalité est de réduire le plus possible l'inventaire radiotoxique à long terme, la séparation et la transmutation ont un rôle essentiel à jouer. Cependant, le succès de la mise en œuvre de la voie de séparation-transmutation dans ce cycle demande des modifications des installations actuelles ou de la conception des usines de retraitement futures qui intégreront des étapes de séparation.

3.2.3.1 Impact du retraitement du combustible REO-UO$_2$

Le rendement de séparation actuellement obtenu (99.9 pour cent) pour les actinides majeurs suffit, dans une première étape, à abaisser leur teneur dans les déchets de haute activité liquides à un niveau acceptable. La seule amélioration qui pourrait avoir des répercussions importantes sur le risque et sur la gestion des déchets à long terme consisterait à éliminer les déchets de moyenne activité, car ils ne sont pas vitrifiés avant d'être stockés et représentent un volume considérable par rapport aux déchets de haute activité.

3.2.3.2 Séparation des actinides mineurs des déchets liquides de haute activité provenant du combustible usé de REO

Pendant le retraitement, la plupart des actinides mineurs se retrouvent dans les déchets de haute activité. C'est ainsi que l'américium et le curium (avec des transuraniens à vie plus courte : Bk, Cf…) sont quantitativement transférés (> 99.5 pour cent) dans les déchets liquides de haute activité. Le neptunium part directement ou indirectement dans les déchets liquides de haute activité, à l'exception d'une fraction (généralement petite) que l'on retrouve dans le flux de plutonium. La séparation du neptunium exige des étapes d'extraction chimique supplémentaires si l'on veut opérer son transfert quantitatif vers un flux unique de déchets ou de produits pour ensuite procéder à de nouvelles séparations.

Avec le procédé PUREX, il est techniquement possible de récupérer le ^{237}Np du flux d'uranium et/ou de plutonium. Il est également indispensable au préalable de séparer l'^{241}Am si l'on veut réduire dans des proportions significatives la radiotoxicité à long terme du ^{237}Np.

De nombreux laboratoires dans le monde entier (Japon, France, Chine et quelques autres pays au parc nucléaire moins important) s'intéressent à l'heure actuelle à la séparation de tous les actinides mineurs des déchets de haute activité liquides, comme l'on fait auparavant les laboratoires nationaux américains (ANL, ONRL, Hanford…). Des études théoriques et des essais en laboratoire chaud ont été effectués selon plusieurs procédés dont les plus importants sont les procédés TRUEX, DIDPA, TRPO, DIAMEX, et CYANEX 301™. Le Japon, de son côté, étudie la séparation en milieu non aqueux.

Si l'on considère la radiotoxicité des éléments, la séparation de l'américium (et du curium) des déchets liquides de haute activité arrive en priorité. Toutefois, la fraction Am (Cm) contient également les éléments du groupe de terres rares qui, suivant le taux de combustion, peuvent représenter des quantités 10 à 20 fois plus importantes que les actinides. Pour un taux de combustion de 45 GWj/t ML, le rapport est de 16 (13.9 kg de terres rares contre 0.870 kg d'Am-Cm). Par conséquent, pour obtenir une fraction de transuraniens qui soit pure à 90 pour cent, il faut que le facteur de décontamination des terres rares dépasse 100. Une pureté de 99 pour cent suppose un facteur de séparation supérieur à 1 000, qui constitue la limite technologique pour des éléments tels que les lanthanides et les actinides dont les propriétés chimiques sont très proches. Les récents progrès des techniques de séparation chimique laissent cependant entrevoir la possibilité de résoudre ce problème.

Adapter les flux annuels de matières et traduire ces adaptations en réalités techniques représente un travail extrêmement complexe qui dépasse le cadre de ce rapport. À l'avenir, les usines de retraitement pourraient bien comporter, dès leur conception, des chaînes de séparation des actinides et des produits de fission à vie longue.

3.2.3.3 Fabrication des cibles d'actinides mineurs pour recyclage hétérogène dans des REO

À moyen terme, l'irradiation des combustibles ou cibles contenant des actinides mineurs ne pourra se faire que dans les réacteurs thermiques, notamment dans les REO. Aujourd'hui, il est difficile de fabriquer des cibles d'irradiation avec des quantités d'actinides mineurs qui soient représentatives d'une production industrielle, et cela même dans des cellules chaudes à l'échelle du pilote.

On possède une certaine expérience de la production de sources de chaleur isotopiques, mais le contexte radiologique actuel, de même que les limites de dose que devraient imposer les organismes de réglementation et contrôle, ont créé pour le recyclage industriel des actinides mineurs une situation très différente de celle qui a prévalu par le passé pour les applications militaires et spatiales.

Étant donné la présence d'importantes quantités d'^{241}Am, auxquelles viennent s'ajouter 1 à 10 pour cent de terres rares, on aura besoin de chaînes de fabrication blindées contre le rayonnement γ ainsi que de systèmes de télémanipulation. La présence de 5 pour cent de ^{244}Cm dans une cible d'^{241}Am et d'^{243}Am exigera des protections neutroniques supplémentaires à cause du taux de fission spontanée et de la réaction α-n caractéristique des cibles isotopiques de type oxyde, ce qui compliquera encore la mise en œuvre du procédé.

3.2.3.4 Séparation des produits de fission et d'activation à vie longue

Plusieurs produits de fission ou d'activation importants d'un point de vue radiologique peuvent avoir des répercussions significatives sur le comportement des dépôts en formations géologiques, de sorte qu'il a été envisagé de les séparer et de les transmuter. L'évaluation doit porter notamment sur les nucléides suivants : produits de fission : ^{99}Tc, ^{129}I, ^{135}Cs, ^{79}Se, ^{93}Zr et ^{126}Sn ; produits d'activation : ^{14}C et ^{36}Cl.

Le technétium-99 (^{99}TC) est un produit de fission dont la période radioactive est de 213 000 ans et que l'on rencontre sous forme de métal et de TcO_2 dans les résidus insolubles et d'ion pertechnétate dans les solutions liquides de déchets de haute activité. Dans du combustible irradié à un taux de 45 GWj/t ML, son activité spécifique est de 6.10^{11} Bq/t ML. Sa radiotoxicité se caractérise par une limite annuelle d'incorporation par ingestion (LAI) de 3.10^{7}Bq. Pour résoudre efficacement le problème de sa radiotoxicité à long terme, il faut combiner les deux fractions avant de transmuter cet élément. Il est relativement facile d'extraire le technétium soluble. En revanche, sa similitude avec les métaux nobles platinoides et la nature des méthodes de séparation (techniques pyrochimiques) rendent très délicate sa séparation lorsqu'il se trouve dans des déchets insolubles. De plus, le technétium ne présente un danger radiotoxicologique que lorsqu'il se trouve dans des conditions géologiques oxydantes (Tuff, Yucca Mountains). Dans les aquifères réducteurs à grande profondeur, la migration du ^{99}Tc est négligeable.

L'iode-129 (^{129}I) est séparé des déchets liquides de haute activité au cours des procédés de retraitement classiques. On le retrouve dans les effluents de lavage du dissolveur. Son activité spécifique dans le combustible est de $1.6.10^{9}$ Bq/t ML et sa LAI de 1.10^{5} Bq. La fraction séparée peut alors être soit stockée sur un adsorbant spécifique ou rejetée dans les océans. Cet élément, qui a une période radioactive de 16 millions d'années, a toutes les chances d'être dispersé dans la géosphère ou la biosphère. Sa forte radiotoxicité le place parmi les nucléides importants lorsque l'on envisage l'installation de dépôts terrestres de combustible usé. En revanche, s'il était dispersé dans la géosphère ou la biosphère, sa radiotoxicité est assez limitée.

Le sélénium-79 (^{79}Se) est un produit de fission dont la période est de 65 000 ans et que l'on trouve dans les déchets de haute activité. Ce nucléide, qui se comporte chimiquement comme un sulfate, sera incorporé aux déchets vitrifiés. Son activité spécifique dans le combustible usé devrait se situer autour de 2.10^{10} Bq/t ML, et sa LAI est de 10^{7} Bq/an. Étant donné sa concentration chimique très faible dans les déchets liquides de haute activité, par rapport aux composés naturels du soufre, sa séparation ne paraît pas évidente.

Le zirconium-93 (^{93}Z) et le césium-135 (^{135}Cs) sont deux nucléides à vie longue (de périodes radioactives égales à 1.5 et 2 millions d'années respectivement) que l'on retrouve dans le combustible usé. Il est pratiquement exclu de séparer ces radionucléides des autres produits de fission pour les transmuter, parce qu'ils sont accompagnés d'autres radioisotopes très radioactifs (^{137}Cs) ou les surpassant en quantité (0.73 ~1 kg de ^{93}Zr et 3.3 ~ 5.0 kg de Zr par t ML). Pour en réduire efficacement le potentiel radiotoxique par bombardement neutronique, il faudrait prévoir, avant la fabrication de la cible, toute une série de procédés de séparation isotopique, stratégie que l'on considère à l'heure actuelle comme pratiquement impossible pour des raisons économiques.

L'étain-126 (^{126}Sn) a une période radioactive de 100 000 ans. Il est en partie soluble dans les déchets liquides de haute activité, mais on en trouve aussi une partie dans les résidus insolubles. Sa concentration dans les déchets de haute activité se situe aux alentours de 4.10^{10} Bq/t ML et sa LAI est de 3.6 Bq/t ML. L'espèce radioactive ^{126}Sn s'accompagne d'une série d'isotopes stables (^{116}Sn, ^{118}Sn, ^{119}Sn, ^{120}Sn, ^{122}Sn, ^{123}Sn et ^{124}Sn), ce qui rend sa transmutation très problématique.

Le carbone-14 (^{14}C), dont la période radioactive est de 5 730 ans, constitue un cas difficile. Étant soluble dans les eaux souterraines, il est susceptible de pénétrer dans la biosphère et présente un risque radiotoxicologique important du fait qu'on le retrouve dans le cycle biochimique. Selon la concentration de l'azote dans le combustible UO_2 d'origine, la concentration du ^{14}C dans le combustible usé devrait représenter environ 3.10^{10} Bq/t ML et sa LAI 4.10^{7} Bq/t ML. Sa contribution à la radiotoxicité à long terme dépendra des conditions physico-chimiques qui prévaudront dans les aquifères souterrains à grandes profondeur ou dans des géosphères non saturées en eau. Sa section efficace de capture dans un spectre neutronique thermique est très faible.

Le chlore-36 (^{36}Cl). La gaine en zircaloy et le combustible UO_2 contiennent du chlore naturel à l'état d'impureté à raison de 5 à 20 ppm. Au cours de l'irradiation, ce ^{35}Cl se transmute en ^{36}Cl dont la période radioactive est de 300 000 ans. Une partie de ce produit d'activation se retrouve dans les effluents liquides du dissolveur, l'autre partie restant dans les coques en zircaloy lavées. Pour un taux de combustion de 45 GWj/t ML, on évalue l'activité de cet élément présent dans les déchets de haute et de moyenne activité à 2.10^{6} Bq. En raison de ses propriétés chimiques, ce nucléide se dissout facilement dans les eaux souterraines et pourrait, par conséquent, contaminer les réservoirs d'eau au voisinage des dépôts. La LAI par ingestion est de 2.10^{7} Bq. Ce radionucléide n'est pas prévu dans les scénarios de transmutation, étant donné que la contamination des concentrés de déchets par le ^{35}Cl naturel produirait des quantités supplémentaires de ^{36}Cl.

Une analyse approfondie de certains radionucléides évoqués dans cette section serait justifiée, afin d'établir les risques qu'ils présentent et leur contribution à la radiotoxicité potentielle par rapport aux transuraniens. En effet, alors qu'ils ne présentent qu'une radiotoxicité de 1 000 à 10 000 fois moins importante que celle des transuraniens, leur contribution au risque à long terme est prédominante, car ils sont susceptibles de migrer beaucoup plus vite dans la biosphère et de provoquer, à très long terme, l'exposition de l'homme à une dose de rayonnement non négligeable.

3.2.3.5 Recyclage quantitatif de l'uranium et du plutonium dans le combustible REO-MOX

Certains pays recyclent sous forme de REO-MOX le plutonium séparé lors du retraitement du combustible REO-UO_2. Il faut savoir cependant que l'irradiation du REO-MOX accroît l'inventaire radiotoxique du combustible, car elle produit davantage d'actinides mineurs. De ce fait, le multirecyclage du plutonium dans les REO-MOX n'est pas efficace.

On s'intéresse depuis peu aux possibilités d'utiliser le plutonium séparé pour en faire du combustible destiné à des RNR dans lesquels « l'incinération » d'isotopes aussi bien impairs que pairs, donne lieu à un taux de fission plus élevé par rapport à celui que l'on obtient dans les REO. Seule la transmutation dans des réacteurs à spectre rapide (RNR ou systèmes hybrides) permet de réduire effectivement l'inventaire radiotoxique des matières nucléaires.

Le recyclage de l'uranium de retraitement dans le combustible des REO se fait à l'échelle industrielle, mais il suppose que l'on surenrichisse en matière fissile le combustible neuf. D'un point de vue purement radiotoxique, l'accumulation d'uranium appauvri ou d'uranium de retraitement a un impact plus important que celle du neptunium par exemple.

3.2.3.6 Retraitement du REO-MOX

Jusqu'à présent, le retraitement du REO-MOX s'effectuait le plus souvent en diluant ce combustible avec du REO-UO_2 en respectant les proportions existantes dans le cœur du réacteur (UO_2/MOX = 2). Dans son usine UP2 de La Hague, la Cogéma a démontré, en 1992 lors d'une campagne spéciale (environ 5 tonnes), qu'il était possible de retraiter le REO-MOX sans le diluer avec du combustible UO_2. Cette opération pourrait être réalisée à l'échelle commerciale, à condition que les usines de retraitement reçoivent l'autorisation de traiter des concentrations plus élevées en plutonium ainsi qu'un inventaire total de plutonium nettement plus important.

L'inventaire radiotoxique du combustible REO-MOX usé représente environ huit fois celui du REO-UO_2. Par le procédé de retraitement classique, on pourra extraire l'uranium et le plutonium qui représentent environ 30 pour cent de l'activité α totale, et les 70 pour cent se retrouveront avec les déchets de haute activité liquides constitués de neptunium, d'américium et de curium. Mais les isotopes du curium et de l'américium sont les principaux responsables de cette radioactivité α.

Si l'on prévoit une étape de séparation et de transmutation, il est indispensable d'éliminer au préalable les transuraniens des déchets de haute activité liquides avant de les vitrifier. Les techniques que l'on utilisera sont en principe identiques à celles adoptées pour le REO-UO_2 (voir 3.2.3.2 Séparation des actinides mineurs des déchets liquides de haute activité provenant du combustible usé de REO). Cependant, les procédés d'extraction sont plus compliqués par l'activité α élevé qui a pour effet d'augmenter le dommage d'irradiation.

Il est possible également de stocker le REO-MOX usé pendant 50 ans ou plus de façon à laisser décroître le ^{244}Cm (période : 18 ans) en ^{240}Pu avant de passer au retraitement. Les procédés d'extraction chimique sont beaucoup plus faciles à réaliser après une période de « refroidissement » prolongée qui permet de réduire d'un facteur 7, voire plus, la chaleur de décroissance α, suivant la composition isotopique. Le multirecyclage du REO-MOX est envisageable si l'on dispose d'une quantité suffisante de plutonium frais issu du retraitement de REO-UO_2 irradié à un taux modéré.

3.2.3.7 Fabrication de RNR-MOX à faible teneur en actinides mineurs

Grâce aux programmes de construction de RNR lancés il y a plusieurs décennies par de nombreux pays nucléaires, on possède aujourd'hui une solide expérience de la fabrication industrielle du combustible RNR-MOX [11]. Le RNR-MOX à 15-25 pour cent de plutonium est aujourd'hui couramment fabriqué à des fins commerciales. Toutefois, jusqu'à présent, le plutonium utilisé de cette manière provenait de combustible UO_2 irradié à des taux peu élevés et contenant de faibles teneurs en ^{238}Pu et en ^{241}Pu.

Depuis, le taux de combustion du REO-UO_2 et du REO-MOX est passé à 50 GWj/t ML. La composition isotopique du plutonium obtenue à l'issue du retraitement de ces combustibles est nettement dégradée et se caractérise par de fortes concentrations de ^{238}Pu et de ^{242}Pu et, au contraire, de faibles teneurs en ^{239}Pu et ^{241}Pu. En 1989, l'AEN a publié une étude approfondie des aspects économiques du recyclage du plutonium; qui reste aujourd'hui une référence généralement valable [12].

Dans l'hypothèse où l'on utiliserait des réacteurs incinérateurs rapides avancés (CAPRA), on envisage des concentrations de plutonium encore supérieures (jusqu'à 45 pour cent). Le recyclage des combustibles contenant d'aussi fortes proportions de ^{238}Pu et de petites quantités d'actinides mineurs est encore plus délicat et exige la conception et la construction d'usines équipées de systèmes de télémanipulation.

Pour le recyclage homogène des actinides mineurs dans le RNR-MOX, on étudie actuellement la possibilité d'ajouter 2.5 pour cent de ^{237}Np et/ou d'^{241}Am. Le ^{237}Np, un émetteur α, a une faible activité spécifique et ne pose pas de problème majeur de manipulation. Néanmoins, l'addition de 2.5 pour cent d'^{241}Am créera un champ γ autour des boîtes à gants et des cellules chaudes. Cependant, le nucléide le plus gênant est le ^{238}Pu à une concentration de 3 pour cent, car il constitue une source de chaleur et de neutrons (5 kWth/t ML ; $5x10^{8}$ neutrons/s par t ML).

Les conditions de fabrication du combustible RNR-MOX contenant de petites quantités d'actinides mineurs dépendront du degré de séparation des terres rares (puissants émetteurs γ) et, et de son contenu en ^{244}Cm qui est accompagne de l'^{241}Am et l'^{243}Am séparés des déchets liquides de haute activité. La présence de petites quantités de ^{244}Cm intensifiera l'émission de neutrons par le combustible RNR-MOX ainsi produit.

Les taux de séparation des terres rares et du ^{244}Cm qu'il faudra atteindre pour pouvoir envisager la fabrication industrielle du combustible dépendront pour une bonne part de la concentration de terres rares que pourra supporter le combustible RNR-MOX frais et de la concentration admissible de ^{244}Cm lors de la fabrication du combustible.

Le recyclage hétérogène des actinides mineurs permet de s'affranchir de l'obligation de diluer les nucléides gênants pendant la fabrication du combustible (par exemple, le ^{244}Cm) et de réaliser cette opération dans de petites installations spécialement conçues pour ces opérations et pourvues des blindages nécessaires.

3.2.3.8 Fabrication de combustibles métalliques pour les ALMR et des combustibles avancés pour les réacteurs incinérateurs

Dans le cadre des travaux sur l'Integral Fast Reactor, une technique de fabrication du combustible a été mise au point et testée à l'échelle du pilote froid (et chaud). Dans l'installation EBR-II, le combustible métallique a été recyclé par fusion d'un alliage U-Pu-Zr à l'échelle du laboratoire et du pilote chaud. Bien sûr, ces procédés en sont encore au stade exploratoire et ne peuvent par conséquent pas être considérés comme des technologies éprouvées. Toutefois, il convient d'en étudier les possibilités étant donné que les combustibles métalliques permettent d'atteindre des taux de combustion très élevés et présentent des propriétés physiques et neutroniques adaptées à la transmutation des transuraniens.

On s'est intéressé très récemment aux possibilités d'utiliser des combustibles nitrure et carbure dans des réacteurs incinérateurs rapides. Il est possible de produire ces combustibles à base de nitrures de transuraniens contenant des quantités macroscopiques d'actinides mineurs en combinant une méthode de gelification interne avec une synthèse carbothermique. Ces combustibles nitrures peuvent être ensuite retraités par des procédés électrolytiques, de la même manière que les combustibles métalliques.

3.2.3.9 Réacteurs incinérateurs rapides

Les études et recherches réalisées pendant 30 ans dans le monde entier sur les réacteurs surgénérateurs rapides (surgénérateurs à métal liquide) nous ont permis d'acquérir une expérience technologique considérable. Cette expérience peut être transposée aux réacteurs incinérateurs rapides. On trouvera à la référence [11] les explications concernant le combustible.

En France, le CEA a lancé, en 1992, le programme de recherche CAPRA consacré à la consommation accrue de plutonium dans les réacteurs incinérateurs rapides, effet qui a été obtenu en augmentant la concentration relative de plutonium dans le combustible. Le programme SPIN sur l'incinération des actinides a été entrepris en parallèle. Actuellement, les recherches s'orientent vers des combustibles à base de transuraniens et sans uranium comportant des matrices inertes afin de réduire le plus possible la formation de transuraniens.

L'autorisation de convertir progressivement le surgénérateur Superphénix en incinérateur de plutonium a été délivrée en 1994 à la suite d'une recommandation d'une commission gouvernementale [11]. À cet effet, on a fabriqué des assemblages équipés de réflecteurs en acier afin de remplacer la couverture fertile radiale. Trois assemblages d'essai ont été fabriqués ainsi : deux assemblages CAPRA contenant du plutonium d'origines différentes, à savoir de première ou de deuxième génération, ainsi qu'un assemblage NACRE dans lequel étaient ajoutés au MOX standard 2 pour cent de neptunium conformément aux résultats du programme SPIN. Après avoir fonctionné en puissance sans problèmes en 1996, Superphénix a été préparé en prévision du changement de cœur. Néanmoins, le gouvernement français a décidé, en 1997, qu'il fallait arrêter définitivement Superphénix, décision qu'il a confirmée en février 1998.

Cette décision implique une importante réorganisation du programme de réacteurs rapides. S'agissant des études relatives à la consommation accrue des transuraniens, il est question de réaliser sur Phénix une partie des expériences initialement prévues sur Superphénix en tenant compte du fait que Phénix a une puissance nettement inférieure et qu'il sera définitivement mis à l'arrêt en 2004. Dans ce nouveau contexte, il a été décidé de sélectionner les expériences pour satisfaire à la plupart des exigences de la Loi française du 2 décembre 1991 concernant la recherche sur les déchets nucléaires.

Le gouvernement japonais a lancé, en 1988, de nouveaux projets de recherche sur la transmutation, dans le cadre de son programme OMEGA, et procède actuellement à leur extension. Loin de se limiter au combustible RNR-MOX, les travaux englobent aujourd'hui des combustibles à base de nitrures et de nitrures-carbures, notamment.

Le projet IFR de l'ANL a évolué peu à peu vers la conception d'un prototype industriel de RNR refroidi au sodium, le réacteur PRISM, qui utiliserait du combustible métallique (UPuZr). Ce réacteur serait associé à des usines de retraitement pyrochimique pour recycler le combustible à base de transuraniens et éliminer les produits de fission. Des études de conception et des analyses des performances du cœur sont en cours. L'objectif est d'augmenter le taux d'incinération des transuraniens.

3.2.3.10 Retraitement du combustible usé de RNR

Il ne sera pas possible de réduire l'inventaire radiotoxique résultant du multirecyclage du combustible RNR-MOX ou RNR-métal que si on intègre au cycle du combustible avancé le multi-recyclage du combustible RNR usé. Les combustibles RNR-MOX ou RNR-métal sont en effet irradiés à des taux très élevés.

On envisage aujourd'hui d'irradier le combustible dans les RNR à des taux doubles ou triples de ceux appliqués au MOX dans les REO. Ces taux s'échelonneraient de 120 à 180 GWj/t ML, voire plus ultérieurement. Or, malgré ces très hauts taux de combustion, le rendement de transmutation ou d'incinération reste assez faible.

Après une campagne d'irradiation prolongée, par exemple de cinq ans à un taux de combustion de 150 GWj/t ML, on n'a consommé que 15 à 20 pour cent de l'inventaire initial total des transuraniens. Pour obtenir un taux de consommation total de 90 pour cent des transuraniens, il faudrait recycler le combustible usé de RNR de nombreuses fois.

En raison de la chaleur de décroissance, il n'est pas possible de retraiter le combustible usé des RNR immédiatement après son déchargement des réacteurs rapides, que ce soit par le procédé PUREX ou par un procédé d'extraction aqueux équivalent. Il faut, en effet, prévoir d'augmenter de 7 à 10 ans, voire à 12 ans, les temps de refroidissement entre le déchargement et le retraitement. Cela porte, par conséquent, la durée totale du cycle avancé à 15 ou 17 ans. On peut, par conséquent, considérer qu'il faudra attendre 100 à 250 ans avant d'obtenir un taux de consommation global de 90 pour cent.

Le retraitement pyrochimique du combustible usé des RNR a été mis au point dans le cadre du projet IFR. Dans ce cas, le combustible métallique irradié dans les RNR à des taux très élevés est transféré sur place à une cellule chaude pyrochimique pour y être retraité. Comme le bain de sels fondus ($CdCl_2$, LiCl, KCl…) n'est pas dégradé par le rayonnement α, il est possible d'envisager des temps de refroidissement beaucoup plus courts. Toutefois, cette technologie de retraitement, qui fait ses tout premiers pas, ne pourra parvenir à une maturité industrielle dans le secteur nucléaire civil qu'après d'importants travaux de recherche et de développement.

Ce procédé repose sur l'affinage électrolytique du combustible métallique usé à l'aide d'une anode, de cathodes, solide et liquide, en cadmium et d'un électrolyte à base de sels fondus (LiCl+KCl) porté à 500°C. Il s'agit ainsi de provoquer la dissolution pyrochimique du combustible usé débarrassé au préalable de sa gaine par des techniques mécaniques.

L'uranium pur (débarrassé du plutonium) est transporté par voie électrolytique à la cathode solide en cadmium. Le mélange de plutonium et d'actinides mineurs contenant des traces d'uranium est transporté vers une seconde cathode en cadmium, liquide cette fois, et séparé de l'ensemble des produits de fission. La fraction plutonium plus actinides mineurs est recyclée sous forme métallique et fondue dans les nouvelles aiguilles combustibles.

Le CRIEPI, au Japon, met au point des procédés pyrochimiques analogues qui pourraient, à la longue, venir remplacer le procédé de retraitement par voie aqueuse (PUREX) pour des combustibles nitrures ou métalliques de RNR fortement irradiés.

Le principal intérêt des procédés de retraitement pyrochimiques tient au fait qu'ils sont insensibles au taux d'irradiation du combustible et peu sujets aux excursions de criticité pendant les opérations. En abaissant à 6 ou 7 ans la durée du cycle avancé, on diminue de moitié le temps nécessaire pour obtenir le même taux de consommation qu'avec un cycle avancé utilisant le procédé de retraitement par voie aqueuse. Toutefois, avant que ce type de procédé pyrochimique puisse être sérieusement envisagé pour retraiter le combustible métallique de RNR, il faudra avoir trouvé des solutions réellement convaincantes aux principaux problèmes en suspens : la corrosion, le degré de séparation des transuraniens et la manutention des déchets et des cibles irradiées.

3.2.3.11 Transmutation des produits de fission à vie longue

Il est extrêmement difficile de transmuter les produits de fission à vie longue parce que leurs sections efficaces de capture, sur lesquelles repose la transmutation des nucléides radioactifs en nucléides à vie courte ou stables, sont très faibles. De ce fait, si l'on veut parvenir à un taux de transformation important, il faut prévoir des périodes d'irradiation très longues. La seule solution, pour réussir cette entreprise très onéreuse, consistera donc à recourir à des réacteurs dédiés à hauts flux de neutrons thermiques et/ou à des systèmes hybrides dédiés.

Du point de vue de la technologie des réacteurs, l'option la plus facile à réaliser est la transmutation du technétium-99 (^{99}Tc), au cours de laquelle la cible de technétium métal se transforme en ^{100}Ru métallique, un isotope stable du ruthénium. Cependant, la section efficace de la réaction (n-γ) pour les neutrons thermiques n'est que de 20 barns (voir 3.2.3.2), donc faible pour la transmutation. La « période » de transmutation, c'est-à-dire le temps nécessaire pour consommer la moitié de la cible varie d'un réacteur thermique à l'autre, mais, dans le meilleur des cas, atteindrait près de 30 ans. La production simultanée de ^{99}Tc par fission dans le combustible nourricier REO-UO_2 fait baisser le taux de transmutation net et nécessite un chargement important en technétium. Le réacteur doit donc être spécifiquement conçu à cette fin. Il faut aussi prévoir de surenrichir le combustible afin de compenser les pertes dues à la réactivité négative des cibles de technétium dans les assemblages combustibles. Par ailleurs, à l'issue de ces longues périodes de transmutation, les structures en acier des assemblages contenant des produits de fission à vie longue irradiés seront si endommagées sous l'effet des rayonnements que les assemblages devront être retraités.

Neutroniquement parlant, la transmutation de l'^{129}I est très proche de celle du ^{99}Tc. Mais le procédé présente beaucoup plus de difficultés d'ordre chimique et métallurgique, étant donné que la cible est chimiquement instable et que le produit de la réaction de capture des neutrons est un gaz rare, le ^{130}Xe, qu'il faut évacuer de la capsule d'irradiation pendant son séjour en réacteur. Toute excursion de température provoquerait en effet le relâchement de l'^{129}I avec les effluents du réacteur.

Les systèmes hybrides caractérisés par des flux de neutrons thermiques extrêmement forts (10^{16} n/cm^2/s) devraient en principe permettre de raccourcir la période de transmutation. Cette technologie fait actuellement l'objet d'études théoriques. Quoi qu'il en soit, toutes les formes de transmutation par neutrons thermiques coûteront très cher en énergie.

À moins de les séparer des autres isotopes du zirconium et du césium avant de les irradier, les radionucléides ^{93}Zr et ^{135}Cs ne se prêtent pas à la transmutation en réacteur.

Il n'a pas encore été envisagé de transmuter le ^{14}C dans un scénario de séparation-transmutation. Théoriquement, il serait possible de le récupérer, dans les effluents gazeux du retraitement, une partie du ^{14}C provenant du combustible usé (environ 50 pour cent). Cependant, on connaît encore trop mal le comportement chimique de cet isotope dans un dissolveur pour pouvoir améliorer ce rendement. Une fois transformé en cible solide, par exemple du carbonate de baryum ($BaCO_3$), cet isotope pourrait être stocké indéfiniment. La section efficace de capture des neutrons thermiques du ^{14}C est proche de zéro. En théorie, on pourrait, dans certains cas, transmuter le ^{14}C à l'aide de particules chargées dans des accélérateurs de haute énergie, mais l'on est en droit de s'interroger sur les possibilités de réalisation et l'intérêt économique de ce type de démarche.

3.2.3.12 Rôle de la séparation-transmutation dans les cycles du combustible avancé

La voie de la séparation-transmutation, intégrée au cycle du combustible avancé comme nous venons de le décrire, représente la démarche la plus complète que l'on puisse proposer et constitue un élargissement important des activités du cycle du combustible par rapport au cycle avec retraitement et, a fortiori, au cycle ouvert.

On peut considérer des opérations de séparation aussi comme une série de procédés isolés, appliqués après un retraitement standard, qui permettraient d'atténuer la radiotoxicité des déchets stockés et d'améliorer les techniques de conditionnement. Cependant, dans ce rapport, nous avons surtout envisagé la séparation comme une étape préalable à la transmutation.

4. ANALYSE CRITIQUE

- Dans l'industrie du cycle du combustible nucléaire de demain, la séparation et la transmutation pourraient soit s'ajouter aux activités du cycle tel que nous connaissons aujourd'hui, soit en remplacer quelques parties.

- La séparation et la transmutation ne peuvent être envisagés sans cycle du combustible classique ou avancé. Toutefois, pendant la phase de recherche et de développement, tous les pays intéressés par l'évolution du cycle du combustible ou la gestion des déchets peuvent participer à l'effort international.

- Toute stratégie de séparation et de transmutation devrait tendre vers l'élimination progressive des radionucléides à vie longue des flux de déchets, dont le mode de stockage reste à décider. Toutefois, la réduction de l'inventaire radiotoxique des déchets que cette solution apporte s'opère au prix d'une légère augmentation de l'inventaire radiotoxique des cœurs de réacteurs et des installations du cycle du combustible, sauf si l'on établit une stratégie exhaustive pour l'incinération des actinides faisant appel, par exemple, à la transmutation dans des réacteurs hybrides.

- Dans un programme nucléaire à long terme, il est possible de passer progressivement du combustible REO-UO_2 au combustible REO-MOX pour ensuite introduire les RNR dans le parc électronucléaire ainsi que les systèmes hybrides.

- La séparation des actinides mineurs est une étape supplémentaire au recyclage de l'uranium et du plutonium dans le combustible MOX et suppose la conception et la construction de nouvelles installations industrielles. À l'heure actuelle, la séparation n'a pas dépassé le stade du développement en laboratoire et en cellules chaudes.

- La séparation ne se conçoit que dans une perspective à long terme. Elle exige des installations de traitement des déchets liquides de haute activité de conception nouvelle ou un cycle d'extraction PUREX avancé qui permettrait de séparer directement tous les radionucléides à vie longue.

- La séparation des actinides mineurs des déchets liquides de haute activité permet de réduire l'inventaire radiotoxique à long terme des déchets de haute activité vitrifiés et, à ce titre, peut ameliorer la perception du danger que présentent le stockage des déchets à vie longue. En fait, elle ne modifie pas les caractéristiques techniques des déchets. Les produits de fission (^{137}Cs et ^{90}Sr) déterminent à court terme la quantité de la chaleur produite dans les déchets de haute activité, et la faible solubilité des transuraniens conditionne leur taux de lixiviation et, partant, leur impact radiologique à long terme.

- La séparation des actinides mineurs (Np, Am, Cm) et de certains produits de fission (Cs, Sr, Tc, etc.) multiplie les possibilités d'améliorer le conditionnement des radionucléides à vie longue en fonction des nucléides. La meilleure stabilité thermodynamique des nouveaux colis de déchets devrait en principe ouvrir de nouvelles perspectives pour le stockage ou la mise en dépôt à long terme des transuraniens par rapport à la vitrification, qui a été spécifiquement conçue pour des mélanges complexes d'actinides mineurs et de produits de fission. On ne saurait trop recommander de poursuivre les recherches sur les méthodes et technologies de séparation si l'on veut disposer de la base technique la plus large possible pour procéder à une évaluation de cette option.

- La transmutation est un terme général qui englobe « l'incinération » d'un nucléide, c'est-à-dire sa transformation en produits de fission, et sa transformation par capture neutronique en un autre radionucléide ou en un isotope stable. La mise au point de cette technologie pourrait avoir d'importantes retombées sur d'autres programmes nucléaires.

- Certaines réactions de transmutation produisent une majorité de nucléides non radioactifs, d'autres une diversité de radionucléides de périodes très variables. Les nucléides ainsi obtenus doivent avoir une période plus courte que celle de la cible ou une radiotoxicité plus faible, voire, dans certains cas, provoquer la production d'un autre nucléide père ayant un impact radiologique moindre.

- Dans un premier temps, la transmutation des actinides mineurs ne se solde pas nécessairement par une réduction de l'inventaire radiotoxique à court terme.

- Dans les réacteurs thermiques, la transmutation s'opère en général par capture neutronique. Le recyclage de quantités industrielles d'actinides mineurs exige de surenrichir le combustible, ce qui a pour effet d'augmenter les coûts de la production nucléaire. Cependant, le recyclage hétérogène des actinides mineurs dans des REO brûlant du MOX pourrait être une solution viable dans la mesure où l'on dispose d'excédents de plutonium permettant de compenser la réactivité négative provoquée par la présence d'actinides mineurs. Cependant, avec ce mode de transmutation, la radiotoxicité globale des matières fissiles et fertiles ne diminue pas de façon significative sur l'ensemble du cycle du combustible.

- Dans les réacteurs à neutrons rapides, la transmutation des actinides mineurs permet de réduire la radiotoxicité dans la mesure où tous les actinides mineurs sont plus ou moins fissiles. Le rendement d'incinération augmente avec l'énergie moyenne des neutrons. Cependant, la sûreté des réacteurs impose une limite aux quantités acceptables d'actinides mineurs et aux rendements de fission et de transmutation résultants. Le potentiel énergétique théorique des actinides mineurs représente à peu près 10 pour cent de celui du plutonium produit dans le combustible UO_2. Pour la transmutation des actinides mineurs, les systèmes à spectre neutronique rapide (RNR, systèmes hybrides) sont plus prometteurs que les REO.

- Il a été envisagé de séparer et de transmuter certains produits de fission à vie longue, dont les plus souvent cités sont l'^{129}I, le ^{135}Cs, le ^{99}Tc et le ^{93}Zr.

- La séparation de certains produits de fission est déjà une réalité industrielle : l'iode est extrait des solutions de combustible usé par barbotage, lavage et traitement séparé avant d'être rejeté dans les océans (pratique interdite dans certains pays). Au cas où il paraîtrait difficile de rejeter encore longtemps l'^{129}I dans les océans, il faudrait réexaminer les possibilités de le récupérer, de le conditionner et éventuellement de le transmuter. Il est impossible de transmuter le ^{135}Cs et le ^{93}Zr sans procéder au préalable à une séparation isotopique.

- Les principes théoriques de la séparation du technétium sont connus, mais n'ont jamais été mis en pratique. Il est possible de transmuter le ^{99}Tc dans des réacteurs thermiques et dans des régions thermalisées de réacteurs rapides. Toutefois, dans un spectre de neutrons thermiques, la période de transmutation est assez longue (de 15 à 77 ans). De ce fait, à moins que l’on ne parvienne à produire des neutrons à un coût marginal dans des installations nucléaires (systèmes hybrides), cette solution ne devrait pas avoir la priorité des travaux de R-D.

5. CONCLUSIONS GÉNÉRALES

- La mise en œuvre de la séparation et de la transmutation passe par des recherches fondamentales de longue haleine et exige des investissements lourds dans des systèmes dédiés produisant des spectres de neutrons rapides (RNR, systèmes hybrides en complément ou à la place des REO exploités) ainsi que la mise au point de nouvelles étapes retraitement et la construction d'usines de fabrication de combustibles et de cibles équipées de télémanipulateurs.

- Les installations de séparation des actinides mineurs et de certains produits de fission à vie longue pourraient être conçues pour être intégrées aux usines de retraitement existantes.

- Des méthodes de séparation des éléments radiotoxiques à vie longue ont été mises au point en laboratoire. Cependant, il reste encore beaucoup à faire pour perfectionner la technologie, passer à l'échelle supérieure, et ainsi la rendre compatible avec les pratiques de retraitement industrielles.

- Dans le court terme, la séparation réduirait l'inventaire radiotoxique à long terme des déchets de haute activité à stocker au prix d'une augmentation des contraintes d'exploitation des installations nucléaires concernées.

- Pour le recyclage et la transmutation des radionucléides à vie longue, les systèmes à spectres neutroniques rapides (RNR dédiés ou systèmes hybrides) sont plus efficaces que les REO de conception actuelle. En tant qu'incinérateurs d'actinides et de certains produits de fission contenus dans le combustible usé REO-MOX ou RNR-MOX, les systèmes hybrides pourraient largement contribuer à réduire l'inventaire radiotoxique à long terme des déchets.

- Avec le recyclage du plutonium et des actinides mineurs, l'inventaire en transuraniens d'un parc électronucléaire pourrait être stabilisé. Le multirecyclage des transuraniens est une entreprise à long terme qui ne permet d'atteindre l'équilibre qu'au bout de plusieurs décennies.

- Au cours des prochaines décennies, on peut envisager de conditionner les nucléides à vie longue séparés dans des matrices adaptées qui soient beaucoup moins solubles que le verre dans les milieux géologiques ou qui puissent servir de matrices d'irradiation au cas où la transmutation serait adoptée ultérieurement.

- La séparation et la transmutation ne sauraient en aucun cas se substituer à l'enfouissement en formation géologique des déchets radioactifs à vie longue, des concentrés de transuraniens irradiés et des combustibles usés résiduels provenant d'un parc de réacteurs composite.

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PART II:
TECHNICAL ANALYSIS AND SYSTEMS STUDY

1. PARTITIONING

1.1 Aqueous separation techniques

This section briefly describes aqueous separation techniques currently used on industrial scale and research activities in the field of new separation methods for more effective separation of minor actinides and fission products. There has been a large number of reports published until now and a selection of the important ones is listed in Annex D.

1.1.1 PUREX process

The PUREX process, see Figure II.1, which is universally employed in the irradiated fuel reprocessing industry, is a wet chemical process based on the use of TBP, a solvent containing phosphorus. As shown in Table II.1 this solvent displays the property of extracting actinide cations in even oxidation states IV and VI, in the form of a neutral complex of the type $M \bullet A_n \bullet 2TBP$ (where M is the metallic cation and A an anion, generally nitrate ion), from an acidic aqueous medium. Conversely, the actinide cations with odd oxidation state are not significantly extracted, at least in the high acidity conditions prevailing during reprocessing operations.

Uranium and plutonium, whose stable oxidation states in nitric medium are VI and IV, respectively, are co-extracted by TBP and thus separated from the bulk of the fission products which remain in the aqueous phase. This is the basic principle of the PUREX process.

Table II.1 **Extractability of actinide nitrates in 3 M nitric acid by TBP**

	Oxidation state			
	III	IV	V	VI
U		(❍)	(●)	❍
Np		(❍)	●	❍
Pu	(●)	❍	(●)	(❍)
Am	●	(❍)		
Cm	●			

❍: extractable by TBP, ●: not extractable by TBP, ( ): unstable in the media

Uranium and plutonium are recovered with an industrial yield close to 99.9% (including losses in secondary wastes).

1.1.1.1 Minor actinides

Americium and curium

Among the minor actinides, americium and curium, which are stable in valency III, are not extracted by TBP and remain in the aqueous phase. They accordingly follow the path of the fission products and are currently managed like the latter by conditioning in a glass matrix.

Neptunium

Another minor actinide, neptunium, whose stable oxidation state is V, is hence very slightly extractable in this species by TBP. However, in the chemical conditions of the first cycle extraction operation of the PUREX process (presence of nitrous acid), part of the Np(V) is oxidised to VI, and accordingly extracted in the organic phase.

The operating results of the UP3 plant reveal that the majority of the Np is extracted by TBP, follows the uranium stream, and is separated from the latter in the second uranium purification cycle. The effluent containing neptunium is currently added to the high-level waste stream. Hence all the neptunium is sent to vitrification apart from the proportion following the Pu product. The behaviour of neptunium is independent of the type of fuel reprocessed.

1.1.1.2 Long-lived fission products

Among the fission products with long-lived isotopes, three elements (technetium, zirconium and iodine) display specific behaviour in the PUREX process, which could be exploited for their separation (see Figure II.1).

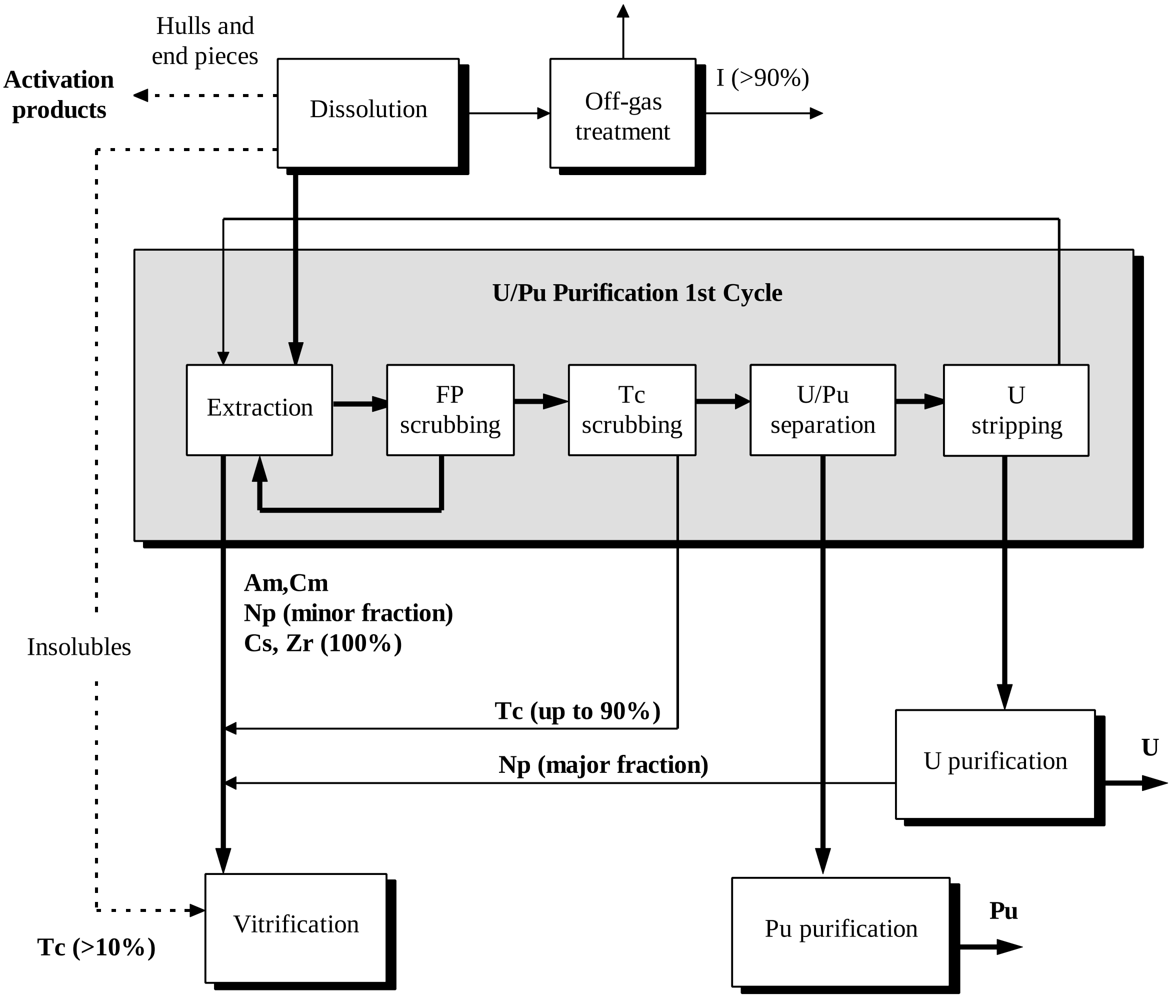
Technetium

During fuel dissolution, part of the technetium, probably in metallic or oxide form, does not go into solution. This fraction, estimated at 10 to 20% of the total Tc for a UO_2 fuel, accompanies the "insoluble residues" essentially consisting of noble metals (Ru, Rh, Pd). These insoluble residues are currently incorporated in the vitrified or cemented wastes.

In nitric medium and in the absence of a reducing agent, the dissolved technetium is in its highest valency (VII) which is the most stable. It occurs in anionic form in the state of pertechnetate ion, TcO_4^-. This species can be extracted by TBP at the same time as a metallic cation by substitution of a pertechnetate group for a nitrate group in the neutral complex extracted.

The occurrence of this co-extraction was observed with the main metallic cations extracted by TBP, and particularly with the cation ZrO^{2+} (see below), which is present in large amounts in the feed solution to the extraction cycle. This extraction of Tc proved to be a serious hindrance because of its interference with the chemical mechanisms of the partitioning operation (U/Pu separation). The PUREX process accordingly had to be adapted to limit the extraction of Tc.

Figure II.1 **Behaviour of long-lived elements in PUREX process (An example of UP3 La Hague)**



The final flow chart, which includes a specific stripping step for the Tc extracted by the TBP, produces an effluent containing a large fraction of the element. This effluent is currently added to the main fission product stream.

Zirconium

Zirconium, which is present in the form of the ion ZrO^{2+}, can be extracted by TBP, but to a lesser degree than uranium and plutonium. This element is effectively stripped in the "FP washing" operation, which immediately follows extraction. The aqueous phase from the washing must be recycled to extraction due to the large amounts of U and Pu which it contains. In the present status of the process, zirconium is not specifically isolated.

Iodine

Iodine presents a special case in so far as this element is extremely volatile in the elemental state. This property is exploited for the containment of this fission product in the "process head end", thus preventing its dispersion in the downstream operations where its controlled management would be quite difficult.

The operating conditions of fuel dissolution are selected to ensure that the iodine is brought to and maintained in the elemental state, and to entrain it in the off-gas. The iodine is recovered in an aqueous solution by caustic scrub of the off-gas.

This specific effluent of iodine, which is discharged into the sea today, thus contains nearly all the iodine initially present in the irradiated fuel. Some reprocessing plants envisage the use of iodine immobilisation by adsorption on silver impregnated zeolites.

Other long-lived fission products

As to the other long-lived fission products, it is clear that the PUREX process cannot be used to separate caesium and strontium, since these mono- and divalent elements are unextractable by TBP. The behaviour in the PUREX process of the other fission products which have long-lived isotopes (Pd, Se, Sn) is not precisely known. A combined electrolytic extraction of Pd^{2+} with the other platinum group elements ($RuNO^{3+}$, Rh^{3+}) and TcO_4^- (and probably SeO_4^{2-}) seems to be promising from even higher acidic PUREX liquors [1].

1.1.1.3 Long-lived activation products

The activation products formed in the fuel element structural metals (stainless steels, inconel and zircaloy) mostly remain in these materials and are found in the corresponding “hulls and end pieces” waste stream. The ^{14}C issue should receive increasing attention because of this isotope’s impact on the biosphere.

1.1.1.4 Conclusions

The behaviour of the minor actinides and long-lived fission products in the PUREX process can be divided into three categories:

- elements already partially separated by the PUREX process: neptunium, technetium and iodine. For these elements, the R&D objective involves process extensions to achieve the desired separation performance. This first aspect is discussed further in Section 1.1.2.
- elements separable by TBP, for which a complementary step to the present PUREX process can be developed. This applies to zirconium.
- elements that cannot be separated by the PUREX process:
  - americium and curium,
  - caesium, strontium and probably the other fission products (Pd, Se, Sn).

To separate these elements, it is necessary to develop new classes of extractants, or to resort to different separation methods. The corresponding developments are discussed in Section 1.1.3 (Am and Cm) and Section 1.1.4 (FPs).

1.1.2 Improved separation of long-lived elements in PUREX process

1.1.2.1 Neptunium

Neptunium is present in the irradiated fuel dissolution liquor (nitric acid medium) in oxidation states V and VI. The extractability of Np(V) by tributylphosphate (TBP) organic solution (the PUREX solvent) is rather poor whereas that of Np(VI) is good, approaching that of U(VI) and Pu(IV). Hence one alternative to separate neptunium from the wastes is to extract this element in the first cycle of the PUREX process together with U(VI) and Pu(IV). This requires the oxidation of the whole neptunium inventory to the VI oxidation state, allowing Np extraction by TBP. Thus the redox reaction to convert Np(V) to Np(VI) is the key point to be addressed to achieve ~100% Np extraction in the first cycle.

Np(V) can be oxidised to Np(VI) by various means including:

- chemical oxidation using nitric/nitrous acid mixture, like those present in the fuel dissolution liquors, intentionally adding oxidants such as vanadium (V) compounds;
- using force field oxidation like: β, γ radiolysis oxidation, photochemical oxidation, sonochemical oxidation.

After its co-extraction with U and Pu, neptunium can be selectively separated from these elements, either using the regular PUREX cycles (for example in the second uranium cycle) or under the action of specific reagents like butyraldehydes.

Computer codes of the PUREX process are available for calculating the behaviour of Np in the PUREX extraction cycles. The current research trend in this area is to define more refined chemical models for Np behaviour that are more elaborate than those hitherto employed. In particular, these new codes incorporate chemical laws which account for deviations from ideality for the main reactions, especially the following:

$$\mathrm{NpO_2^+ + 1/2\,NO_3^- + 3/2\,H^+ \Leftrightarrow NpO_2^{2+} + 1/2\,HNO_2 + 1/2\,H_2O} \qquad (1)$$

Reaction (1) is autocatalytic (nitrous acid catalyst). Its control is certainly the key to success for a near-quantitative extraction of Np in the first cycle of the PUREX process.

The quantitative extraction of Np in PUREX process was demonstrated by PNC with counter-current extraction test for FBR spent fuel solution of 5 M HNO_3. This is probably due to the oxidation of Np to hexavalent oxidation state by nitric acid in high temperature condition [2].

1.1.2.2 Technetium

Intensive technetium separation must be considered from two different viewpoints because technetium is divided into a soluble form and an insoluble form.

The behaviour of solubilised technetium in the PUREX process is now clearly understood, and separation flow charts can be proposed to isolate 97 to 98% of dissolved Tc. These flow charts have already been used in the facilities at La Hague and the results obtained confirm the validity of the behaviour model used to develop these flow charts.

By contrast, information about the insoluble fraction is much more limited, beginning with the quantities of technetium present in this form. The necessary steps include, in the following order, developing analytical methods to measure technetium in the dissolution insolubles for different types of fuel, followed, if necessary, by the development of methods to treat the insolubles to recover the technetium. Ideas concerning this separation are still in the embryonic state for the time being. Presently, the insoluble residues are separated from the dissolution liquor before entering the PUREX process. The insoluble residues are stored and eventually mixed with the other FPs for vitrification.

1.1.2.3 Iodine

The process applied today in the La Hague plants helps to recover over 95% of the iodine theoretically calculated in the fuel. In the absence of a demonstrated need to improve this recovery, no specific research is currently conducted on this topic.

Distribution of iodine in the Tokai Reprocessing Plant has been balanced between two discharge effluents (to the sea and to the atmosphere), and both have been kept lower than domestic regulatory discharge levels. In order to reduce further the discharge to air, silver-impregnated adsorbents (using carrier such as inorganic zeolite, organic hydrophobic polymer) have been developed in the course of commercial campaigns [3].

1.1.2.4 Carbon

In dissolving irradiated fuel, carbon is in the form of CO_2 and is partly transferred to the dissolution off-gas. In BNFL, the off-gas is scrubbed with sodium hydroxide solution in the off-gas treatment process where carbon is fixed as sodium carbonate. A specific conditioning rig using barium carbonate precipitation ($Ba^{14}CO_3$) has been investigated and could be installed [4].

1.1.3 Separation of minor actinides

1.1.3.1 Neptunium separation

As described in 1.1.2.1, the separation of Np in PUREX process has been investigated mainly aiming at the valence adjustment of Np by redox reactions. Other attempts were carried out to separate Np from the raffinate of the PUREX process.

DIDPA

In the DIDPA process developed by JAERI, neptunium in any valence state present in the High Level Liquid Waste (HLLW) is extracted by the solvent (0.5 M DIDPA – 0.1 M TBP) together with other actinides and lanthanides if hydrogen peroxide is added during the extraction. This is due to the reduction of Np(V) to Np(IV). The extracted Np is then selectively stripped by 0.8 M oxalic acid solution with Pu. More than 99.95% recovery of Np was demonstrated with a simulated HLLW [5].

HDEHP

Yang *et al.* of KAERI [6] demonstrated the separation of Np by HDEHP from simulated HLLW solution in which the valence of Np is adjusted to tetravalent by 1.0 M hydrogen peroxide. More than 99% of Np was extracted by HDEHP and more than 99.4% was back-extracted by 0.5 M oxalic acid solution.

TRUEX

The distribution coefficient of Np, valence state of which was equilibrated in feed solution of 5 M nitric acid, to the TRUEX solvent (0.2 M CMPO – 1 M TBP) was observed to be more than 10. PNC demonstrated the quantitative extraction of Np from HLLW solution in hot test where extracted Np was stripped by dilute nitric acid solution [7].

1.1.3.2 Actinide/lanthanide separation

The present oxidation state of Am and Cm in nuclear fuel dissolution liquors is III. The oxidation state of lanthanide fission products, which represent about one third of the total mass of the FP inventory in the spent fuel, is also III. Thus, the major problem here is to separate An(III) from Ln(III). This separation is difficult because:

- these two series of elements (5f and 4f) have very similar chemical properties;
- the mass ratio R = Ln/An is high (R is ~20 for 47.5 GWd/tHM burn-up uranium oxide fuels).

Two alternatives are available to separate the Ans from the other elements present in the wastes. The first is based on co-extraction of An and Ln. The processes such as TALSPEAK, DIDPA, TRUEX, TRPO and DIAMEX are classified in this category. It is possible in the former three processes to separate actinides from lanthanides by selective stripping with complexants such as DTPA, however, further separation of Am/Cm and chemical purification require further separation steps. Another is based on selective extraction of An where an extractant with high selectivity, such as TPTZ and CYANEX 301, plays an important role.

The concept of two-cycle separation process requiring two different solvents are also promising for An separation. In the first step, An(III) and Ln(III) are co-extracted, e.g. by DIAMEX, and separated from the remaining 2/3 of the FPs, while in the second step, An(III) is separated from Ln(III) by selective extraction. The advantage of this process is that the purity of An product is high and that the volume of secondary waste is reduced.

Co-extraction of actinides and lanthanides

TALSPEAK and DIDPA

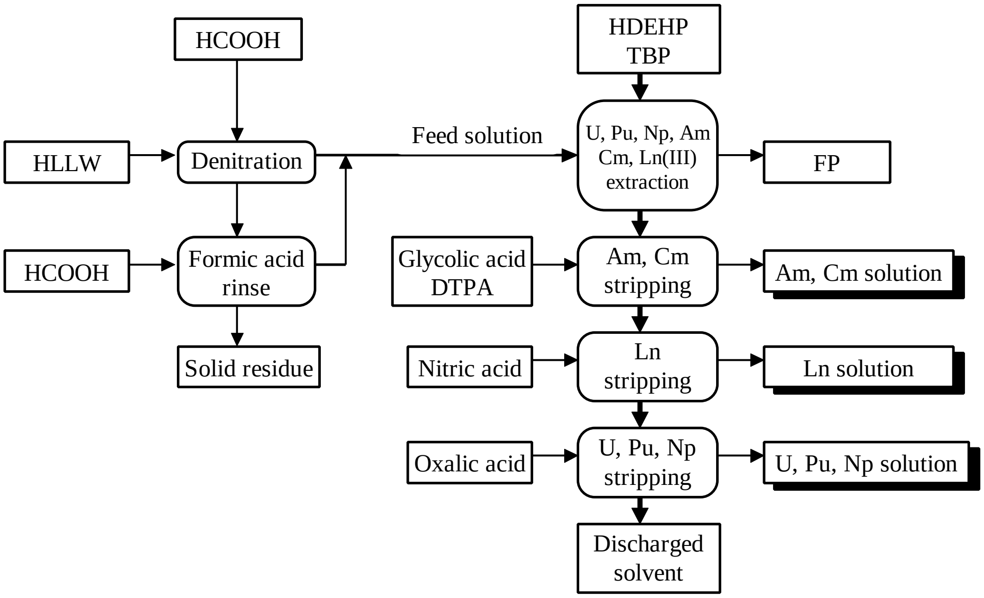
The principle of the minor actinide separation here is the co-extraction of the MAs with other elements present in the high active raffinate or in the HLLW, such as lanthanides, and then to partition the different extracted elements by selective stripping. Two processes are known in this category, both based on very similar or identical chemical reagents: the so-called TALSPEAK [8] (see Figure II.2) and

DIDPA [5] (see Figure II.3) processes. For both processes the MAs and other elements are extracted with an acidic organophosphorous extractant, di-2-ethyl-hexyl-phosphoric acid (HDEHP for TALSPEAK) or di-isodecylphosphoric acid (DIDPA), and An(III)/Ln(III) partition is achieved by selective stripping of An(III) from the loaded solvent with the help of aqueous stripping solutions containing the following complexing agents: alcohol-carboxylic acid (traditionally lactic or glycolic acids, or citric acid as proposed recently) and diethylenetriaminopentaacetic acid (DTPA). It is generally believed that the selective stripping of Ans is due to the fact that An(III)/DTPA complexes are more stable than the corresponding Ln(III) complexes.

Since HDEHP and DIDPA extractants are cation exchangers, the nitric acid concentration of the HLLW to be treated must be drastically reduced. This can be done, for example, by denitration with formic acid, as investigated by JAERI scientists [9]. A definite advantage of the DIDPA process compared with TALSPEAK resides in the higher affinity of the extractant for the metal ions to be extracted, so that they can be extracted from a more acidic aqueous solution (HNO_3 = 0.5 M) than in the TALSPEAK process. The amount of secondary effluents is of the same order as the TRUEX process.

Consequently, a major drawback of these processes, i.e. the precipitation of some FPs in the form of hydroxides which can carry a fraction of the TRU present in the waste, can be minimised.

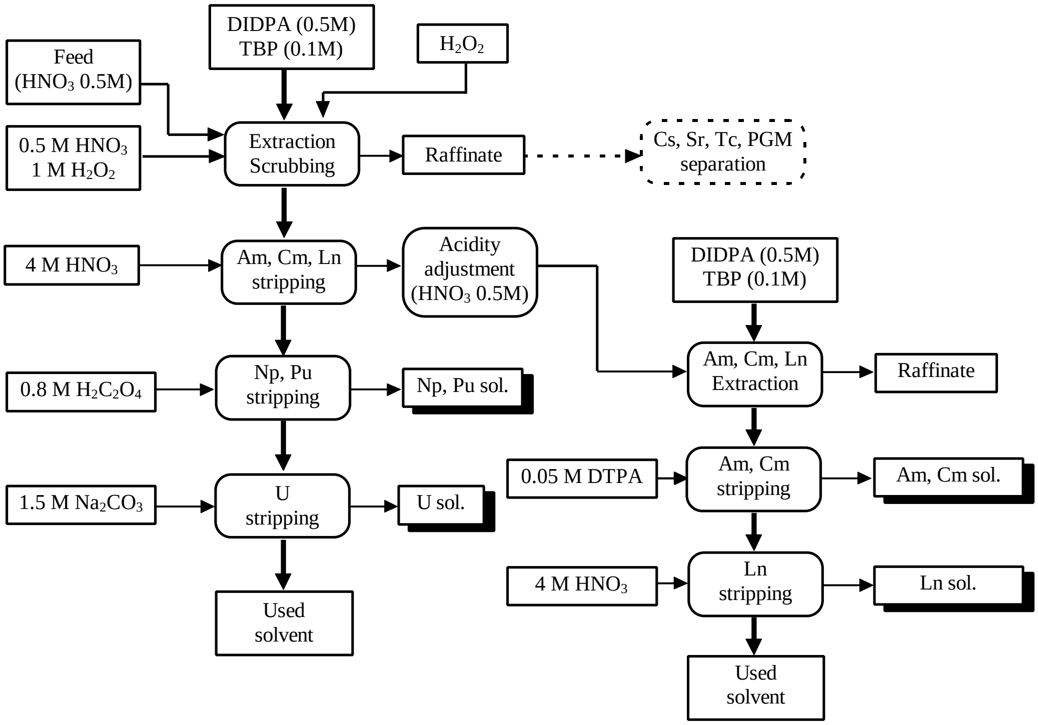
Figure II.2 **TALSPEAK process**



To prevent the formation of extracted polymers of metallic species which are difficult to strip, such as Ans and Lns for example, it is necessary to limit the concentration of the metallic species in the solvent. Consequently, the solvent inventory required for these processes is rather high.

In DIDPA process (see Figure II.3) more than 99.95% recovery of all actinides was demonstrated with a simulated HLLW and 99.99% recovery of Am and Cm with real HLLW [5]. Recent activities of JAERI’s study are devoted to the confirmation of the effectiveness of the four group partitioning process (see Annex B) including DIDPA extraction with real HLLW and to the fundamental study on its practical application.

Figure II.3 **DIDPA process**



TRUEX

The TRUEX (TRansUranium EXtraction) process is based on the use of neutral organophosphorus bidendate extractant: n-octyl-phenyl-di-isobutyl-carbamoylmethyl-phosphine-oxide (named CMPO). It was developed in the 80s by Horwitz *et al.* [10] to decontaminate the huge amounts of TRU waste accumulated in the US during the Cold War in defence nuclear material production sites (Hanford, Idaho etc.). This process is also studied by Japanese (PNC), Italian and Indian scientists for partitioning commercial wastes. CMPO displays high and low affinities for An(III) and Ln(III) nitrates at high and low aqueous nitric acid concentration, respectively. Consequently, an extraction-stripping cycle can easily be designed. Of course, other metallic species are also extracted by the solvent. To separate these metallic species from the An(III)+Ln(IIII) fraction, it is possible to:

- add complexing agents to the feed (e.g. oxalic acid);
- scrub the loaded solvent with aqueous complexing solutions (e.g. oxalic acid);
- strip them selectively after stripping the An(III)+Ln(III) fraction.

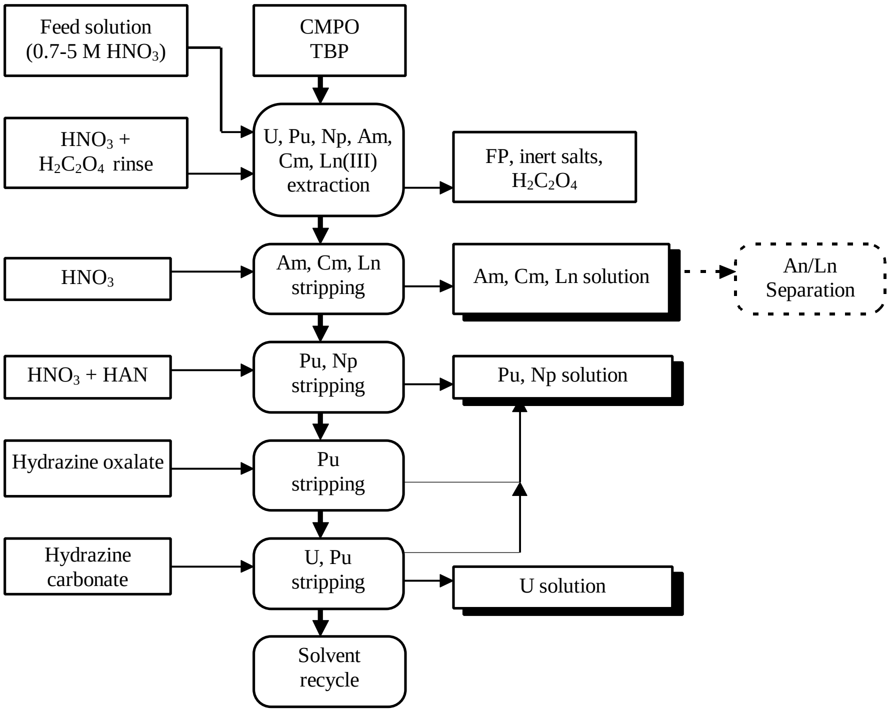
To cope with the third phase formation problem, the solvent contains a high concentration of TBP (1 to 1.4 M) used as a modifier. CMPO mixed with TBP allows the extraction of the most important actinides, except Np(V), and the process is directly applicable to HLLW solutions with a HNO_3 concentration of 0.7 to 5 M as it results from the conventional PUREX process. CMPO is such a powerful extractant (distribution ratio of 10^4) that quantitative stripping of extracted actinides, especially U and Pu, is difficult. Am extracted by CMPO can be recovered quantitatively but it is accompanied by Cm and the bulk of rare earths. The TRUEX process needs to be complemented by an Am-Cm/rare earth separation (TALSPEAK process) and possibly by an Am/Cm separation. The secondary waste production in the TRUEX process is not negligible due to the additional steps.

The generation of secondary effluents starts beyond the TRUEX extraction step:

Stripping of the Am-Cm-RE fraction from loaded CMPO-TBP:	500 l/tHM equiv.
Washing of the TRUEX solvent:	250 l/tHM equiv.
Separation of rare earths from Am-Cm (e.g. TALSPEAK):	3 000 l/tHM equiv.
Am/Cm separation (method unknown):	large volumes

Compared to the classical PUREX process (5000 l/tHM) this additional TRUEX sequence will increase the bulk volume of effluents to be treated.

Figure II.4 **TRUEX process**



One drawback of this process is the effect of solvent degradation products in the process. Some of these are cation exchangers which prevent the efficient stripping of the An(III)+Ln(III) fraction. Another problem is the difficulty in stripping U(VI) and An(IV) from the solvent due to their high affinity with CMPO. In order to eliminate this problem, the solvent clean-up step must be performed thoroughly with strong complexing agents e.g. hydrofluoric acid or diphosphonic acids (TUCS), but it causes the increase of the secondary waste. PNC proposed the use of “salt-free” reagents such as HAN, hydrazine oxalate and hydrazine carbonate which were adopted to the TRUEX flowsheet [7] (see Figure II.4).

In addition, CMPO has been considered not to be effective for An(III)/Ln(III) separation. Currently, a new separation scheme has been proposed by PNC utilising DTPA as a selective strip reagent for An(III). It was demonstrated that An(III) could preferentially be co-stripped with Cm(III) and heavier Lns by DTPA-$NaNO_3$ solution and that partial fractionation was possible by this system named SETFICS [11].

A computer code of the TRUEX process (GTM) was developed by G.F. Vandegrift at ANL (US) and successfully tested not only for US experiments but also for PNC (Japan) experiments. Hot tests of the TRUEX process were successfully completed in the US and Japan (PNC) in recent years.

Carbamoylphosphine oxides (CMPO) known for their ability to remove actinides from high activity (HA) liquid waste are used in the TRUEX process. Horwitz showed that the actinides are included in complexes with several CMPO molecules (two to four). It seems interesting to synthesise molecules in which several CMPO moieties are combined in a suitable arrangement, this may lead to more efficient and selective extractant. Calixarenes bearing four diphenyl acetamido phosphine oxide functions on the upper rim, synthesised by Böhmer display at low concentration (10^{-3} M) in nitro-phenyl-hexyl-ether (NPHE) an higher extracting ability than CMPO used at a concentration ten times higher towards trivalent and tetravalent actinides [12]. In contrast to CMPO which displays low selectivity towards lanthanides, a strong decrease of lanthanide distribution coefficients is observed from 140 for lanthanum to 0.19 for ytterbium when calixarenes CMPO are used. One has to point out that this discrimination is suppressed when phenyl borne by phosphorus are replaced by hexyl groups [13].

Trialkyl phosphine oxide (TRPO)

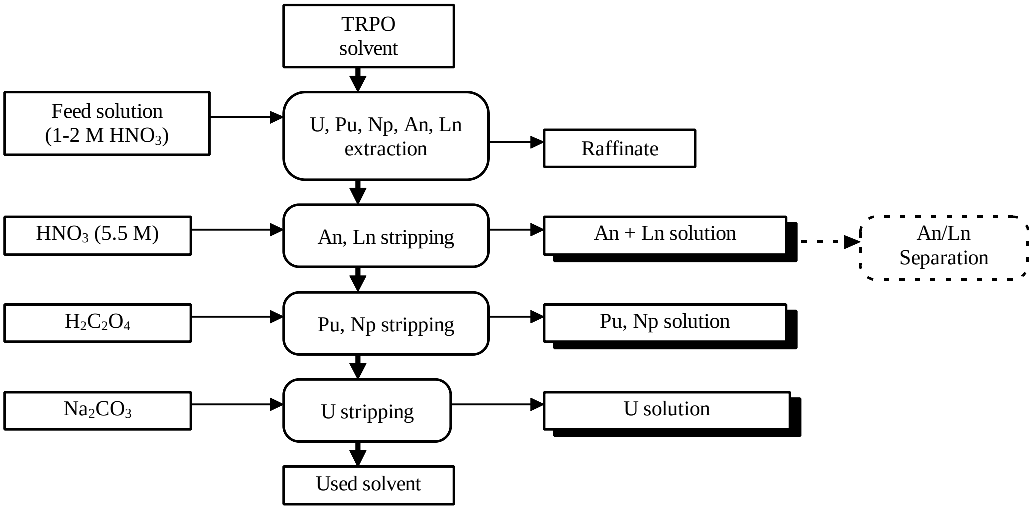
The TRPO process, developed by Zhu, Song *et al.* in Tsinghua University (China), is based on the use of liquid mixtures of TRPO soluble in aliphatic hydrocarbon diluent (kerosene) [14].

The affinity of the TRPO extractant for trivalent actinides and lanthanides is high for moderate aqueous nitric acid concentration (1 M) and low for high acidity (5 M), respectively. Consequently, the An+Ln TRPO extraction cycle is performed after neutralisation of the nitric acid in the feed to 1 M and the An+Ln mixture is stripped from the solvent using a HNO_3 = 5.5 M aqueous solution. The process was invented in the People’s Republic of China and successfully tested at the ITU of Karlsruhe with diluted HLLW solution. The decontamination factors for TRUs range from 10^3 to 10^4 in 1 M HNO_3. In principle the method requires a slight acid reduction step but can operate in 1 to 2 M HNO_3. The great advantage of the method is its reversibility in extraction and stripping, its miscibility with TBP and especially its loading capacity. However, the extraction requires additional separation steps similar to those described for TRUEX and DIDPA. Certain fission products (Zr, Mo, Ru and Tc) interfere with the separation. The presence of the high nitric acid concentration in the An+Ln mixture obtained after implementation of the TRPO process is a drawback for the subsequent An/Ln separation cycle, which requires rather low acidity for almost all the systems studied today (see below).

The TRPO process (Figure II.5) was the subject of many developments in China as well as Europe at the ITU (Karlsruhe).

The formation of secondary waste streams from the primary extraction process results mostly from the acid destruction and from the use of Na_2CO_3 as solvent washing agent. It may be expected that the overall secondary waste volume will be higher than 10 000 l/tHM equivalent.

Figure II.5 **TRPO process**



Diamide extraction (DIAMEX)

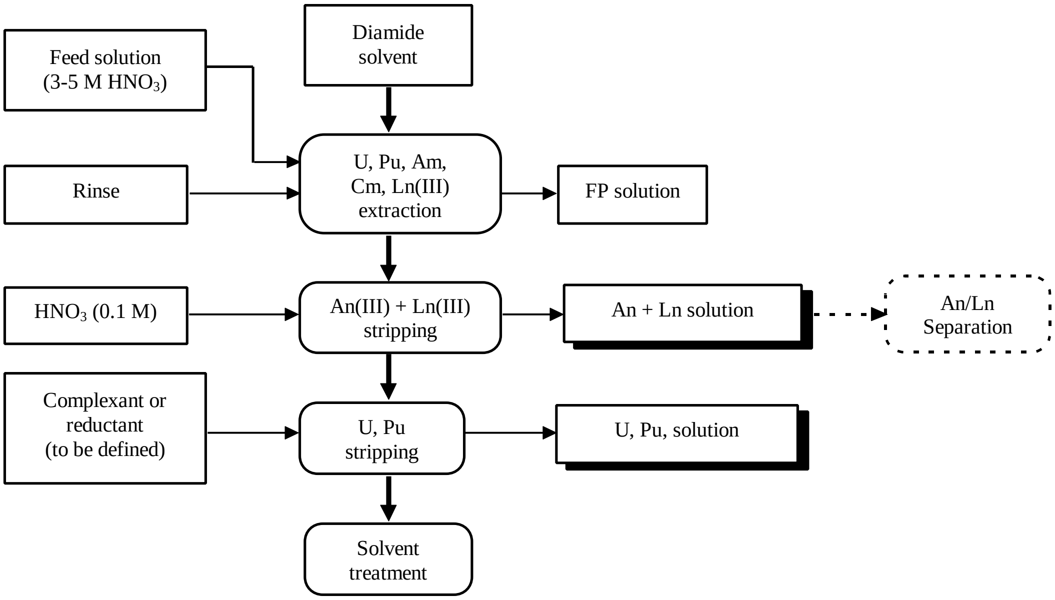
The DIAMEX (DIAMide EXtraction) process was first developed by Musikas *et al.* [15] at the CEA Fontenay-aux-Roses Research Centre (France) and by C. Madic and M.J. Hudson in a joint European research programme involving the CEA (Fontenay-aux-Roses) and the University of Reading (UK) [16]. This process is based on the use of malonamide extractants. So far, the reference substance developed for the definition of the first version of the DIAMEX process is di-methyl-di-butyltetradecylmalonamide (DMDBTDMA). This reagent has up to now the most attractive properties as actinide extractant but has to be considered as a compromise between its behaviour as chemical extractant and its physical behaviour in extraction conditions (viscosity, emulsion, settling time, etc.). The diamide extractant is used in solution in an aliphatic diluent. The extracting properties of DMDBTDMA are, to some extent, similar to those exhibited by CMPO (TRUEX process), hence the extraction-scrubbing-stripping cycle of the DIAMEX process resembles TRUEX. DIAMEX offers the following advantages over TRUEX:

- DIAMEX degradation products are less troublesome than TRUEX products;
- no secondary solid waste is expected from the use of the solvent because it consists of hydrogen, carbon, nitrogen and oxygen and is fully incinerable. On the contrary, in TRUEX, the high phosphorus concentration (in TBP and CMPO) in the solvent causes the production of secondary solid waste.

The DIAMEX process (Figure II.6) was tested successfully in 1993 on real waste at Fontenay-aux-Roses Research Centre. The process continues to be developed as part of a European co-operation project. Optimisation of the diamide formula is underway. Diamide extractants have also been investigated in Japan, the UK, the US, Switzerland and India.

Since the diamide extractants are fully incinerable no solid secondary wastes are expected.

Figure II.6 **DIAMEX process**



Selective extraction of actinides

Tripyridyltriazine (TPTZ)

Tripyridyltriazine ligand (TPTZ) is a terdendate nitrogen-donor ligand which can selectively extract An(III) from An+Ln mixtures when used in synergistic combination with an organic cation exchanger. This system was first studied in the 80s by Vitorge [17]. Good separation factors were obtained for An(III) *vs* Ln(III) using TPTZ+HDNNS (di-nonylnaphthalenesulfonic acid) or TPTZ + alpha-bromocapric acid. Tests of the process using synthetic spiked solutions were successfully conducted at Fontenay-aux-Roses Research Centre. Improvements to the process are underway in a European research programme. One way to improve the system is to use the lipophilic alkyl derivative of TPTZ developed jointly by the University of Reading (UK) and the CEA (France). Recently, Kolarik *et al.* at the FZK (Germany) designed a very efficient family of molecules related to TPTZ able to extract selectively An(III) over Ln(III) from aqueous nitric acid solution [18].

CYANEX 301ä

Zhu *et al.* in Tsinghua University (China) recently published extraordinary results for the separation of An(III) over Ln(III) using CYANEX 301 extractant [19]. CYANEX 301 consists chiefly of bis (2,4,4-trimethylpentyl) dithiophosphinic acid. The commercial product contains many impurities some of which are detrimental to the extraction of metal ions, and especially to the separation of An(III) over Ln(III). In the case of crude CYANEX 301, no An/Ln separation is observed unless the extractant is saponified (i.e. neutralised with an alkali). In this case, very high An(III)/Ln(III) separation factors can be obtained. After purification of CYANEX 301 by precipitation of its ammonium salt, there is no need for saponification of the extractant to obtain tremendously high An(III)/Ln(III) separation factors, up to 5 900. Nevertheless, one of the drawbacks in using CYANEX 301 for An/Ln separation is the fact that the aqueous solution must be adjusted to a rather high pH of 3.5 to 4.

According to Jarvinen *et al.* in LANL (US), CYANEX 301 can be used in synergistic combination with TBP for An(III)/Ln(III) separations [20]. Hill *et al.* recently found in the frame of CEA/JNC collaboration programme that purified CYANEX 301/TBP synergistic mixture yielded higher separation ($SF_{Am/Eu}>10^4$) than CYANEX 301 alone, especially at lower $pH_{eq} = 2$ [21].

The separation of Am(III) over Ln(III) from a real waste was successfully demonstrated with bench scale 4 stage cross-flow hot test by Zhu *et al.* [22]. An empirical distribution ratio model and a method of counter-current extraction process parameter calculation were established and the results of calculation were verified by a five-stage mixer-settler experiment with tracer amount of ^{241}Am and macro amount of Lns.

Counter-current tests of the separation Am(III)/Ln(III) from actual HLLW using CYANEX 301 plus TBP synergistic mixture were also carried out successfully by Glatz *et al.* at the ITU Karlsruhe, Germany [23].

One drawback of CYANEX 301 is the very low acidity (pH~4) required for the feed solution. Modolo *et al.* synthesised extractants by replacing the 2,4,4-trimethylpentyl group of CYANEX 301 with halogenated phenyl. The extractant $(Cl\text{-}C_6H_4)_2PS(SH)$ showed good extraction properties and chemical stability if used with trioctylphosphine oxide (TOPO) as synergist. The distribution ratio of Am(III) was higher than 400 and that of Eu(III) was less than 0.5 (Separation Factor = 88) even with 0.4 M of nitric acid [24].

These results related to An(III)/Ln(III) separation are certainly the most important in this field in many years. A great deal of work can be expected in the near future in various countries using this extractant.

Other new methods

Among the new methods proposed recently for trivalent An/Ln separations is the one reported by Kulyako *et al.* in Moscow (Russia), who announced at the NRC4 Conference held at St-Malo (France) in September 1996, that An(III)/Ln(III) separations can be carried out simply by the selective precipitation of the Ans from acidic aqueous solution after the addition of potassium ferricyanide [25]. Moreover, the separation between Am and Cm could be achieved by contacting the Am + Cm ferrricyanide precipitates with a basic solution (soda) containing ferricyanide ions: Am dissolves as Am(V) species whereas curium remains as a Cm(III) ferricyanide precipitate. More work is needed in this area to check these results and consider their potential applications.

Picolinamides are N and O chelating agents which can selectively complex or extract An(III) over Ln(III). These extractants were studied by Cordier *et al.* at the CEA, Fontenay-aux-Roses [26], and then Marcoule (France).

The selective extraction of trivalent Ans requires high nitrate concentration and low proton concentration, making it difficult to consider the development of a process based on this system. On the other hand, high distribution ratios for An(III) and high An(III)/Ln(III) separation factors are obtained from an aqueous solution containing pertechnetic acid. It is thus possible to consider the development of a separation process based on this system.

Nevertheless, Goto *et al.* recently reported, by using novel organo-phosphorus bi-functional ligands in which no soft nitrogen or sulphur donor atoms are incorporated, high selectivity for inter-lanthanide separation and ^{152}Eu/^{241}Am separation ($SF_{Eu/Am}>10$) at acidic medium (pH<1). A promising candidate is ODP (1,8-octanediol-O,O'-diphenyl phosphonic acid) [27].

1.1.3.3 *Americium/curium separation*

Two methods are available for the separation between americium and curium:

- the first exploits the existence in americium of oxidation states higher than III, unlike curium, for which these oxidation states are practically inaccessible in aqueous medium. The SESAME process, described below, uses this difference in oxidation states to separate Am and Cm.

- the second consists in using the difference between the partition coefficients of americium and curium in the DIAMEX solvent. With a sufficient number of separation stages, a flow-chart could in principle be developed to extract the americium and leave the curium in the aqueous phase.

SESAME process

Americium exhibits the specific property to exist in nitric medium in oxidation states IV and VI. Since this property is not shared by lanthanides (except for cerium), it offers a basis for an original separation process. Curium can not be separated by this process because it is very difficult to obtain the oxidation states higher than III in aqueous medium.

In the SESAME process [28], developed at the CEA, an electrochemical method is used to oxidise the americium to IV or VI, and then separate it. However, it is difficult to obtain and keep the higher oxidation states of americium in aqueous medium because of their instability. This makes it necessary to use a complexing agent which has the effect of reducing the apparent normal potentials of the REDOX pairs implicated.

The choice fell on cage-like heteropolyanions, which offer high complexing capacity for tetravalent elements. Potassium phosphotungstate, $P_2W_{17}O_{61}^{10-}$, is the compound employed so far. A test campaign conducted in 1994 at the CEA on real solutions effectively demonstrated that americium could be stabilised in this way in the oxidised state.

The oxidised species of americium can be separated either by selective extraction:

- by organophosphorus solvents or amides for Am (VI);
- by amines for americium (IV) in complexed form, or;
- by nanofiltration of the complexed species of Am (IV).

This second step of the SESAME process has not yet gone beyond the exploratory stage.

Three applications have been planned for the SESAME process (see Figure II.7).

- Separation of americium from the high-level solution produced by the PUREX process (*SESAME A*). Technically speaking, this operation is complex because the solution contains many chemical elements that interfere with the oxidation and complexation of americium. These include ruthenium, which must be eliminated prior to the oxidation step.

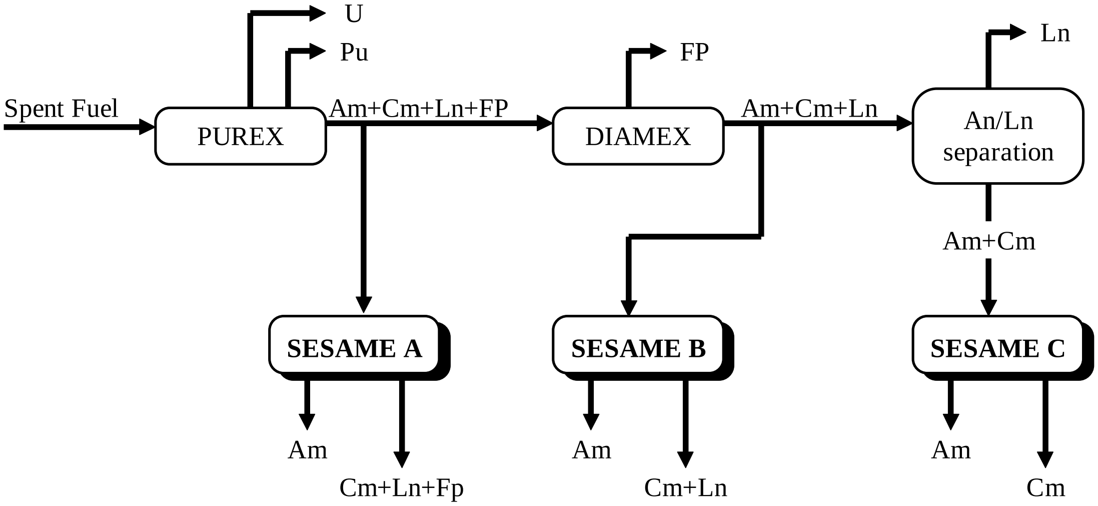
- Separation of americium from the stripping solution of the DIAMEX process (*SESAME B*). This operation is significantly easier than the previous one, because the interfering elements are present in much smaller quantities.

- Americium/curium separation (*SESAME C*), which is probably the simplest application of this process, but which might generate large volumes of waste.

It may be observed that the first two applications can be used to isolate *americium* but not *curium*.

Recently, Adnet *et al.* reported the results of the hot test using Am-Cm or Am-Cm-Nd mixture solutions [29]. The effective oxidation of Am(III) to Am(IV) in the presence of $P_2W_{17}O_{61}^{10-}$ by electrolytically generated Ag(II) was examined. It was also demonstrated that Am(VI) was extracted to HD(DiBM)P (27 wt%) - silica gel column with the yield of ca. 90% and the purity of more than 95%.

Figure II.7 **Possible separation schemes related to SESAME process**



As a conclusion we might say that great progress has been made in the development of separation techniques useful in partitioning operations. It is possible that in the next decade a fully acceptable partitioning technology, based on a single method or on a mixture of different separation techniques will be developed and tested in hot demonstration facilities.

1.1.4 Separation of long-lived fission products

1.1.4.1 Separation of strontium and caesium

The adsorption method with inorganic exchangers, titanic acid and zeolite has been developed for the separation of heat generating nuclides such as ^{90}Sr and ^{137}Cs in many countries during the sixties and the seventies, and recently reinvestigated by JAERI [30]. More than 99.9% recovery from real HLLW was successfully demonstrated. This adsorption method adopted in the four group partitioning process (see Annex B) greatly contributes to the reduction of the waste volume after the partitioning of HLLW because the inorganic exchangers loaded with Sr and Cs can be solidified into a very stable form by direct calcination at high temperature. This also contributes to the overall reduction of secondary waste because the material used for the adsorption can be used as the mother material for the Sr and Cs solidification.

Cobalt dicarbollides were first synthesised and produced in Czechoslovakia for application to caesium and strontium extraction [31]. From 600 kg of hexachloro derivative of dicarbollide, synthesised by KATCHEM (Prague), diluted in nitrobenzene, the extraction of strontium and caesium has been tested on a plant scale in the former USSR [32]. The use of highly toxic nitrobenzene is a drawback for the use of dicarbollides, so in the framework of a project supported by the European Commission, efforts were focused on the synthesis of dicarbollides soluble in diluents other than nitrobenzene. Promising results were achieved with bis-ylene cosan (BISPHECOSAN) diluted in nitrophenyl-alkyl-ether (NPHE or NPOE) or in solubilizers such as diethylpropanesulfonamide (DEPSAM) or dibutylmethanesulfonamide (DIBUMESAM). The presence of two phenyl groups enhances the caesium extraction from acidic media [33].

1.1.4.2 Separation of caesium

The separation of caesium at the CEA (Dozol *et al.* [34]) was first approached in connection with a ^{137}Cs decontamination study on highly saline low- and medium-level effluents, rich in sodium nitrate.

The problem accordingly demanded highly selective separation chemistry, capable of differentiating between two alkaline cations, sodium and caesium, chemically very similar but possessing very different ionic radii.

The basic idea was to use macrocycles of the calixarene type, functionalized by etheroxide chains. Calixarenes are cyclic oligomers produced by the condensation of phenolic units on formaldehyde (the name is derived from their shape which resembles a calyx). They only display weak complexing properties, and must be functionalized, for example, by grafting one or two etheroxide chains on either side of the macrocycle cavity.

These compounds, called calix-crowns, thus display pre-organised co-ordination sites, which can be perfectly adjusted to the dimensions of the Cs^+ ion, giving them strong affinity, and, above all, outstanding selectivity in terms of caesium/sodium separation. Separation factors in the range of 30 000 in favour of caesium have been obtained.

These calix-crowns were naturally selected in an attempt to separate the caesium contained in a real high-level effluent. The results obtained were highly satisfactory, particularly in terms of selectivity, because no other fission product, actinide or chemical element, except for rubidium, was extracted in more than 1 to 2% [36].

1.1.4.3 Separation of strontium

The SREX (Strontium Extraction) process complemented the TRUEX process for the strontium removal from acidic HA liquid waste. Horwitz chose among the dicyclohexano 18-C-6 derivatives the lipophilic di-t-butylcyclohexano 18-C-6 (0.2 M) diluted in octanol [36]. Tests carried out on simulated waste show the selectivity of crown ether since only barium and technetium are appreciably extracted with strontium by the crown ether. Subsequently Horwitz proposed as diluent a variety of phase modifier/paraffinic hydrocarbon mixtures, among them TBP (1.2 M) in Isopar L [37]. In Bhabha Atomic Research Centre (India), Kumar optimised the extraction of strontium by diluting dicyclohexano 18-C-6 in a mixture butanol (80%)-octanol (20%) [38].

1.1.4.4 Separation of technetium and platinum group metals (PGM)

Two chemical methods have been developed at JAERI for the Tc and PGM separation from the DIDPA raffinate generated from actinides extraction from HLLW: the precipitation method by a denitration with formic acid and the adsorption method with active carbon. The former method might be more effective in practical use because it avoids generation of secondary waste even though the recovery of Tc is estimated to be about 95% as compared with more than 99.9% in the latter method [35]. The demonstration of these methods with real HLLW is proceeding at JAERI.

1.1.5 Conclusions

Table II.2 shows the status of R&D under way on the various elementary processes considered.

Table II.2. **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)			❑	95% separation
(PUREX)		❑		>95% separation
(DIDPA)		❑		
(HDEHP)		❑		
(TRUEX)		❑		
Am + Cm separation:				
* based on An/Ln co-extraction				
(TALSPEAK)			❑	
(DIDPA)		❑		
(TRUEX)		❑		
(TRPO)		❑		
(DIAMEX)		❑		
* based on An selective extraction				
(TPTZ)	❑			
(Picolinamides)	❑			
(CYANEX 301)	❑			SF=5 900
* based on precipitation				
(Ferricyanide)	❑			
Am separation in the oxidised state				
(SESAME)		❑		Am/Cm separation
Tc separation (PUREX)			❑	soluble Tc
(PUREX)	❑			insoluble Tc
Tc - PGM separation				
(Denitration precipitation)		❑		
(Active carbon adsorption)		❑		
I separation (PUREX)			❑	95% separation
Zr separation (PUREX)		❑		
Cs separation (Calixarenes)		❑		
(Zeolite)		❑		
Sr separation (Titanic acid)		❑		
Cs and Sr separation (Dicarbollides)			❑	
Pd (PGM), Se, Ru separation				
(Electrolytic extraction)	❑			soluble Pd, Se, etc.

Three phases can be 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 corresponds to 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 corresponds to 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.

Note that the current approach is based on separation element by element. The conditions of a possible grouping of certain elements would simplify the overall management. Moreover, some aspects of the separation processes have not yet been covered by the R&D programmes. These include the preparation of solid products for the fabrication of targets and fuels, for which the specification (product type and purity) remains to be determined.

In addition, the development of new extractant molecules such as diphosphine dioxide is carried out in Russia. Researchers in India are working on the examination of sulfoxide type extractants. These research activities have also the potential to contribute to the development of advanced separation technology.

1.2 Non-aqueous methods

An advanced nuclear fuel cycle is under development based on pyrochemical processes in order to achieve a compact, economic and non-proliferative recycling process. R&D on oxide fuel, metal fuel and nitride fuel are carried out to assess the technological feasibility of pyrochemical process with electrorefining and reductive extraction. This process has a feature that complete separation is difficult.

1.2.1 Pyrochemical reprocessing of oxide fuels for oxide fuel cycle

The Research Institute of Atomic Reactor (RIAR) in Russia has been developing a pyrochemical reprocessing method based on the electrorefining of oxides combined with vibro-pack fuel fabrication since the early 1960s [39]. Pyrochemical reprocessing can be carried out by two ways: separation of UO_2 and PuO_2 during process and joint co-deposition of $(U, Pu)O_2$.

Figure II.8 shows the flow diagram of the first pyrochemical reprocessing option. After decladding of fuel pins, granulated or powdered oxides are chlorinated at 600~650 °C in a pyrographite crucible by passing chlorine gas in order to dissolve uranium and plutonium as UO_2^{2+} and Pu^{4+} in molten NaCl-KCl. In the second stage, uranium oxychloride is electrolysed to deposit UO_2 at the cathode by using a potential at which PuO_2 is not co-deposited. In the third stage, crystallised PuO_2 is precipitated from the salt by circulating a mixture of oxygen and chlorine gas. More than 99% of plutonium from fuel can be precipitated by this process. After recovering plutonium, UO_2 is deposited at the cathode by supplementary electrorefining.

The second option is production of fresh MOX fuel from spent MOX fuel. In this case the spent MOX fuel is chlorinated by a mixture of the eutectic salt NaCl-KCl at 600~650°C. After chlorination of the spent fuels, a special electrolysis under molten salt oxidation is carried out for electrolytic co-deposition of UO_2 and PuO_2. This process is used for production of granulated fuel.

In RIAR this process has been demonstrated with highly irradiated spent oxide fuels with burn-ups of 21% and 24% discharged from the BOR-60 reactor. Good results were obtained during the demonstration. The PuO_2 recovered will be mixed with UO_2 and processed by a vibro-packing fuel fabrication method into fresh MOX fuel [40].

The pyrochemical process allows fuel decontamination from fission products with a factor of more than 100.

It is planned to incorporate the separation process of minor actinides, which is called the DOVITA Programme (Dry reprocessing, Oxide fuel, Vibropac, Integral, Transmutation of Actinides) in this pyroreprocessing method [41]. This R&D programme includes the demonstration studies for minor actinides recycling in the fast reactors. The DOVITA flow diagram (see Figure II.9) includes the following technological steps:

- reprocessing of irradiated MOX fuel and targets containing minor actinides by two methods:
  - pyrochemical reprocessing;
  - partial vacuum-thermal reprocessing.
- manufacture of fuel pins and targets by the vibro-packing method.
- waste and product treatment with the purpose of complete recycle and use of Pu, Np, Am and Cm.

The chemical similarity of neptunium with plutonium in molten chloride salts is used for its pyrochemical reprocessing. Neptunium (as oxychlorides or tetrachloride) will co-deposit with UO_2 or co-precipitate with PuO_2 by supplementary electrorefining for UO_2-PuO_2 recovery. That behaviour was demonstrated during the BOR-60 spent MOX fuel reprocessing [40]. The process for Np pyrochemical co-deposition with UO_2 and UO_2-PuO_2 from molten chloride salts was developed for Np fuel production and experimental fuel pins are under irradiation in the BOR-60 fast reactor [41].

Americium and curium behave in molten chloride systems similarly to the rare-earth elements. So their recovery and separation from molten salt with rare earth elements is difficult. It could be possible for americium to exist in the salt as AmO^+ species under high oxygen partial pressure. This phenomenon is used for the co-deposition of americium with UO_2-PuO_2 during electrolysis. A method for americium co-deposition with MOX fuel by electrolysis is under development.

1.2.2 Actinides recycling combined FBR and LWR cycles by a pyrochemical process

The Central Research Institute of Electric Power Industry (CRIEPI) is investigating the technology of recycling actinides from metallic FBR or reduced oxide fuels, and recovering them from high-level liquid waste [42]. The three processes make use of pyrochemical techniques in molten chlorides (LiCl-KCl) in contact with liquid metal (Cd or Bi) as shown in Figure II.10. The process is essentially based on a metal fuel cycle previously developed at the Argonne National Laboratory (ANL) [43]. The advantage is that it can be applied to either oxide or metal fuel, of any composition whether uranium only or mixed with plutonium, and to fuels of very high burn-up.

Figure II.8 **Schematic presentation of pyrochemical reprocessing for oxide fuel (RIAR)**

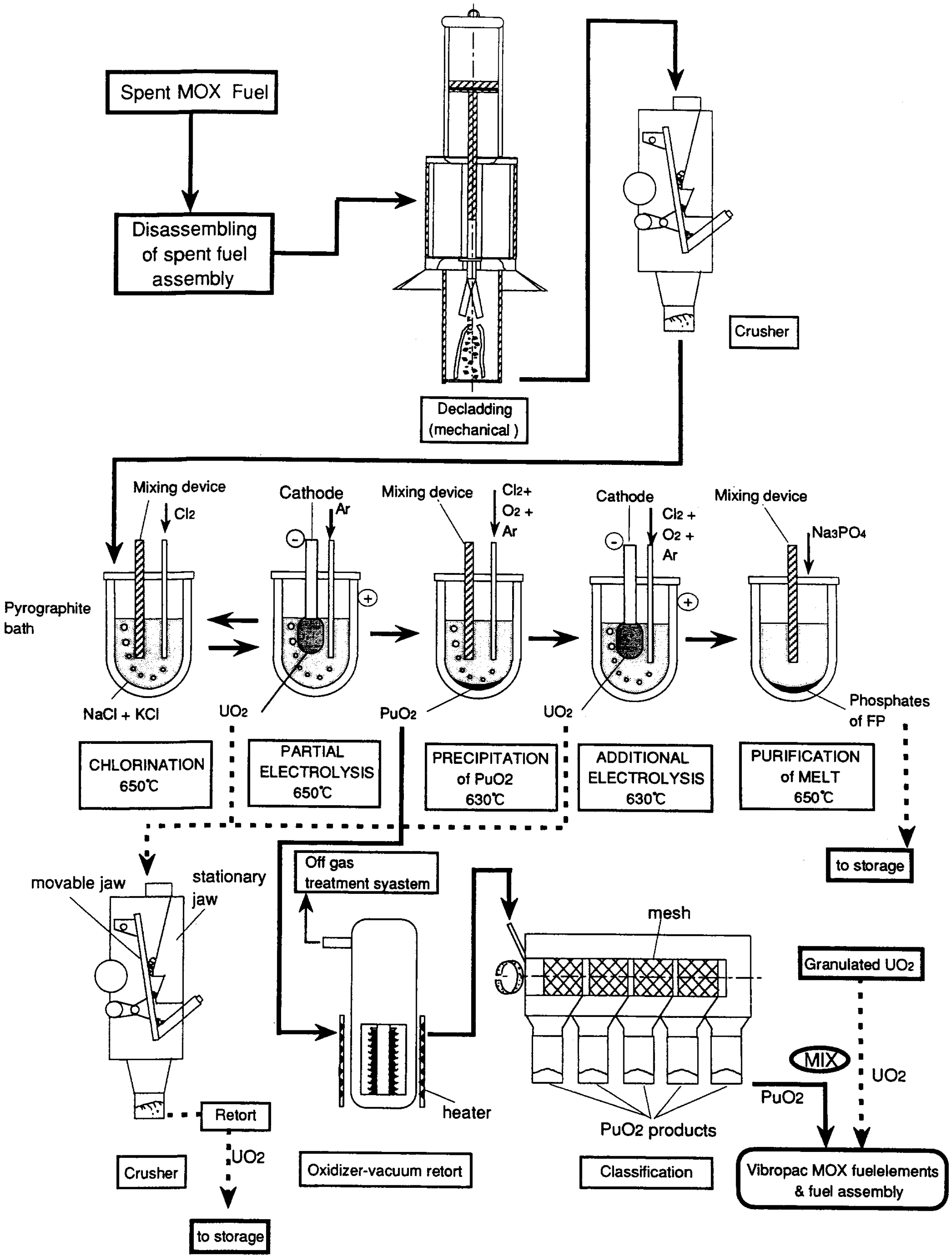
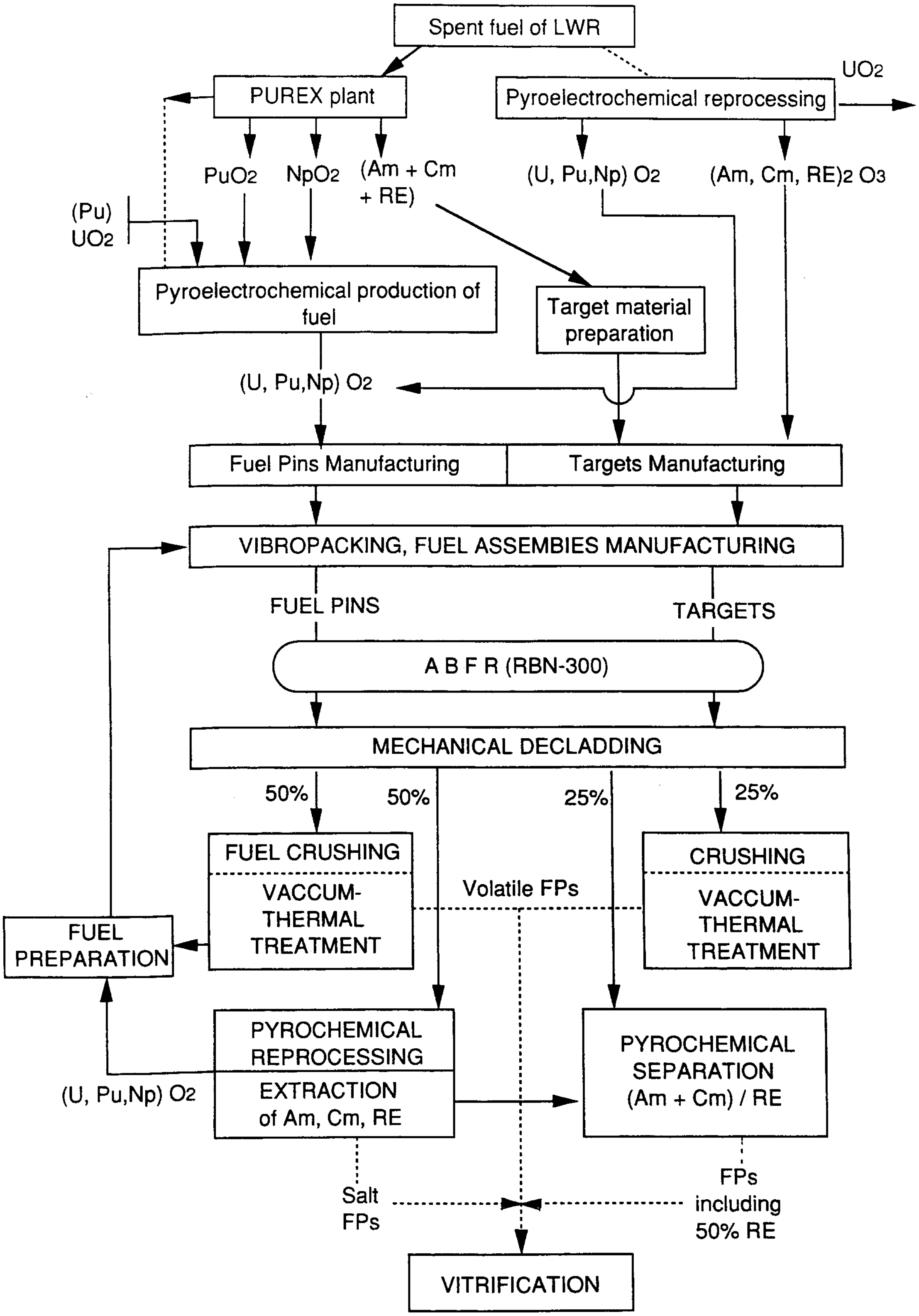
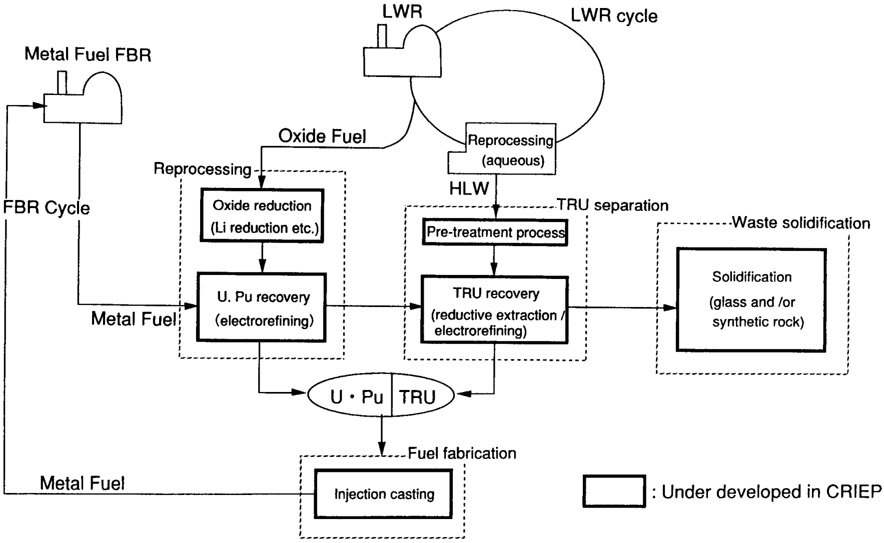


Figure II.9 **Fuel cycle actinide burner reactor (DOVITA Fuel Cycle)**



This recycling system can be operated flexibly by combining LWR and FBR cycles. Its main features are the electrolytic transfer of uranium and plutonium to solid and liquid double cathodes and recovery of transuranium elements from the salt by reductive extraction or electrorefining. Oxide fuels are first reduced to metals with lithium or calcium. Expected advantages are improved proliferation resistance, simplified criticality control and economically compact equipment. The disadvantage is lack of industrial experience with a high-temperature process using corrosive materials.

Figure II.10 **Actinide recycling system based on a pyrochemical processing of HLW and FR metal-fuel FR-irradiation**



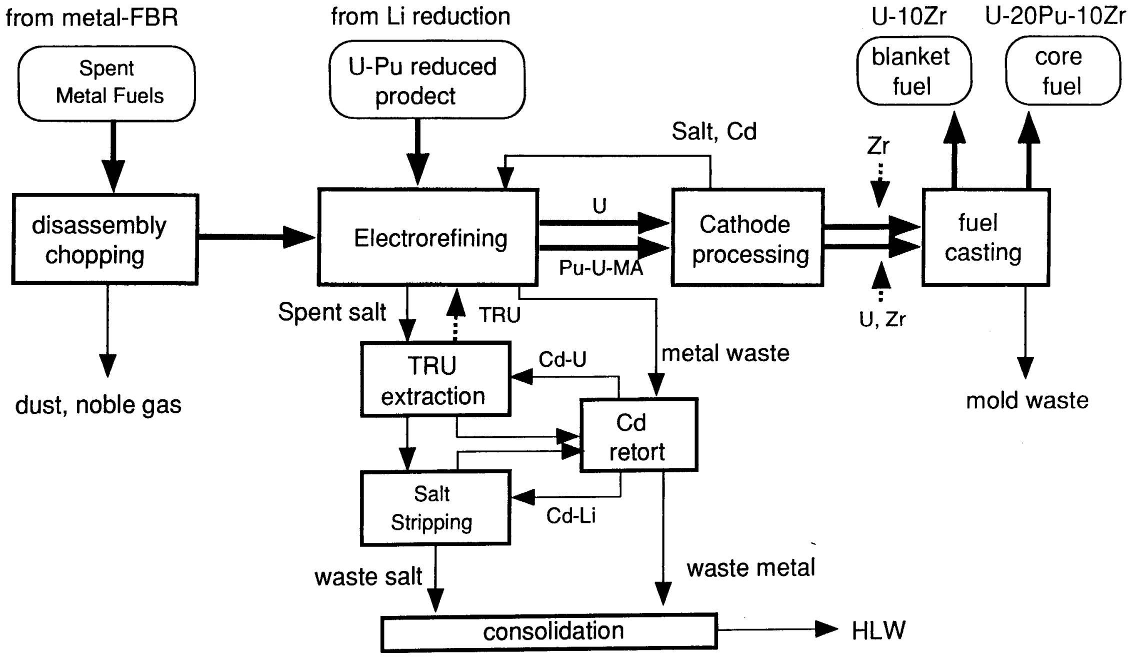
1.2.2.1 *Pyrochemical reprocessing of metal and oxide fuels*

The basic pyrochemical process developed at ANL [43] consists of anodic dissolution of spent fuel, partial recovery of uranium on a solid cathode, and electrolysis of plutonium with the remaining uranium into a liquid cadmium cathode, with an electrolyte of LiCl-KCl above 500°C [44]. The flow diagram for metal fuel is shown in Figure II.11. An anodic dissolution test on EBR-II fuel showed that most of the alloy was dissolved into the salt while a little cladding material remained as residue [45]. Recovery of uranium on the solid cathode has been thoroughly demonstrated with 10 kg deposited in a dendritic formation [46]. The condition of effective deposition on the solid cathode was investigated by changing the ratio of surface area of cathode to anode at CRIEPI [44].

Oxide fuels to be treated by this process must first be reduced to metal. ANL experimentally evaluated several methods, such as salt transport, the Zn-Mg process, Mg extraction and lithium reduction. The lithium process has the advantages of a lower operating temperature, less corrosive environment and small amounts of salt waste. Single element tests with uranium, plutonium and other elements, demonstrate the reduction to metallic form in LiCl or LiCl-KCl above 500°C, resulting in the formation of lithium oxide which will be electrolytically decomposed for recycling.

Multi-element tests of the whole process, preferably with real fuels, are going on to find out the technical feasibility of lithium reduction, proceeding to find the best operating conditions for pilot- and larger-scale trials at CRIEPI.

Figure II.11 **Flow sheet of pyro-reprocessing of spent fuel (ANL/CRIEPI)**



1.2.2.2 Pyrochemical separation of transuranic elements from high-level liquid waste

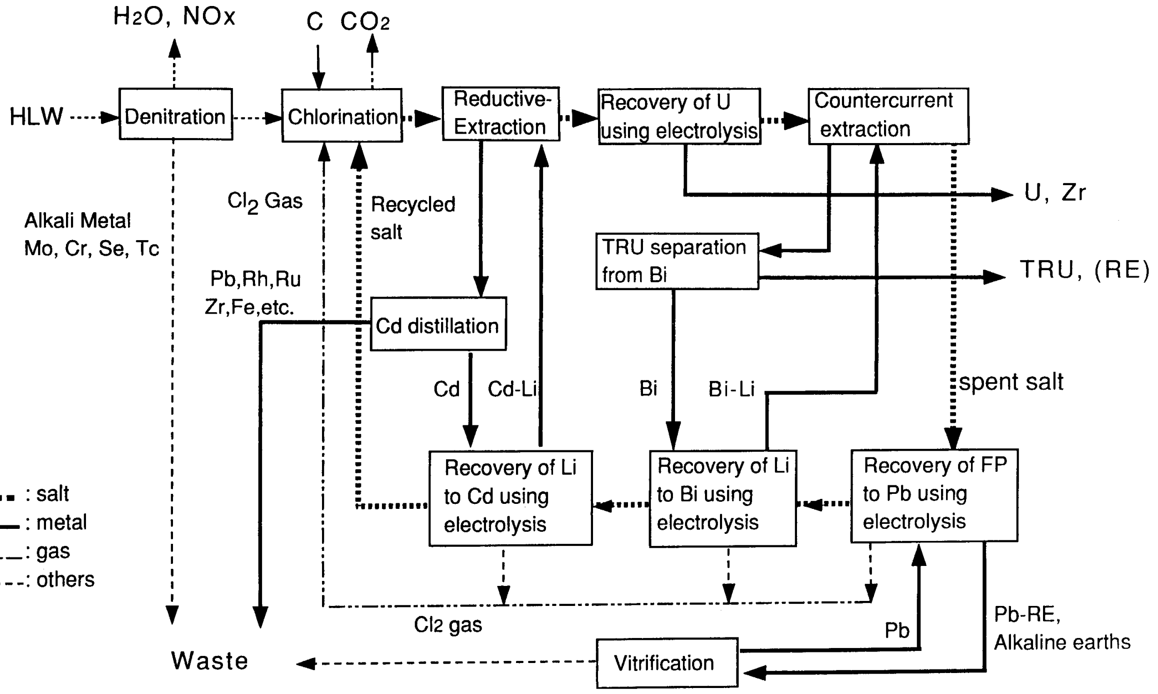
The process investigated at CRIEPI to recover transuranic elements from high-level PUREX wastes consists of denitration to oxides, chlorination, reductive extraction and electrorefining in a LiCl-KCl/Cd or LiCl-KCl/Bi system. After denitration and leaching with water to remove the soluble alkali-metal nitrates, the undissolved oxides (mainly of actinides, rare earths and transition metals) and platinum-group metals are converted into chlorides in a bed of LiCl-KCl at above 700°C.

The mixed chlorides are reductively extracted or electrorefined in contact with liquid cadmium or bismuth [43,47]. Thermodynamic data for actinides and rare earth elements in this system are needed to establish the separation process. The flowsheet illustrated in Figure II.12 is based on the results of small-scale tests.

Electrorefining may be used merely for a rough separation of uranium before counter-current reductive extraction, but can afford a more complete separation between transuranic and rare-earth elements. A small-scale experiment showed that over 99% of each actinide could be recovered from a simulated waste. The treatment of highly-active salt waste is also shown in the figure. The LiCl-KCl mixture can be recycled after purification, while the salts of fission products are electrolytically decomposed and converted to oxides for vitrification in borosilicate glass [48].

The waste produced throughout the process is expected to be minimal, since most of the materials (such as the eutectic salt, cadmium and bismuth metal, and chlorine) will be recycled. As an alternative treatment, waste might be solidified directly into an artificial rock such as zeolite or sodalite with high integrity and leach resistance [49]. Further technological assessment requires a pilot-scale demonstration with the full range of actinides.

Figure II.12 **Flow sheet of pyrochemical partitioning of TRUs from HLW (CRIEPI)**



1.2.3 *Condensed actinide-burner cycle: double-strata concept*

The Japan Atomic Energy Research Institute (JAERI) has proposed a P&T scheme based on a double-strata concept in which MAs from the familiar fuel cycle pass to an "actinide burner cycle" for total fission [50]. Here, concentrations of actinides are kept high in nitride fuels to be reprocessed pyrochemically. Nitrides have excellent properties, allowing (a) low fuel temperatures that reduce release of fission gas, and (b) a thinner cladding with consequently harder neutron spectrum than with oxide. Moreover, actinide mononitrides, unlike the metals, are expected to be mutually soluble.

The outline of the burner cycle is shown in Figure II.13:

- actinide salts from the first stratum are converted to mononitride microspheres by sol-gel techniques. A very high yield is expected.
- irradiated nitride fuel is reprocessed by a molten-salt electrorefining technique [51], basically the same as for metal fuels. The highly-enriched ^{15}N, necessary to minimise production of ^{14}C, is easily recovered and recycled.
- recovered metallic actinides are converted to nitride by direct reaction between liquid cadmium alloys and nitrogen [52].

In a recently devised alternative to this last step, called LINEX (Lithium Nitrate Extraction of Actinides), actinide nitrides are produced in a single step by addition of Li_3N to the molten salt [53]. It is obtained by direct reaction of Li metal with the ^{15}N evolved on dissolution of fuel in molten salt. Thus recycling of ^{15}N is also facilitated.

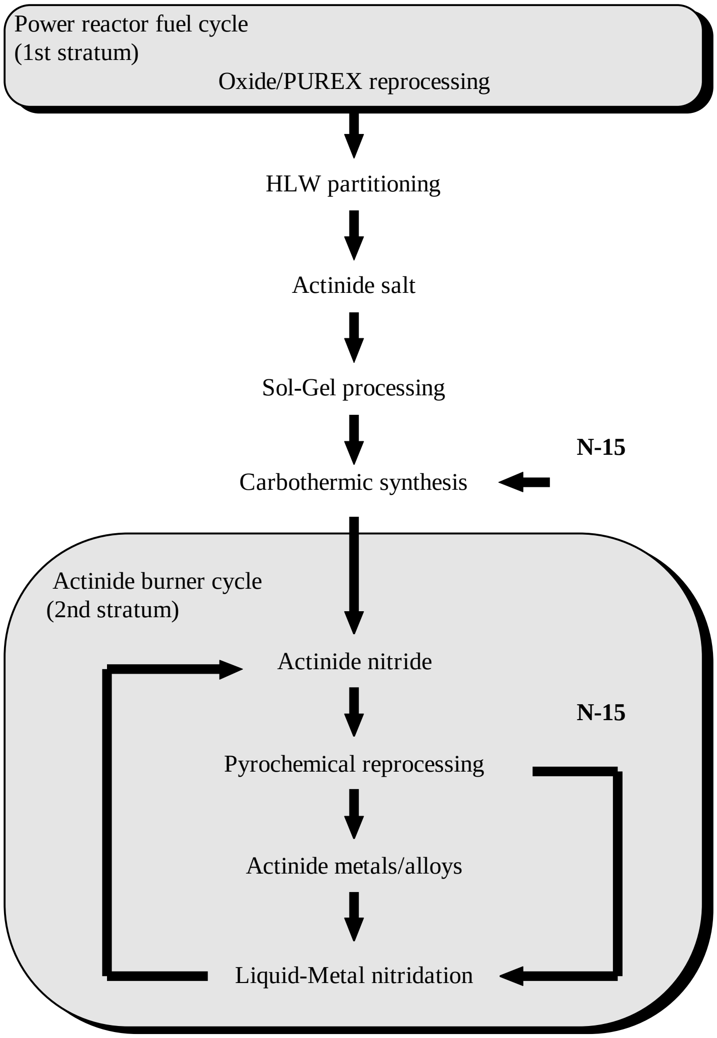
The metathetical reaction between Li_3N and actinide chlorides ($AnCl_3$) yields AnN and LiCl in the salt. Since the stability of actinide and lanthanide mononitrides varies relatively little from element to element, the nitrides are formed in the reverse order of stability in the corresponding halides. Thus most of the lanthanides remain in the salt until most of the actinides are separated as nitrides. Further addition of Li_3N then removes lanthanides from the salt if necessary.

The same reaction can also be used to remove lanthanides from the salts after other pyrochemical processes.

1.2.4 Conclusion

The pyrochemical process is particularly suited for processing high-burnup FR fuels and irradiated targets in order to shorten the cooling times in the processing cycles. The separation ability is presently limited to groups of elements and in order to achieve higher separation factors and element separation, multi-stage separation will have to be developed.

Figure II.13 **Actinide burner cycle with nitride/pyrochemical process**



2. TRANSMUTATION

2.1 Introduction

Nuclear power generation is inevitably accompanied by the formation of neptunium, plutonium and higher actinides from uranium (see Annex E). The long half-lives of some isotopes of these elements, and of a few fission products, give rise to concern about possible long-term radiological effects.

When plutonium is multi-recycled, the minor actinides will dominate the long-term radiotoxicity of the wastes. The reprocessing and separation processes give rise to a mixture of Am+Cm+lanthanides (or rare earths) which is difficult to further separate, because of the similarity of these elements' chemical properties. The impact of the separation performance on the americium transmutation should be investigated. Since reprocessing losses of plutonium are low (about 0.1%) compared to those expected for minor actinides, the latter will account for the major part of the long-term radiotoxicity of the wastes. In these conditions, the complete recycling of plutonium offers no advantage from the standpoint of reducing potential radiotoxicity, unless the minor actinides are also reduced with a view to minimise the radiotoxic inventory of the wastes to be stored.

Three minor actinide elements to be transmuted in reactors are considered: neptunium, americium and curium.

The activity of neptunium and americium is low enough to consider them for recycling in reactors without prior interim decay storage. Two options are available for transmutation: in the homogeneous mode, the element is mixed in a suitable chemical form with the standard reactor fuel; in the heterogeneous mode, the element is placed in the reactor separately from the fuel in a device known as a "target". The choice between these options depends on the behaviour of the particular nuclide in the reactor and in the fuel cycle.

Two other aspects of the minor actinides must be taken into account: the effect of their presence on reactor operation – primarily from a safety standpoint – and their transmutation yield. The principal core characteristics liable to be affected by the presence of actinides are the reactivity and the safety parameters (transient over-power and loss-of-coolant incidents).

The initial reactivity value is modified, as is the rate at which it diminishes. A positive value must be maintained throughout the reactor cycle. The initial fuel enrichment in fissionable isotopes (^{235}U or ^{239}Pu) or the absorber content of the core may be modified to compensate for the variations compared with the standard core resulting from the presence of minor actinides for incineration.

Recycling of the minor actinides (neptunium and americium) is possible in thermal reactors and in fast neutron reactors, either in homogeneous or heterogeneous mode. The mass balance shows the advantage of a fast neutron spectrum over thermal spectrum in allowing a higher burn-up to be reached.

This is confirmed by the radiotoxicity balance. In any case, incineration of americium generates a large amount of curium that must be processed to diminish the overall radiotoxicity of the waste. Moreover, the SUPERFACT experiments in the Phénix fast reactor revealed considerable helium production in targets containing americium, which could limit the permissible concentration.

Transmutation of pure curium in reactors is a highly problematic operation. Curium is the most intensely radioactive of the actinides for both neutron emission and α-activity which interfere with handling operations in fuel and target fabrication. In addition, the most abundant of its isotopes is ^{244}Cm, which decays with a half-life of 18 years to form ^{240}Pu and has a low fission cross-section which makes it relatively unsuitable for transmutation in a reactor. Separating the curium after removal from the reactor and placing it in interim storage for a sufficiently long period to allow significant radioactive decay (only 2% of the initial ^{244}Cm remains after a century) should be considered as one among the several options for handling curium. The daughter nuclides, mainly ^{240}Pu, could then be recycled. This solution, however, involves the interim storage of large quantities of a highly radioactive element, and will require further assessment from a safety standpoint in particular.

In addition to the minor actinides, three long-lived fission products were considered; technetium (^{99}Tc), iodine (^{129}I) and caesium (^{135}Cs). The ^{135}Cs is found only in small amounts. Caesium irradiation in reactors would be inefficient, as more ^{135}Cs would initially be formed by neutron capture from lower isotopes than would be eliminated. Isotopic separation would be necessary for transmutation of this element to be of any interest.

The destruction rate of ^{99}Tc and ^{129}I by neutron capture is low because of their low capture cross-sections and particular resonances, which make it necessary to recycle these elements in a high flux of appropriate spectrum.

Transmutation studies on long-lived radioactive wastes have been performed:

- to define acceptable recycling conditions, considering the effects of recycling on the core properties (effects on reactivity and safety parameters) and on the fuel cycle (radioactivity levels, neutron sources, and residual power);
- to assess the potential for radiotoxicity and mass reduction prior to disposal of long-lived radioactive waste from nuclear reactors;
- to identify the data required for fuel cycle studies (isotopic composition, mass flux).

In this systems study, the recycling of the minor actinides is considered in thermal reactors (standard PWRs loaded with UO_2 and MOX, High Moderating PWRs, etc.), in fast neutron reactors (oxide, metal and nitride fuels), and in dedicated systems (accelerator-driven systems, and MA burner reactors). The transmutation of fission products is also considered in thermal reactors, notably the heavy water reactors (CANDU), in thermal high flux reactors and in fast neutron reactors.

2.2 Target and fuel fabrication technology

2.2.1 Fuels for homogeneous recycling

2.2.1.1 Fabrication of fuel including neptunium

^{237}Np and particularly its daughter ^{233}Pa have a considerable γ emission requiring appropriate shielding in the powder blending section of the fabrication plant. In the remainder of the fuel plant, the effect is very small and can be neglected.

In principle, neptunium could be mixed with the standard UO_2 fuel of current PWR cores. However, the reference ^{235}U enrichment should be significantly increased, which is seen as a major penalty [54].

It is more advisable to mix NpO_2 with the usual MOX fuel $(U, Pu)O_2$. This has been done already for irradiation in fast reactors: first of all for the SUPERFACT experiment in Phénix, the fuel of which was manufactured by ITU Karlsruhe, and more recently in view of the NACRE experiment in Superphénix, the fuel of which was fabricated, with 2% Np, at the CFCA plant of Cogéma Cadarache.

In a MOX fuel factory, with respect to the pure PuO_2 reference case, the presence of NpO_2 does not affect the α and neutron emission but increases the γ source, due to ^{233}Pa (the daughter of ^{237}Np). The powder blending glove box should be protected by some 2 mm-thick Pb layers, to keep the external dose rates unchanged [55].

In case of a multiple recycling of ^{237}Np, ^{238}Pu is progressively built-up, and this additional source of neutrons (mostly from (α,n) reaction) and of heat affects further re-processing and refabrication steps. Np should therefore be irradiated preferably in fast reactors with a lower capture to fission ratio and consequently reduce ^{238}Pu yield from irradiation. On the other hand, a multiple recycle of ^{237}Np in fast reactors also increases the tiny ^{236}Pu fraction, and the radioactive chain ^{236}Pu - ^{232}U - ^{208}Tl brings a further emission of high-energy γ-rays, so that the 2 mm-thick Pb layer quoted above would become about 5 mm.

As a conclusion, Np recycling affects fuel refabrication in a UO_2 factory substantially but a MOX factory to a limited extent, so that the present MOX plants can afford it without major modification.

2.2.1.2 Fabrication of MOX fuel containing americium for LWR

For the implications of recycling, both plutonium and americium as MOX fuel in light water reactors have been considered, with reference to the current $(PuO_2\text{-}UO_2)$ fabrication of MOX fuel.

Recycling of plutonium and americium simultaneously in LWRs

It is assumed that not only plutonium, but also americium can be recovered from spent fuel reprocessing, and recycled in the form of MOX fuel (Pu+Am) in the same PWR under the same conditions. A recovery yield of 99.5% for Pu and 98% for Am has been assumed.

For the same burn-up as in the pure Pu recycling case, the computation has been repeated for the sum of plutonium and americium. The enrichments required in this Pu+Am case are still higher than the enrichments needed to recycle only the plutonium [56]. Recycling of americium together with plutonium in PWRs is limited due to the deterioration of the void reactivity coefficient

Fuel fabrication aspects of plutonium and americium recycling in PWRs

A study has been made on radiation dose protection and criticality safety in a MOX fuel refabrication plant.

As a reference case, the MOX fabrication plant of Belgonucléaire at Dessel [57] has been taken. It was already known that the front-end stages are the most critical ones with respect to handling operations on the pure oxide powders. Currently, PuO_2 powders from the reprocessors are taken from storage. Subsequently, these powders are introduced into a glove box to be milled and blended together with UO_2 powders to produce the so-called primary blend. Such dose intensive operations are followed by secondary blending, pressing and sintering, before the sintered pellets are put into fuel pin cladding: dilution first and canning afterwards lower the dose rates. Therefore, primary blending is retained as the most typical source of the dose rate due to handling operations.

Neutron and γ dose rates at a distance of 30 cm from the external glove box wall have been calculated for a given configuration of blending devices.

The dose rates at 30 cm from the glove box have been calculated for the reference PuO_2 powder (MOX 1; 7.3% Pu) and for the various powders, which correspond either to a second plutonium recycling step (MOX 2; 10.4% Pu) or to a first recycling of plutonium and americium (9.8%; MOX 1-Am).

A value of 20 mSv/h (2 mrem/h) is taken as a guiding value for these comparisons, although it is no real limit in the plant as the staff will not stay longer than needed near the glove box of primary blending, according to the ALARA principle.

The results [58] show that conditions for the MOX 2 fuel fabrication with 10.4% Pu are very similar to those of the MOX 1 with 7.3% Pu. There is indeed only a marginal increase in the main neutron source, which is induced by the α radiation from ^{238}Pu by (α,n) reactions. This favourable result is caused by the dilution of the plutonium from the MOX fuel with the plutonium from UO_2 fuel.

Addition of americium to the plutonium powder for its recycling would induce a strong increase of the γ dose (by a factor of 4.5). This increase could be mitigated by the addition of shielding with a layer of 25 mm of steel. It thus appears that dose rates could be controlled at the expense of extra shielding. Of course, the extra shielding would hinder the fabrication and would increase the cost of operations. Still the operations seem a feasible extension of the standard MOX fabrication conditions especially if one were to consider remote fabrication with automated processes.

2.2.1.3 Fabrication of MOX fuel containing MAs for fast reactor

The first experience of fabrication of oxide fuels containing high contents of Am and Np (up to 20% of Am) has been done by the Institute of Transuranium Elements Karlsruhe (ITU) during 1984-1986. The fabrication process used was the SOL-GEL one (GSP, Gel-Supported process) followed by pressing and sintering of the spherical particles in order to obtain pellets. These fuels have been irradiated in the reactor Phénix (SUPERFACT Experiment) [59].

In France, one complete subassembly has been fabricated successfully by Cogéma using industrial facilities in 1997, the fuel was (U, Pu)O_2 containing 5% of Np.

Currently, laboratory studies on fabrication of innovative fuels for MA transmutation are performed in the ATALANTE facility at Marcoule (CEA). The CERCER manufacturing process qualification is in progress based on the mechanical mixing of MgO and AmO_2 and granulation. The preliminary tests have been done during 1998. The pellets to be fabricated will be irradiated in Phénix (ECRIX experiments) in 2000.

A systematic programme has been planned in JNC for fabrication and investigation of irradiation behaviour of MOX containing MAs. Two fabrication methods, pellet-pressing and vibro-packing have been studied for neptunium-based fuel pins. The pellet type Np-based fuel will be fabricated at Tokai Works of JNC, and the fabrication of Np-based fuel by vibro-packing will be performed at PSI in collaboration with JNC. For Am-based fuels, the Alpha-Gamma Facility (AGF) at Oarai Engineering Center of JNC has already been adapted to fabricate MOX fuel pins containing Am at first and Am and Np afterwards. Remote assembling will be conducted in the Fuel Monitoring Facility (FMC). Both facilities will provide test beds for the post irradiation examination. Irradiation of Np- and Am-containing MOX fuel is planned in JOYO. In step with the JOYO MK-III schedule, the irradiation test will be initiated from around 2003.

2.2.1.4 Fabrication and irradiation of metal alloy fuel including MAs

Since the 1960s, Argonne National Laboratory (ANL) has been engaged in developing metal alloy fuels. The initially-developed U-5% fissium alloy fuels with 85% smear density for commercial use were found to fail by swelling. In the 1970s, by lowering smear density to 75% and by increasing the plenum gas volume, over 10 at% burn-up was attained on U-5 wt% fissium and U-10 wt% Zr alloy fuels. As plutonium-containing alloy, U-Pu-10 wt% Zr alloy was selected as having a high melting point and compatibility with stainless steel cladding. Since 1984, U-Pu-Zr alloy fuels have been further investigated as part of the IFR Programme [43]. More than a thousand fuel pins were fabricated by injection casting and irradiated in EBR-II, some of them to a maximum of 18 at% burn-up with cladding temperature <600°C and linear heat rate <500 W/s without failure [60].

High-level waste from the PUREX process has been converted to metallic form and the actinides together with residual uranium and plutonium recycled by the pyrochemical process proposed by CRIEPI. The products recovered contain equivalent amounts of REs to MAs, because of the similarity in the thermodynamic properties.

The multi component alloys U-Pu-Zr-5MA-5RE and U-Pu-Zr-2MA-2RE were selected by CRIEPI for irradiation study and metallographic characterisation. Properties related to reactor performance, and compatibility with stainless steel cladding and sodium coolant, have also been studied. The melting points of the alloys with minor actinides appear to be similar to those of reference alloy U-Pu-Zr. Similarly, no large difference in thermal conductivity was observed between U-Pu-Zr-5MA-5RE and U-Pu-Zr. A significant migration of Am and rare earths was observed in a temperature gradient above 700°C [61].

CRIEPI selected the same fuel alloys, with U-Pu-Zr as reference, for comparative irradiation in a fast reactor. The alloys with minor actinides are sandwiched between U-Pu-Zr pins in order to economise the MA-materials. The irradiation conditions will be chosen to obtain a fuel centre temperature not exceeding 750°C and a linear heat rating of maximum 400 W/cm. Irradiations are planned for a burn-up range of 1.5 to 6 at% and up to 10 at% as soon as difficulties in restarting the Phénix reactor have been overcome.

2.2.1.5 Fabrication of nitride fuel including MAs

Nitride has many advantageous properties as advanced fuel such as high-thermal conductivity, good FP retention, high heavy metal density and mutual solubility.

In 1970s and 1980s, laboratory studies on a fabrication process for mixed nitride fuels has been performed at CEA. The fuel was seen as the best advanced fuel for FR application mainly due to its higher breeding gain and its absence of fuel/sodium reaction in the case of clad failure. The process developed was the carbothermic reduction of mixed oxide in an atmosphere of nitrogen, followed by decomposition of higher nitrides to mononitride. This dry preparation process was the most capable for industrial applications. Nitride pellet fuel pins have been fabricated for irradiation test (NIMPHE programme) in the Phénix reactor. The ITU fabricated, at the same time, nitrides by sol-gel process for irradiation in the HFR reactor (NILOC experiments). The fabrication technology for mixed nitride fuel based on the experience of carbide fuel has been developed, in both the Paul Scherrer Institute in Switzerland [62] and the Bhabha Atomic Research Center in India [63].

During the past 10 years, research on fabrication technology for mixed uranium-plutonium nitride fuel has been performed at JAERI with a view to an advanced fuel cycle system. Nitride pellets have been fabricated and supplied for measurement of their characteristics and for irradiation tests [64]. PNC has also fabricated some nitride fuel pellets and measured the fuel characteristics.

Pellet fuel

In 1960s and 1970s, actinide mononitride was mainly synthesised by a hydriding-nitriding route from the actinide metal, but recently the conversion of oxide to the nitride by carbothermic reduction has been improved [65,66]. Uranium-plutonium mixed nitride and neptunium-bearing nitride have been synthesised by reduction of the dioxide with graphite, usually in a nitrogen-hydrogen mixture. Pellet-type fuel is fabricated by milling, compacting and sintering in a similar way to MOX fuel. No important problems appear to remain except in the enrichment and recycling of ^{15}N.

The use of ^{15}N would be preferable because of the massive formation of ^{14}C by the $^{14}N(n,p)^{14}C$ reaction. Both ^{15}N and ^{14}C would be difficult to retain during the nitride dissolution in the PUREX reprocessing, while they could be easily recovered during the pyrochemical reprocessing with fused salt electrorefining. In view of the cost for the enrichment of ^{15}N, the content of which is only 0.365% in natural nitrogen, the recycling of ^{15}N would be one of major issues for feasibility of nitride fuel.

Particle fuel

Remote fabrication from particles has advantages with materials of high radioactivity, so techniques for the fabrication of nitride particles by a sol-gel process have been developed [62-67]. Minor actinides separated from the high-level waste (HLW) as nitrates are converted into solids. The feed solution is prepared by mixing actinide nitrate solution, carbon powder, hexamethylenetetramine (HMTA) and urea. HMTA decomposes to form ammonia on heating to about 80-100°C, so gel particles are formed as a mixture of the actinide oxide and carbon, which is converted to mononitride by carbothermic reduction in N_2-H_2 mixture. The particles of pure UN with low oxygen and carbon contents have been successfully fabricated in Japan and Switzerland, and the method has also been applied to uranium-plutonium mixed nitride. The technology for the fabrication of dense and pure nitride particles would be a future R&D subject.

Targets with an inert matrix

Nitride is also suitable as target compound for burning americium by diluting it with inert materials such as ZrN and TiN. The fabrication of the solid solution of ZrN and UN, which is a substitute of AmN, has been studied by a sol-gel route [62]. Such fuel is also considered to be a candidate for plutonium burning in the CAPRA reactor [68].

2.2.2 Targets for heterogeneous recycling

2.2.2.1 Fabrication of AmO_2 target pins

A selection of inert-matrix material, actinide support alone (Am based), and compound materials, heterogeneous Am targets have been studied for Am transmutation [69]. The selection of possible candidates was based on a number of criteria concerning their neutronic, physical and chemical properties in relation to fabrication, performance and reprocessing.

The following classes of conceivable candidates were investigated:

- pure Am compounds: Am_2O_3, AmO_2, AmN and Am_2C_3 ceramics.
- ceramic solid solution materials: they are obtained by a chemical reaction between Am compound and the inert material. The final product forms a single phase which is physico-chemically different from that of the initial Am compound and from the inert material used.
- two-phase materials: are compounds of two distinct phases in which the two components keep their chemical form. In this case, Am compound is dispersed in the inert material, either in the form of fine particles or in the form of macro-particles.

Two categories are to be distinguished:

- the CERCER material (dispersion of a Am CERamic in an inert CERamic) of oxide, nitride or carbide;
- the CERMET material (dispersion of an Am CERamic in an inert METal).

It was found that MgO, Y_2O_3, Al_2O_3, $MgAl_2O_4$, and $Y_3Al_5O_{12}$ are good potential candidates as inert matrices for fast reactor.

CERCER composite materials with Am, MgO-AmO_{2-x} [70] have been fabricated in ITU, after sintering the pellets are a two-phases mixture. $MgAl_2O_4$-AmO_{2-x} pellets containing 10% ^{241}Am by weight have been fabricated by the impregnation method by ITU. A reaction between the Am oxide and $MgAl_2O_4$ occurs to form a new compound. Taking into account the difficulties in preparing and controlling Am oxides of well-defined composition, it may be advantageous to use a dispersion of a (M, Am)O_2, (M, Am)$_2O_3$ type solid-solution in $MgAl_2O_4$ [71].

On the basis of the study, it is not yet possible to select a single U-free material as the "best" target. Interesting candidate materials are (Am, Zr)N solid solution, MgO-AmO_{2-x}, CERCER and CERMET Am composites.

Comparative Dose Rate Values [72]

Storage of powders

Dose rates have been calculated for pure PuO_2 and for AmO_2 powders. The γ-dose rate due to AmO_2 increases by a factor of 185 compared to PuO_2, and the neutron dose by a factor of 3. Compensating for these increases needs the addition of 7 cm lead to the shielding.

Handling and transportation of Pins

The γ-dose rate of AmO_2 is 2 780 times higher than that of conventional MOX fuel. For neutrons, the ratio is about 7. Compensation needs typically the addition of about 4 cm lead and 4 cm resin to the shielding is required, and compared with plutonium only half as many pins must be transported at the same time.

2.2.2.2 *Effect of curium on target fabrication*

For one tonne of uranium loaded in a PWR, approximately 85 g of Cm are formed at the end of a 45 GWd/t irradiation; the ratio Cm/Pu is about 0.8%. The isotopic composition of this curium fraction (just after irradiation) is as follows:

$$^{242}Cm : ^{243}Cm : ^{244}Cm : ^{245}Cm : ^{246}Cm = 19 : 1 : 62 : 3 : 0.2.$$

The major effect of curium on the storage of nuclear wastes in the long-term is that the decay of ^{244}Cm ($T_{1/2}$ = 18 years) adds some 2.5% to the ^{240}Pu quantities.

Both isotopes ^{242}Cm and ^{244}Cm are intense neutron sources. In case of a refabrication of fuel or targets five years after core discharge, ^{242}Cm has mostly decayed while ^{244}Cm remains the predominant neutron source.

The addition of Cm to the MOX fuel would increase the neutron dose rates around the blending glove box by about a factor 100. This would require such thick protection layers (0.3 to 1 m polyethylene shields) as to exclude its use in present MOX fuel production plants. Curium targets containing gram quantities have been produced in laboratory conditions at ITU Karlsruhe.

2.2.2.3 *Fabrication of targets for transmutation of fission products*

The fabrication and irradiation of targets for transmutation of fission products has been studied by the EFTTRA group in Europe [73-75]. The metallic form was selected for Tc. Rods of the metal reduced from ammonium pertechnetate were prepared by arc melting, and casting in a water-cooled copper mould. The targets have been irradiated in a thermal neutron flux in the High Flux Reactor (HFR) in Petten, in a first test to a transmutation yield of about 6% and in a second test to a yield of about 20%. Post-irradiation examinations of the first test have shown that technetium metal has a good irradiation behaviour, showing negligible swelling and no microstructural changes [74]. The measured transmutation yields were found to be in reasonable agreement with those obtained from post-test calculations [76].

For the transmutation tests of iodine, metal iodides containing natural ^{127}I were considered by the EFTTRA group [75]. Cerium iodide (CeI_3), lead diiodide (PbI_2) and sodium iodide (NaI) were selected and powders of the pure compounds, enclosed in stainless steel capsules, have been irradiated in the HFR. Both CeI_3 and NaI showed a reasonably good irradiation behaviour but it was noticed that the fabrication of pure CeI_3 (i.e. water-free) is a troublesome process. NaI was therefore considered the most promising target material. Further work in this field is going on, especially to clarify the discrepancies between measured and calculated transmutation rates.

2.3 Transmutation of minor actinides

2.3.1 Physics and methods of transmutation

2.3.1.1 Physics

When heavy nuclei are placed in a reactor, they are subjected to a neutron flux which induces two main types of reaction:

- fission, most products of which are β and/or γ emitters with much shorter average half-lives than the initial heavy nuclei. These reactions are preponderant in some nuclei: ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu, ^{242m}Am and ^{245}Cm. This type of reaction is called incineration.
- transmutation by neutron capture (generally followed by β decay) leads to heavier nuclides whose half-lives and α activities are accordingly different. These transmutation reactions are preponderant on all the heavy nuclei other than those mentioned above.

A preliminary analysis of these mechanisms leads to a number of important conclusions:

- the incineration rate will remain low, in comparison with the transmutation rate, and it will generally be impossible to reduce significantly the heavy nuclei by a single recycle to the reactor, unless sufficiently long residence times are applied for the successive transmutations culminating in fissile nuclei which are destroyed by incineration. In most cores multiple recycling will be necessary to achieve quantitative elimination.
- during this multi-recycling, the proportion of heavier nuclei (Am, Cm and even Bk etc.), produced by successive captures, is significantly increased. The composition of the fuel thus changes substantially both with regard to chemical composition and to the isotope fractions for each element.
- since plutonium is the principal contributor to radiotoxicity, it is necessary to start by recycling plutonium alone, followed by the minor actinides, americium, curium and neptunium.
- multi-recycling of the MAs is feasible in various types of reactor (BWR, PWR, FBR) in various modes: homogeneous, in which the MAs are diluted in the fuel with low contents, and heterogeneous, in which they are concentrated in targets with high contents.

In a given reactor system, the different characteristics necessary to achieve equilibrium between actinide production and consumption can be determined (proportion of the different reactors, recycling mode, actinide content). These studies were conducted by a two-step approach:

- in the first step, the capacity of a single type of reactor to incinerate and transmute the plutonium and long-lived nuclear wastes was characterised;
- in a second step, the results of the first phase were used to draw-up feasible scenarios involving a group of different reactors producing and consuming plutonium and minor actinides.

Table II.3 compares the mean actinide cross-sections of capture σ_c and fission σ_f in a thermal reactor and in a fast breeder reactor. It shows that the ratio, α, is higher in a thermal spectrum than in a fast spectrum. Hence, a fast spectrum is preferable because it promotes fission.

Table II.3 **Mean cross-sections of actinides**

Nuclide	f**: PWR thermal reactor**			f**: FBR fast reactor**		
	$\bar{\sigma}_f(b)^{a)}$	$\bar{\sigma}_c(b)^{a)}$	$\alpha^{b)}$	$\bar{\sigma}_f(b)^{a)}$	$\bar{\sigma}_c(b)^{a)}$	$\alpha^{b)}$
^{235}U	38.8	8.7	0.22	1.98	0.57	0.29
^{237}Np	0.52	33	63	0.32	1.7	5.3
^{238}Np	134	13.6	0.1	3.6	0.2	0.05
^{238}Pu	2.4	27.7	12	1.1	0.58	0.53
^{239}Pu	102	58.7	0.58	1.86	0.56	0.3
^{240}Pu	0.5	110.6	221	0.36	0.57	1.58
^{241}Pu	94.8	36.7	0.38	2.49	0.47	0.19
^{242}Pu	0.43	29	67	0.23	0.44	1.9
^{241}Am	1.1	110	100	0.27	2.0	7.4
^{242}Am	159	301	1.9	3.2	0.6	0.19
^{242m}Am	595	137	0.23	3.3	0.6	0.18
^{243}Am	0.44	49	111	0.21	1.8	8.57
^{242}Cm	1.14	4.5	3.9	0.58	1.0	1.7
^{243}Cm	88	14	0.16	7.2	1.0	0.14
^{244}Cm	1.0	16	16	0.42	0.6	1.4
^{245}Cm	116	17	0.15	5.1	0.9	0.18

a) average cross-section,

b) $a = \overline{s}_c / \overline{s}_f$.

Destruction of the long-lived elements by capture or by fission requires neutrons. The following was evaluated to determine whether sufficient neutrons are available in the reactors.

- The neutron surplus of each type of reactor defined as the difference between neutron production by fission and the losses by capture in the structural materials and absorbents and by leakage from the core. The estimates show that, in thermal reactors, this surplus is small (0.1 neutron/fission for a PWR with 6% enrichment in ^{235}U) and that it is about 0.4 to 0.6 neutron/fission in fast breeder reactors. Hence a fast spectrum is more favourable from this standpoint.

- The neutron consumption of the recycled nuclei is defined as the number of neutrons required to pass from the initial nucleus to stable nuclei. The calculation accounts for the probabilities of capture, fission, decay, and the number of neutrons involved in each case. Table II.4 gives a number of values obtained in this way. They show that it is always possible to incinerate the minor actinides in a fast spectrum (they also produce neutrons), whereas, in a thermal spectrum, the neutron surplus is not always sufficient.

Table II.4 **Neutron consumption normalised to 1 fission (*)**

Nuclide or element	**Thermal spectrum** f = 10^{14} n/cm^2·s	**Fast spectrum** f = 10^{15} n/cm^2·s
^{237}Np	1.12	-0.59
^{241}Am	1.12	-0.62
^{243}Am	0.82	-2.51
^{244}Cm	-0.15	-1.39
^{245}Cm	-1.48	-2.51
Pu unloaded from a standard PWR	-0.2	-1.1
Long-lived fission products		
without isotopic separation	2	
with isotopic separation	0.3 to 0.5	

(*) negative values correspond to neutron production

As for long-lived fission products, there are not enough neutrons to transmute them in the reactor, even if isotopic separation were possible. This is why hybrid systems have been considered, combining a particle accelerator and a sub-critical multiplier medium, which would offer an additional source of neutrons.

2.3.1.2 *Methods*

The recycling of the minor actinides in fission reactors is mainly considered in two ways:

- a so-called homogeneous method in which the recycled elements are mixed with the conventional fuel and are hence subjected to the same irradiation conditions;
- a so-called heterogeneous method in which the minor actinides are separated from the standard fuel and placed in specific target sub-assemblies or pins and managed independently from the standard fuel.

In homogeneous mode, the impact on the physical properties of the core due to the introduction of minor actinides like neptunium and americium is roughly proportional to the actinide content initially present.

Generally speaking, for fast reactors and for PWRs, the following is observed:

- positive effects, such as a reduction in the loss of reactivity in the cycle;
- negative effects, such as:
    - deterioration of the fuel temperature coefficient;

- deterioration of the effectiveness of absorbents (control rods, soluble boron);
- an increase in the reactivity effect associated with the coolant void reactivity coefficient.

For thermal reactors, it is also necessary to over-enrich the fuel, which is a heavy economic penalty for a UO_2 fuel.

For thermal reactors, the influence of the minor actinides on the safety parameters depends also on the moderation ratio. The variations are less pronounced in the case of the HMR (high moderation reactor) so that a slightly higher minor actinide content can be accepted.

In heterogeneous mode, if the targets are placed in the core, the consequences are similar to those in homogeneous mode. However if the targets are placed at the periphery of the core, the impact on the physical properties of the core remains slight if the actinide concentration has been fixed so as to limit the power release during irradiation to a value compatible with the cooling possibilities in order to avoid local power peaking.

The effectiveness of transmutation is characterised by three values which are used in the Tables II.5 to II.9.

- depletion/consumption:

$$\frac{\text{(initial mass – final mass) of MA}}{\text{initial mass of MA}}$$

- fissioned fraction rate:

$$\frac{\text{mass of fissioned MA}}{\text{initial mass of MA}}$$

- specific consumption:

$$\frac{\text{(initial mass – final mass) of MA}}{\text{energy produced by the reactor}}$$

2.3.2 Transmutation of minor actinides in thermal reactors

2.3.2.1 Present day PWR reactor [77-79]

Homogeneous recycling of minor actinides in UO_2-fuelled PWRs

The Np and Am recycling was studied in a N4 type PWR, rated at 1 470 MWe, with UO_2 fuel enriched at 4% ^{235}U, average burn-up of 47.5 GWd/tHM and fuel management of 1/5.

The irradiated fuel is cooled for 5 years before reprocessing. The minor actinides (^{237}Np or Am) are mixed with the UO_2 fuel at a content between 0.5% and 5%. Then, the manufactured assemblies are stored for 2 years before use.

The introduction of minor actinides, homogeneously mixed in the fuel, induces a reduction both of the initial reactivity, caused by neutron absorption capacity, and of the loss of reactivity over the cycle, due to the generation of more reactive isotopes resulting from their transmutation. To keep the cycle management unchanged, these effects are compensated by over-enriching the fuel and modifying the boron content.

In the presence of minor actinides, whether Am or Np, the fuel temperature coefficient decreases by about 10%.

The moderator temperature coefficient is reduced to some extent by the addition of Np, or more particularly by Am, because these minor actinides have resonances, greater for americium than for neptunium, at energies under 6.7 eV (the first resonance of ^{238}U). The spectrum hardening due to the temperature and density variation thus increases the reactivity with respect to the reference case.

The soluble boron efficiency decreases with the use of thermal neutrons absorbent isotopes, more in the case of americium than in the case of neptunium. Although increased by the presence of MAs, the impact of a total voiding of the moderator remained very clearly negative.

The MA content of the fuel must be restricted on safety grounds.

Homogeneous recycling of minor actinides in MOX-fuelled PWRs

This recycling is considered in comparison with the N4 reactor using MOX fuel enriched to 9% Pu. The 1% americium or neptunium recycling requires increases in the initial Pu contents of 3.5% and 3%, respectively.

The net production of MAs in a thermal reactor is less with some MAs initially present in the fuel than without them.

Unlike the operation of UO_2-fuelled N4 PWR, the MA recycling results in a less negative moderator temperature coefficient (-67 pcm/°C for the reference case, -50 pcm/°C for the 1% Am recycling and - 53 pcm/°C for the 1% Np recycling).

The initial plutonium content is very close to the maximum content allowed by its effect on the void coefficient. Then the allowed initial MA content, in the case of recycling in a MOX-fuelled N4 PWR, must be limited to less than 1%.

Table II.5 gives the calculated results of homogeneous recycling of the americium and neptunium in two thermal reactors with moderation ratios 2 and 3 (see definition in Annex G).

Table II.5 **Mass balances for homogeneous recycling in thermal reactors**

		PWR-UOX* moderation ratio 2	PWR-MOX** moderation ratio 2	PWR-MOX*** moderation ratio 3
Burn-up (GWd/t)		47.5	47.5	47.5
Initial minor actinide content (%) (mass of heavy metal)		1	1	1
Np	Initial mass (kg)	1 104	1 104	1 104
	Specific consumption (kg/TWhe)	12	10	11
	Depletion (% of initial mass)	53	43	45
Am	Initial mass (kg)	1 104	1 104	1 104
	Specific consumption (kg/TWhe)	19	5	10
	Depletion (% of initial mass)	83	20	42

* ^{235}U enrichment of 5.5% to recycle Np, or Am (standard enrichment 4.5%)

** Pu enrichment of 12% to recycle Np and 12.5% for Am (standard enrichment 9%)

*** Pu enrichment of 7.7% to recycle Np, 8.2% for Am (standard enrichment 5.7%)

Calculation of the effect of lanthanides on homogeneous recycling of minor actinides [79]

The reprocessing and separation processes give rise to an Am+Cm+lanthanides (or rare earths) mixture which is difficult to further separate, because of the similarity of these elements' chemical properties. In these calculations it has been supposed that Am and Cm would not be separated, but that in any case the REs have to be removed to a predetermined extent in order not to affect the reactivity balance in a thermal reactor.

The effects on the recycling of a mixture (Am+Cm+RE) with a 1% initial content in a MOX-fuelled N4 reactor was studied. The decontamination factor (DF) varies from 1 (natural mixture) to 1 000 (mixture practically without lanthanide taken as reference).

When DF varies from 1 to 1 000, the reactivity balance varies by 49%, the fuel temperature coefficient changes only slightly, the boron efficiency decreases by 19% at the end-of-life. The moderator temperature coefficient becomes less negative with a variation of 13% at end-of-life thanks to the presence of MAs which hardens the spectrum and makes this coefficient less sensitive to the moderator density variations. The global voiding effect gets less negative (it is even positive at the beginning of life for DF = 1 000). In order to limit the adverse effect of the lanthanides on the incinerated mass to less than 10%, there should be no more than 1% of RE in the mixture (Am+Cm+RE) resulting from the reprocessing of a UO_2 fuel, and this requires a minimum decontamination factor of 640, which remains a quite high value.

Heterogeneous recycling of minor actinides in UO_2-fuelled PWRs

The targets are made of neptunium or americium oxide mixed with an inert matrix. The concentrations are chosen to limit the effects on both reactor and the fuel cycle. The matrix material must be stable under irradiation, fairly compatible with water at 300°C (in case of clad failure), with satisfactory thermal properties (good conductivity, high melting point) and suitable for reprocessing. In consideration of these requirements, the theoretical studies were first conducted with Al_2O_3.

To study the influence of recycling conditions, standard 900 MWe PWRs supplied with UO_2 fuel at 3.7% ^{235}U enrichment and with 4-batch management were considered as reference. The recycled actinides are supposed to be contained in 24-rod clusters loaded in the guide tubes of some fresh assemblies and irradiated for a few annual cycles.

In the case of neptunium, initial target contents from 5% to 100% were considered. The results show that the initial NpO_2 content has a significant effect: for a one year irradiation period, the neptunium consumption increases from 10% to 25% as the initial content decreases from 100% to 5%.

In the case of americium, large radius rods (0.413 cm) were considered, with initial AmO_2 contents between 20 and 70%. The content was purposely limited to avoid difficulties at manufacture (dose rate) and after irradiation (helium (He) production emphasised by the SUPERFACT experiment). A comparison shows that for identical initial contents americium transmutation is much faster than that of neptunium. This result, already noticed in the study of homogeneous recycling, results from the more favourable neutron characteristics of americium. The total americium consumption considerably depends on the initial AmO_2 content. For a one-year-irradiation period, it decreases from 41% to 17% as the initial content increases from 20% to 70%.

The neutron linear heat rating in the target increases from a few W/cm at the beginning of the cycle to 138 W/cm (Np) and 130 W/cm (Am) at the end of irradiation. In the determination of the maximum local power in the target, the 3-dimensional shape factor of the assembly must also be taken into account.

Table II.6 shows the calculated transmutation performance in heterogeneous recycling mode for two cases: recycle of targets placed at the periphery of a PWR N4, and recycle of targets placed in the thimble guides of a PWR 900 (TIGRE). The second case is different from the first in that the targets undergo the same irradiation as the fuel. The initial masses involved are different because of the number of possible locations and the permissible contents, which are limited by the power release during irradiation. The irradiation times are related to the currently permissible irradiation damage rates.

Multiple heterogeneous recycling of minor actinide targets in UO_2-fuelled PWRs

Achieving a sufficiently high incineration ratio is not possible with once-through recycling, and so the targets must be recycled. Since the production of plutonium in the targets is significant, the operation is beneficial only if the plutonium is also destroyed. For the same reasons as in the case of homogenous recycling, curium recycling is not considered.

The neptunium or americium is recycled in assemblies with target pins (with a 70% content for Np or a 30% content for Am). The ^{235}U enrichment of fuel assemblies is 4.5%.

After a 4 year irradiation in a standard PWR, the target pins are reprocessed to eliminate the fission products and curium which are disposed of with the losses (10% for Np and Am, 0.3% for plutonium). Neptunium, americium andplutonium are recycled with adding of minor actinides recovered from the fuel to keep a fissile material content of about 3%. The time for the reprocessing and manufacture is 2 years. 12×24 targets are loaded-unloaded each year and are irradiated for three successive campaigns of each 43 GWd/tHM.

Table II.6 **Mass balances for heterogeneous recycling in thermal reactors**

		PWR-UO_2	**PWR 900 (TIGRE)**
Irradiation time EFPD(*)		2460	1120
Np	Content (%) (mass)	20	70
	Initial mass (kg)	736	236
	Specific consumption (kg/TWhe)	4.2	15
	Depletion (% of initial mass)	42	38
	Fissioned fraction (% of initial mass)	7	3
	Fissioned mass (kg)	52	7
Am	Content (%) (mass)	20	30
	Initial mass (kg)	726	71
	Specific consumption (kg/TWhe)	5.8	8.6
	Depletion (% of initial mass)	58	74
	Fissioned fraction (% of initial mass)	10	13
	Fissioned mass (kg)	73	9

(*) Effective Full Power Days

The results from these calculations show a relative stabilisation of performances between the second and third campaign. The annual neptunium consumption decreases from 37% to 27% between the first and third irradiation campaign because of the increase in the neptunium content in the targets. During the third campaign, the annual neptunium consumption of 166 kg gives rise to 140 kg of plutonium. The fissile Pu content in the total plutonium is 15.7% at the end of the irradiation period.

The annual americium consumption at equilibrium is 60%. As the resulting plutonium is recycled, the results are similar to those of neptunium except for a production of curium (of about 20% of the loaded americium amount) which is directly disposed of.

2.3.2.2 Molten salt reactor

The preliminary design of a TRU burner utilising a molten salt fuel and a graphite moderator has been performed. The molten salt reactor will be able to burn transuranic material at a rate of 1.2 kg/MWe-y [80]. The transuranic inventory of the molten salt reactor at a thermal power of 1 600 MWth is about 700 kg. Therefore, the time to reduce an amount equal to this transuranic inventory is about 10 years.

2.3.3 ***Transmutation of minor actinides in fast reactors***

2.3.3.1 MOX-fuelled fast reactor

Homogeneous recycling of minor actinides in fast reactor [77]

Np and Am recycling was studied in a large FBR core (see Figure II.14 (a)), for example, EFR (European Fast Reactor with a 1 500 MWe nominal power rating) type supplied with mixed oxide fuel. The plutonium isotopic composition in the fuel corresponds to the plutonium vector in a standard PWR UO_2 fuel irradiated at 33 GWd/tHM.

The parametric study was conducted for 2%, 5% and 10% contents of ^{237}Np or ^{241}Am, and for a 5% content of a mixture with equal proportions of ^{237}Np and ^{241}Am.

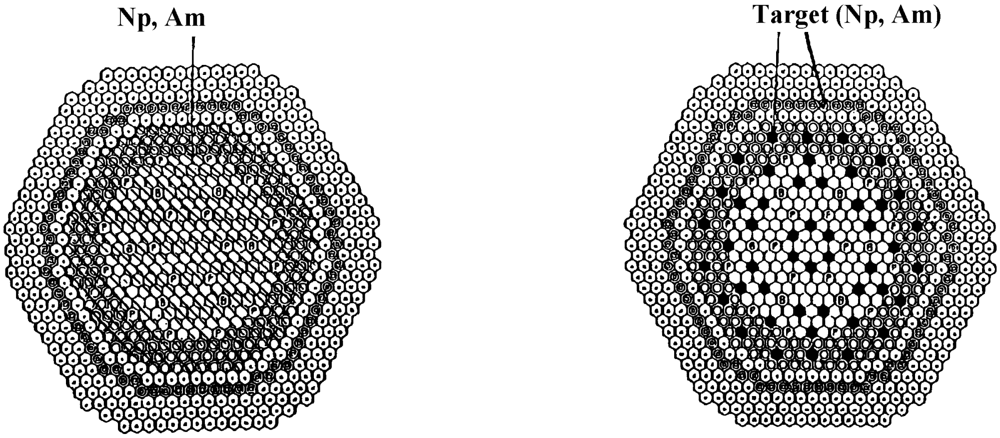
Multiple recycling of TRU components from reprocessed spent LWR-MOX fuel was investigated to assess the incineration capabilities of a FBuR [81].

Effects on core reactivity

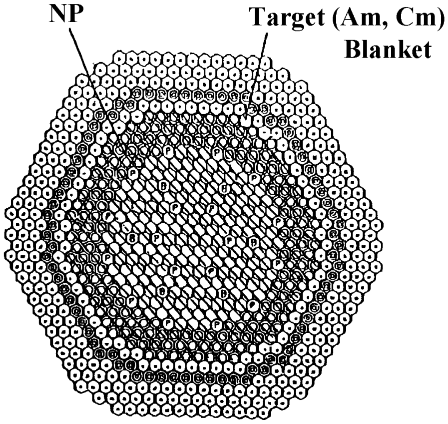
These effects are roughly equivalent for americium and neptunium. The introduction of americium and neptunium instead of ^{238}U causes:

- a reduction of the initial core reactivity due to the high minor actinide capture rate;
- a reduction of the reactivity loss over the cycle due to the transmutation of ^{237}Np and ^{241}Am into more fissile isotopes. The effect is more marked for ^{241}Am because of its partial transformation into highly fissile ^{242m}Am.

Figure II.14 **MA-loading methods in fast reactor**



Homogeneous (a) **Heterogeneous (b)**



Mixed (c)

For an initial minor actinide content of 1%, the combination of these two effects leads to a reduction of about 2% of the initial enrichment in plutonium, in order to obtain the same reactivity at end of cycle as in the reference case without minor actinides:

- a reduction of the absorber negative reactivity due to the spectrum hardening;
- a reduction of the effective β value due to the lower delayed neutron yields for ^{237}Np and ^{241}Am than for ^{238}U (factor of approximately 5), a very significant drop in the Doppler effect (approximately 10% for an initial minor actinide content of 1%) due to the spectrum hardening and to the depopulation of the energy range corresponding to ^{238}U resonance caused through a strong absorption by ^{237}Np and ^{241}Am;
- a significant increase in the reactivity effect resulting from ^{237}Np and ^{241}Am tends to amplify the variation of absorption during voiding.

However, as regards sodium voiding, the most restrictive situation corresponds to the end-of-life configuration, since the void effect coefficient increases throughout the cycle due to the gradual accumulation of fission products. Conversely, the penalty due to the presence of minor actinides is highest at the beginning of the cycle since their concentration decreases afterwards.

So, the initial content of minor actinides is limited by their influence on the Doppler effect and on the sodium void effect which can create the difficulties in reactivity control on coolant voiding, specially in a large core. In large cores however, penalties resulting from these effects can be limited by a suitable design:

- preferential disposition of actinides in the outer part of the core in order to limit the effect of sodium voiding;
- moderator introduction in the core in order to reduce the voiding effect and simultaneously increase the Doppler effect.

Mass balances

Table II.7 gives the performance obtained by homogeneous recycling of the americium and neptunium in an EFR type fast breeder reactor.

The incineration ratio of ^{237}Np does not vary either with its initial content nor with the size of the reactor.

On the other hand, the incineration ratio of ^{241}Am is closely related to the initial content as this isotope is produced in the core by radioactive decay of ^{241}Pu. The ratio is similar to that of ^{237}Np (approximately 50%) for a 5% initial content, but decreases significantly with a reduced initial content. The overall incineration ratios for all the minor actinides are lower, and the difference between ^{237}Np and ^{241}Am is no longer significant.

The ^{238}Pu fraction in the total plutonium is approximately 4.5% for the EFR, with a 2% initial content of minor actinide (^{237}Np or ^{241}Am). As initial minor actinide contents increase, the proportion of ^{238}Pu also increases. Solvent radiolysis problems may occur at reprocessing if the ^{238}Pu content of the irradiated fuel exceeds 5%. The initial content of minor actinides must, therefore, be limited to 2.5% in an EFR type core.

In the case of a 1 000 MWe-class FR core with mixed oxide fuel, MA transmutation has no serious drawbacks in terms of core performance, provided that the homogeneous loading method can be employed with a small ratio of MAs to fuel (~5 wt%) [81]. Since a 1 000 MWe-class LWR produces about 26 kg of MAs per year, a MOX fuel fast reactor with 5 wt% MA loading can take up to the output from six LWRs.

These values represent acceptable limits for the core safety parameters, in particular in relation to the Doppler and sodium void effects. The acceptable content is thus higher than in a UO_2-fuelled N4 PWR.

Multiple homogeneous recycling of Pu+MAs present in spent LWR-MOX fuel after advanced reprocessing was assessed [81] with respect to mass balance. The TRU components were assumed to be incorporated into fresh fast reactor fuel in a ratio 66% depleted-U and 33% TRU (Pu+MAs). This was submitted to multiple recycling in a sequence of 5 years irradiation and 12 years cooling. The results show a steady increase of the ^{238}Pu and ^{244}Cm concentration in the fuel discharged after having reached a burn-up of 150 GWd/tHM. In order to fully incinerate 1 tHM Pu+MAs from LWR-MOX, 15 FBuR sequences are necessary. The overall Pu+MAs destruction during the multiple recycling operations amounts to 88.4% which leads to an actinide reduction factor of about 12 in a period of 255 years.

Table II.7 **Mass balances for homogeneous recycling in fast reactors**

		FBR[*]
Burn-up (GWd/t)		120
Initial minor actinide content (%) (mass of heavy metal)		2.5
Np	Initial mass (kg)	1 010
	Specific consumption (kg/TWhe)	10
	Depletion (% of initial mass)	60
	Fissioned fraction (% of initial mass)	27
	Fissioned mass (kg)	273
Am	Initial mass (kg)	1 174
	Specific consumption (kg/TWhe)	9
	Depletion (% of initial mass)	45
	Fissioned fraction (% of initial mass)	18
	Fissioned mass (kg)	211

[*] EFR type reactor with three enrichment zones (15.32, 18.18 and 22.08%)

Separate recycling of minor actinides

The calculated effects of recycling 2.5% neptunium, americium or curium in a EFR-type reactor are compared with those of recycling 1% neptunium or americium in a UO_2-fuelled PWR.

Recycling ^{237}Np in a fast reactor has no impact at the beginning of cycle, except for an increase in dose rates at fuel fabrication (see Section 2.2.1 of PART II) due to the presence of ^{233}Pa in equilibrium with ^{237}Np (^{233}Pa is a strong γ emitter in the vicinity of 300 keV). After irradiation in a fast reactor and 5 years of cooling, a limited increase in the γ source and of the residual power can be observed. Homogeneous recycling of neptunium with a content of approximately 2.5% in a EFR type core therefore does not raise any major problem for the fuel cycle installations.

Recycling americium has more significant effects at the beginning of cycle with, in particular, a 4.5-fold increase in γ emission. After five years of cooling, all forms of radiation are tripled compared with standard EFR-MOX. The unfavourable consequences of homogeneously recycling of Am in fast reactors, although less than PWRs, imply a preference for heterogeneous recycling.

As regards curium, the consequences of homogeneous recycling are so unfavourable that it cannot even be considered.

Effect of lanthanides

Studies have been undertaken to investigate the impact of lanthanide impurity levels on the reactivity of the FR core loaded with MA and on the required decontamination factor between lanthanides and actinides in the fuel mixture.

The replacement of uranium by lanthanides increases the consumption of plutonium by reducing the formation of ^{239}Pu via capture by ^{238}U. The reaction ratio of heavy nuclei is approximately 38 times that of lanthanides on account of the latter's lower cross-sections.

The introduction of Am induces the usual reduction in both initial reactivity and loss of reactivity during irradiation. Part of the americium is replaced by curium (which is less absorbent) whereas the proportion of rare earths is low, and therefore has little influence, hence the increase in the initial reactivity.

The lanthanides have a low average absorption cross section (about 0.21 barn), which explains their limited impact on initial reactivity. However, as they are not fertile, their introduction results in an increase in the loss of reactivity during irradiation owing to competition with ^{239}Pu formation.

In order to maintain the same fuel management as in the reference case, it would be necessary to increase the plutonium enrichment if the decontamination factor between Am and lanthanides falls below 15. If DF>15, the effect of lanthanides is compensated by the influence of the minor actinides.

The increase in the void effect ($\Delta\rho_{void}$) is +2.4% per vol.% of minor actinides and +0.89% per vol.% of RE in the core. The reduction of the Doppler effect ($\Delta\rho_{dopp}$) is 5.3% per vol.% of minor actinides and 1.3% per vol.% of RE in the core. Lower DFs are permissible for recycling in fast reactors than in PWRs.

Heterogeneous recycling of minor actinides in fast reactor [77]

Mass balances for heterogeneous recycling

Table II.8 shows the calculated transmutation performance in heterogeneous recycling mode with target positioned in the first row of the radial blanket of an FR (see Figure II.14 (b)).

Specific consumption values range from 4 to 15 kg/TWhe, which is encouragingly higher than in standard PWRs. On the other hand, the fission rates are very limited. Any improvement in these rates requires either a lengthening of the irradiation time, which may be limited by the behaviour of the targets, or a multi-recycling mode.

Americium recycling

Table II.9 shows a comparison between the different alternative strategies for recycling Am in FRs.

The three cases correspond to the irradiation of targets placed in the first row of the radial blanket of the EFR-type core. The targets have identical dimensions to the fertile UO_2 assemblies. UO_2 is replaced by americium mixed with a matrix, as required. The cases differ in the initial mass of americium loaded. All the available space is occupied by americium in case 1. In cases 2 and 3, the targets are loaded with mixtures of americium and inert matrix, Al_2O_3 in this case. The common limiting criterion is the radiation damage to the cladding materials which is set at 200 dpa NRT for very radiation resistant steel types.

Depletion and fission percentages increase sharply with irradiation time when the initial content is reduced. In contrast, the absolute value of specific consumption decreases sharply, depending on the initial mass loaded.

Table II.8 **Mass balances for heterogeneous recycling in fast reactors**

		FBR
Irradiation time EFPD		4 500
Np	Content (%) (mass)	40
	Initial mass (kg)	3 574
	Specific consumption (kg/TWhe)	13
	Depletion (% of initial mass)	60
	Fissioned fraction (% of initial mass)	24
	Fissioned mass (kg)	858
Am	Content (%) (mass)	40
	Initial mass (kg)	3 523
	Specific consumption (kg/TWhe)	14
	Depletion (% of initial mass)	63
	Fissioned fraction (% of initial mass)	22
	Fissioned mass (kg)	775

The average power levels in the 100% americium targets are higher than in fuel assemblies, and their changes substantially with consequent irradiation are control problems.

The americium production rate of a FR ranges from 5 to 10 kg/TWhe depending on the cooling time. In order to confer on a FR a positive capacity of incinerating americium, exceeding simple self-consumption and preserving satisfactory fission, the optimum Am content of a target lies around 40%.

To avoid significantly changing core characteristics, up to 40% of Am is transmuted into Pu. It therefore appears wise to irradiate targets at the periphery of the core and to use the leakage neutron flux.

Mixed MA-loading in fast reactor [82]

The mixed MA loading method as shown in Figure II.14 (c) is a combination of the homogeneous and heterogeneous methods: Np is uniformly loaded in the core region and a small number of subassemblies containing Am, Cm and RE nuclides is loaded into the blanket region. Parameters were surveyed systematically to investigate the basic characteristics of MA transmutation in a 1 000 MWe-class FR core with mixed oxide fuel.

The mixed MA-loading method can transmute a large amount of MAs without serious drawbacks in terms of core performance. The transmuted mass of MAs is ~530 kg/cycle, which is almost 16 times the mass produced by an LWR of the same power output.

It was found that a combination of homogeneous and heterogeneous loading methods has the potential to achieve the maximum transmutation of MAs with no special design consideration.

Table II.9 **Irradiation of americium targets in FR**

CASE	1	2	3
Content (%)	100	40	20
Matrix	-	Al_2O_3	Al_2O_3
Numbers/positions	72 in blanket	72 in blanket	72 in blanket
Mass of americium (kg)	15 673	3 523	1 470
Residence time (EFPD) →200 dpa NRT	1 500	4 500	5 100
Am depletion consumption rate (%)	20	63	81
Fission rate (%)	6.6	22	38
Am specific consumption (kg/TWhe)	58	14	6.6
Mean target power (MW)	15	2.2	1.2
Mean burn-up (GWd/HMt)	93	80	62
Initial isotopic vector of Am: ^{241}Am = 63.6%, ^{242}Am = 0.2%, ^{243}Am = 36.2%			
This concerns an FBR 1400 MWe with two enrichment zones (17.52 and 23.87%). Regardless of the irradiation time considered, the table does not take account of any intermediate cooling time corresponding to shutdowns for core refuelling.			

2.3.3.2 Minor actinides transmutation in metal-fuelled fast reactor

Studies have been made on minor actinides homogeneously distributed in U-Pu-10% Zr metal fuel. In any of the technologies currently proposed for pyrochemical reprocessing, the recovered minor actinides are to some extent accompanied by rare earth fission products. According to the current pyrochemical reprocessing target value, a DF of 20 is assumed for the process. With this decontamination factor, the weights of rare earth fission products and minor actinides will be nearly equal.

In a metal fuel FBR, the minor actinides are homogeneously loaded in core.

There are two modes of fuel recycling [83-85], the self-recycle mode and that with additional actinides, with respective mass flow pattern as follows:

- in the self-recycle mode, the minor actinides recovered from the spent core fuel in each FBR are recycled without addition. Consumed plutonium is replaced from LWRs;

- in the minor actinide-enriched mode, the plutonium and minor actinides recovered from the spent core fuel of each FBR are recycled. Consumed plutonium and minor actinides are both made up from LWRs. In the particular example, pyrochemical reprocessing was assumed.

Core characteristics

The following conditions are assumed for neutronic assessment in the two modes defined above:

- the proportions of minor actinides, ^{237}Np, ^{241}Am, ^{243}Am and ^{244}Cm in the material recovered from the LWR spent fuel, are 54, 23, 17 and 6 wt%, respectively;
- a quantitative Pu recovery and 98% of minor actinides to be recovered in reprocessing.
- the core is composed of inner and outer regions with a combined thermal output of 1 600 MWt;
- the refuelling interval is 15 months with 3 batches, and the refuelling time is 60 days;
- the average discharge burn-up is ~90 GWd/tHM.

The performance parameters [83] of the FBR core at equilibrium recycle in the two modes described above are summarised in Table II.10 with the non-recycle core included for comparison. Here, the feed plutonium comes only from LWR and is used once-through. In the self-recycle core, the minor actinides and rare earths amount to 0.6 and 0.3 wt%, respectively, at equilibrium recycle.

In the minor actinide-enriched mode, the following significant characteristics appear:

- the burn-up reactivity swing becomes smaller with the increase in minor actinide enrichment;
- the minor actinide composition shifts to higher masses with recycling;
- the fast fission contribution causes an increase in void reactivity;
- the minor actinide-enriched core has a smaller Doppler constant due to the reduced uranium content;
- at full-power, the power coefficients which relate to fuel, cladding, structural components and coolant, and to the Doppler effect are reduced.

Taking the decreased β_{eff} of minor actinide-enriched core into account, it is quite difficult to use metal fuel with minor actinides of higher than a few percent.

Transmutation performance

In the self-recycle core, the minor actinides amount to 0.6 wt% at equilibrium. In an FR with 2 wt% minor actinide-enriched fuel, the transmutation rate is 31% at each refuelling the reactor can consume the minor actinides and plutonium recovered from 2.5 LWRs per year.

Effect of lanthanide nuclides

In nuclear reactors, lanthanide nuclides (i.e. rare earth isotopes) are created by fission of actinides. But transmutation rates in fast reactors are not changed substantially by their presence [86,87]. Rare earth fission products have capture cross-sections that cause them to act as a poison. With low distribution factors there will be large burn-up reactivity loss. The weight ratio of the total rare earth fission products to minor actinides in pyrochemical process is ~1. The reduction of reactivity due to rare earth fission product must be compensated by increased plutonium concentration [86]. Therefore, the increase of rare earth fission products content causes the decrease of Doppler coefficient and β_{eff} [87]. But the transmutation rate is not changed markedly by the content of rare earth fission products.

Table II.10 **Core performance at the equilibrium cycle of metal -fuelled fast reactor**

	Non-recycle	**Self-recycle**	**2%MAs enriched**	**5% MAs enriched**
MAs/RE concentration[1)] (wt%)	0.0/0.0	0.6/0.3	2.0/0.9	5.0/2.0
Pu concentration (wt%, IC/OC)[2)]	13.5/21.2	14.0/22.8	14.2/23.3	13.8/23.0
Burn-up reactivity (%Δk/k)	2.76	1.24	1.09	0.33
Internal conversion ratio	0.85	0.89	0.89	0.9
$ß_{eff}$ ($\times 10^{-3}$)	3.65	3.47	3.39	3.24
Void reactivity (%Δk/k)	1.78	2.03	2.19	2.5
Doppler constant ($\times 10^{-3}$Tdk/dT)	-4.62	-4.18	-3.70	-2.94
Power coefficient (¢/%-power)	-0.191	-0.163	-0.118	-0.036

1) concentration: wt% of (heavy metals +RE)
2) IC: Inner core, OC: Outer core

2.3.4 Transmutation of minor actinides and fission products in dedicated systems

Dedicated transmutation systems are being studied at JAERI [88-90] and CEA [91] based on a strategy named the double strata fuel cycle concept (or multi component concept). The double strata concept is to consider a P&T fuel cycle (second stratum) separated completely from the conventional fuel cycle for commercial power reactors (first stratum) as illustrated schematically in Figure II.15.

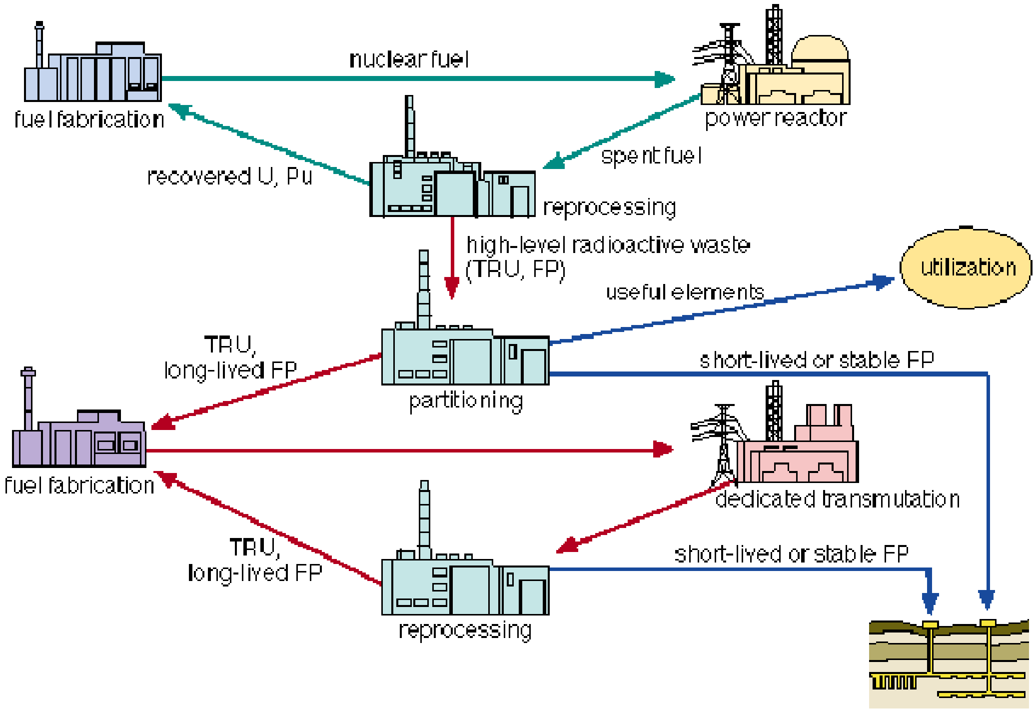
The first stratum is devoted to the electricity production and consists of standard power reactors (LWR-UO_2, LWR-MOX and FR), fuel fabrication plants and reprocessing plants. The U fuel irradiated in LWRs is reprocessed and the recovered Pu is recycled in LWR-MOX and in fast reactors. HLW from the reprocessing goes to the second stratum.

In the second stratum, MAs (Np, Am, Cm) and long-lived fission products are partitioned from HLW to be fabricated into fuels and targets. They are irradiated in dedicated systems for transmutation, and then reprocessed for multiple recycling.

There are several advantages in using dedicated transmutation systems compared with recycling MAs into commercial power reactors. Because of much lower mass flow in the P&T cycle, it could be on a correspondingly smaller scale than the main cycle. Its separation from the main power-producing cycle, and the small number of units required, would allow the extensive innovation in fuel fabrication, core design and reprocessing technology needed to optimise this part of the overall system [91-93]. It would also avoid burdening the main cycle with the problems associated with higher radioactivity and decay heat, and reduced safety margins in reactor physics parameters [94].

The P&T cycle could be made very compact by co-locating the entire facilities. This would minimise the transportation of nuclides that are troublesome with respect to waste management, and confine them effectively in the P&T fuel cycle.

Figure II.15 **Concept of double stratum fuel cycle**



Both critical reactors (burner reactors) and sub-critical reactors (hybrid systems or accelerator-driven systems) are potential candidates as dedicated transmutation systems used in the second stratum. MA transmutation in critical reactors can be either homogeneous or heterogeneous. The extreme case of heterogeneous recycling is a core loaded only with MAs (or with MAs and Pu). This could maximise the transmutation rate in dedicated systems. Systems loaded with MA based fuel, however, pose crucial problems related to reactivity coefficients (increase in the void reactivity coefficient and decrease in the Doppler effect), and to the small delayed neutron fraction.

To mitigate these problems for critical systems, a considerable amount of U should be added to the MA based fuel [94], preferably highly-enriched U to avoid undesirable accumulation of MAs, but this results in reduced transmutation rate. For hybrid systems, its sub-criticality mitigates the problems without adding U and thus allows the maximum transmutation rate.

2.3.4.1 *Dedicated minor actinide burner reactor* [95,96]

Two types of actinide burner reactors (ABRs) are designed at JAERI. The fuel material of these ABRs is a MA-U nitride mixture. One type is lead-cooled with pin fuel (L-ABR) and the other is He-cooled with pellets-type fuel (P-ABR). Nitride forms were selected as fuel material for these ABRs because of good thermal properties, applicability to very high burn-up, expected stability and mutual solubility of the various heavy-element component. The reactor core design parameters of these ABRs are given in Table II.11. Their neutron energy spectrum is very hard with the core-averaged neutron energy around 720 keV. These hard neutron spectra are very effective for direct fission of those MAs which have fission thresholds above 600 keV. The MA fission in the ABRs is 190 to 200 kg/GWt-year.

Table II.11 **Reactor design parameters of actinide burner reactors**

	L-ABR[1]	P-ABR[2]
Fuel concept	pin-bundle	coated-particle
material	(64NpAmCm-36U[3])$_{1.0}$N[4]$_{1.0}$	(65NpAmCm-35U[3])$_{1.0}$N[4]$_{1.0}$
MAs initial loading, kg	918	2870
MAs/U	588/330	1 865/1 005
Reactor power, MWth	180	1 200
Coolant material	Lead	Helium
Neutron flux, 10^{15} n/cm^2·sec	3.1	6.6
Core averaged mean neutron energy, keV	720	720
Reactivity (% $\Delta k/k$)		
Coolant-void reactivity/core	-1.3	–
Doppler reactivity/core (Δt=300°C)	-0.01	-0.01
Kinetic parameters		
β_{eff}	2.6×10^{-3}	2.6×10^{-3}
l_P (sec)	1.3×10^{-7}	1.5×10^{-7}
Cycle length, full-power days	550	300
MA burn-up, %/cycle	11	13

1) L-ABR: MA-nitride fuel with lead coolant burner reactor.
2) P-ABR: MA-particle fuel burner reactor.
3) 90% enriched uranium.
4) ^{15}N enriched.

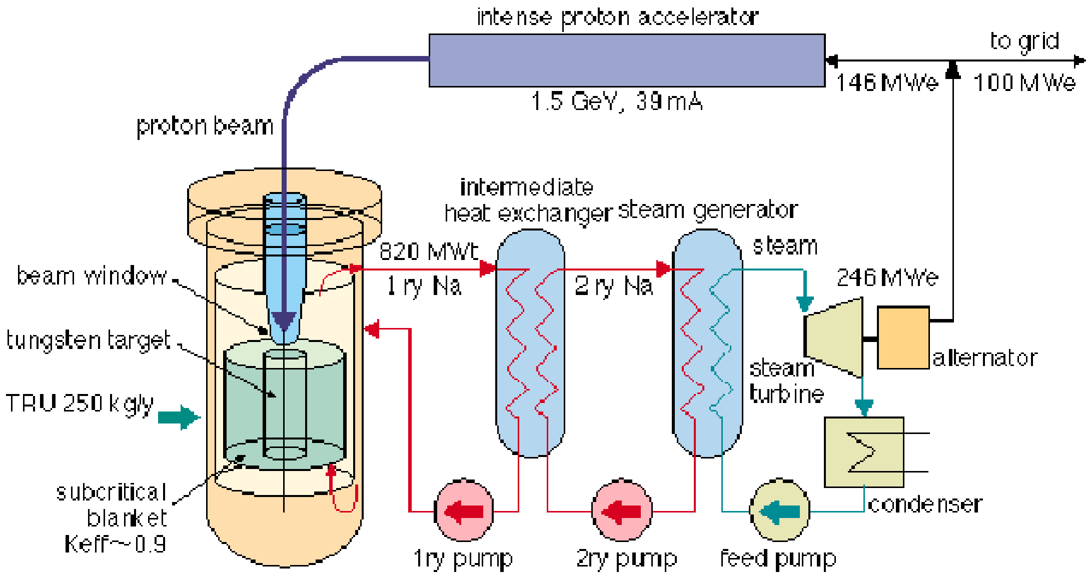
2.3.4.2 Accelerator-driven transmutation systems

Accelerator-driven systems (ADS, frequently called hybrid systems) combine high-intensity proton accelerators with spallation targets and a subcritical core with or without blanket (see Figure II.16). The proton accelerator will be either a linear accelerator (linac) or a circular accelerator (cyclotron). The high-intensity continuous-wave (CW) proton beam with an energy around 1 GeV and a current of several tens mA are injected into a target of heavy metal. This results in spallation reaction that emits neutrons, which enter the subcritical core to induce further nuclear reactions. The subcritical core can, in principal, be operated with either a thermal or a fast neutron spectrum.

ADSs have unique features to burn MAs and FPs, preferably in the double strata option. They operate in a subcritical mode and can more easily address safety issues associated with criticality than in critical systems. They also offer substantial flexibility in overall operation. ADSs can provide more excess neutrons compared to critical reactors. The excess neutrons may be utilised for transmutation, conversion, and breeding purposes. These features may be exploitable to prepare a safe and efficient mean of transmuting nuclear waste. Both homogenous and heterogeneous fuel recycling is possible.

Various concepts of ADS have been proposed with different goals and approaches. Relevant R&D programmes are being pursued at CEA, JAERI, LANL, CERN, etc. In recent years, all the system concepts proposed by these groups have converged on a fast neutron spectrum because of its large neutronic advantage over the thermal one, and the reduced production of higher actinides.

Figure II.16 **Concept of accelerator -based transmutation plant**



Technical issues

High-intensity accelerator

The high-intensity proton accelerator technology required for industrial applications has been under continuous development for the past decades. Beam powers in the range 10~100 MW are assumed in the current proposals of accelerator-driven systems. The maximum beam power that could be achieved within a decade would be up to around 100 MW for a linac and around 10 MW for a cyclotron.

The LANSCE accelerator at LANL is the most powerful operational proton linac (around 1 MW) in the world. It is operated in pulsed mode (10% duty), and well below the limits of space charge and radio frequency (RF) bucket filling. It is estimated that a 40~160 MW CW (100% duty) beam is possible in principle with simple extension of proven technologies.

Cyclotrons have the advantages of much smaller physical size and lower cost than linacs with the same beam power. The PSI proton accelerator is the most powerful operational cyclotron (around 1 MW) in the world. The PSI group made a preliminary design of a cyclotron with a relatively modest beam power of 10 MW [97].

In the high-intensity accelerator, beam loss should be minimised to avoid resultant damage and activation of accelerator hardware. For industrial applications, primary issues are the stability, efficiency, reliability, operability and maintainability of the accelerator. Recent reviews of the operation reliability of accelerators have shown that beam trips and proton source failures are very frequent. Substantial improvement is required and R&D priority should therefore be given to that essential part of ADS. Reducing the cost and size of the accelerator should also be included in the design priorities.

Spallation target

Reliable nuclear data and codes for the intermediate energy region are required for the design of an ADS. At present, most of the data and codes are available for an approximate evaluation of conceptual designs and for feasibility studies. Detailed designs will need much higher accuracy. If a ±2% accuracy on the system's energy balance is required, the spallation neutron yield should be calculated with an accuracy of ±2%. Uncertainties still seem large in predicting the spallation product yields and the high energy component of the neutron spectrum to evaluate activation and damage in materials.

Injection of the intense proton beam into the target causes high fluxes of protons and fast neutrons in the beam window, target, and wall material surrounding the target. These, particularly for the beam window, suffer irradiation damage and are degraded in mechanical properties and dimensional stability. The exposure of the materials to high fluxes and energies would be more severe than in normal reactors. Research on the interaction between high-energy proton and neutron beam and window as well as structural materials is required.

An intense proton beam deposits heat in the target. Heat removal requirements for the target are essentially identical to those for the fuel.

Subcritical core

A subcritical core can be very similar in principle to a critical core except that the effective neutron multiplication factor is less than unity. A subcritical core cooled by liquid metal can fully utilise existing LMFR technologies.

Subcritical operation provides great freedom in design and operation. Criticality in a conventional reactor imposes tight constraints on the fuel specifications and cycle length. Accelerator-driven systems can accept fuels that would be impossible or difficult to use in critical reactors, and can extend their cycle length if necessary.

Trips and fluctuations of the incident proton beam are inevitable, causing thermal shocks in the core components. The design must take this into consideration; power distribution, effective neutron multiplication factor, the neutron flux shape transient response and the size of the system.

Safety features [98]

The subcriticality of an ADS has clear safety advantages for severe reactivity accidents. It can cope with fast ramp rate accidents which could occur too rapidly for scram systems in critical reactors. A margin to accommodate fast reactivity insertions is important to avoid super-criticality accidents.

The consequences of cooling failure for ADSs are similar to critical reactors. A reliable beam shut-off system is, therefore, required for an ADS, just as a reliable scram system is required for a critical reactor. A reliable emergency decay heat removal system is required for both.

Performance assessment of accelerator-driven systems

Over the past few years a number of different ADS concepts have been developed. For the purpose of illustration, the performance of an ADS described in References 102 and 103 is discussed here. The objective is to incinerate Np, Am and Cm and transmute ^{99}Tc and ^{129}I in spent LWR fuel.

In Reference [99], the sodium cooled, fast spectrum ADS employs a very high power accelerator of up to about 200 mA of proton beam with an energy of 1.6 GeV. The spallation target is MAO_2 or PuO_2 fuel itself. The proton beam is spread over the entire surface of the target to avoid the hot-spot problem. In this scenario, three different cores are required; the first one is for the incineration of MA from 14 units of LWRs; the second core is loaded with ^{99}Tc and ^{129}I and plutonium produced in 19 LWRs. ^{129}I eventually left over from this core is transmuted in the third core, fuelled by a fraction of Pu coming out of the second core. At certain stage, the 2nd and 3rd cores require feed of grid electricity, but on the average, no core is a net consumer of grid electricity. In these calculations, a 12.5% conversion ratio of core thermal power to a given proton beam power is assumed. It was shown that the considerable power swings between BOC (k_{eff}=0.95) and EOC (k_{eff}=0.80) stages could be smoothed out by adjusting the beam current of 58 mA at BOC to 243 mA at EOC in the 2nd core, if deemed desirable. A technical difficulty of this concept is the direct use of MAO_2 or PuO_2 as a spallation target, the need of beam intensity adjustment by threefold during one cycle of ADS operation, and the use of spread proton beam.

In Reference [100], two types of fast spectrum ADS with nitride fuels were designed. One type is Na-cooled core with a solid tungsten (W) target at the core centre and the other is Pb-Bi target and coolant system. The spallation targets are bombarded by high energy and intense proton beams of 1.5 GeV and 45 mA. The parametric studies were conducted to obtain the optimal neutronic characteristics of the subcritical core to maximise the MA transmutation rate and to minimise the burn-up reactivity swing during irradiation by adjusting the MA and Pu fraction of nitride fuels with ZrN inert matrix as thermal diluent. These core design parameters are given in Table II.12. The coolant void reactivity is negative in the Pb-Bi cooled core, though it is positive in the Na-cooled one. The MA transmutations in both cores are 250 kg/year and this amount corresponds to the annual production of MA in about 10 LWRs. FP (^{99}Tc and ^{129}I) transmutation is calculated for loading the FP target assemblies with ZrH moderator pins in the core-reflector region. The core performance for FP loading is also given in the table.

Table II.12 **Characteristics of the Na and Pb-Bi cooled 820 MW -ADS cores with (MA, Pu) nitride fuels (Proton beam 1.5 GeV – 45 mA, 30 spallation neutrons/proton)**

Type	**Na cooled MA transmutor**	**Pb-Bi cooled MA transmutor**	**Pb-Bi cooled (MA, FP) transmutor**
Target	Solid Tungsten	Liquid Lead-Bismuth Alloy	
Initial core inventory (kg) (MA/Pu/FP)	1950/1300/0	2500/1660/0	2500/1660/1000
MA compositions (%) (^{237}Np/^{241}Am/^{243}Am/^{244}Cm)	56.2/26.4/12.0/5.11		
k_{eff} (Initial/Max./Min.)	0.93 / 0.94 / 0.90	0.95 / 0.95 / 0.94	0.93 / 0.93 / 0.92
Coolant void reactivity (%Δk/k)	+4.5	-4.8	-7.1
Transmutation rate (kg/year) (MA/FP)	250/–		250/40

Calculation: Code system: ATRAS [101], Nuclear Database: JENDL-3.2 Library [102].

Review of the existing projects

Active projects for the accelerator driven transmutation systems exist in France, Japan, USA, and CERN. Furthermore, there are a number of research activities in many other countries as well as within the international programmes of OECD/NEA, IAEA, and EC.

The Belgian MYRRHA project

The MYRRHA project has been started at the end of 1996 as a conceptual study aiming at the development and realisation of a new radiation source based on accelerator driven neutron generation for multiple purposes.

The accelerator part would consist of a 25 mA proton source, an accelerator of 25-30 MeV, and a proton-cyclotron with an exit proton energy of 250 MeV and a proton current of 2 mA to be upgraded to 10 mA. The multipurpose research facility would be used in materials research, radioisotope production (^{99}Mo), proton therapy and last but not least for the study of spallation induced transmutation of long-lived radionuclides. The total energy would not exceed 30 MWth. The fast neutron flux could reach 1.5×10^{15} n/(cm^2·s) in a core volume of 35 L.

The spallation source would be a windowless liquid lead-(bismuth) target surrounded by a subcritical assembly. The scale of the MYRRHA system is limited to remain a prototype research facility. The basic engineering study is currently going on.

Czech activities [103]

The Czech Republic started a national R&D programme on accelerator-driven transmutation technologies. A project LA-0 is proposed for testing the subcritical modular assemblies with fluoride salts on the experimental reactor LR-0.

CEA project, France

Within the framework of the French SPIN Programme [104], the CEA ISAAC programme has been set-up to investigate the physics of subcritical ADSs. The programme includes the MUSE experiments at MASURCA [105] and the spallation experiments at SATURNE [106].

Recently, a research group GEDEON made up of the CNRS, CEA, EDF and Framatome has also been set-up to intensify and co-ordinate research in these areas. An experimental fast system called HADRON has been proposed for experimental validation and demonstration of an ADS. The concept is based on a subcritical core with a thermal power of 50-100 MW.

German activities

At the Technical University Munich the design of a separated-orbit cyclotron with superconducting channel magnets and superconducting RF cavities for 1 GeV proton beams of up to about 10 MW beam power is under development (TRITRON). The distinguishing feature of this type of cyclotron is the strong transverse and longitudinal focusing [107]. Recently it was demonstrated that the principle works as anticipated with operation well above the design values [108].

In Germany, some small activities related to the application of ADSs for the back-end of the nuclear fuel cycle are in progress since several years [99,109,110,111]. 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 ADS investigations have been performed for thermal systems with dispersed fuel in lead coolant at FZJ Juelich and for Phénix like fast systems at FZK Karlsruhe.

Italian activity

ENEA (Ente per la Nuove Tecnologie l'Energia e l'Ambiente) and INFN (Instituto Nazionale di Fisica Nucleare) set up a basic R&D programme TASCO aiming at the study of physics and technologies needed to design an ADS for nuclear waste transmutation. The programme consists of research subprogrammes on proton accelerator, neutronics, thermal-hydraulic 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 [112].

JAERI project, Japan

JAERI is carrying out studies on accelerator-driven transmutation systems and development of a high-intensity proton accelerator [113,114] under the Japanese OMEGA Programme. Two types, using respectively solid and a molten-salt fuels have been proposed as dedicated transmuters.

Engineering tests for accelerator-driven transmutation are planned under the JAERI proposed Neutron Science Project based on a 1.5 GeV-5.3 mA superconducting linac. Demonstration tests on individual components will be made for a 30-60 MWt integral target/core system and technical feasibility of the window/target assembly will be also tested with a 7 MW beam power.

KAERI HYPER programme, Korea

KAERI has initiated a study on the transmutation since 1992 [115] and is now setting up a long-term research programme called HYPER on ADSs. KAERI is also trying to launch a programme for the development of a 1 GeV-20 mA multi-purpose linear proton accelerator called KOMAC.

Russian activities

Several research institutes in Russia are involved in a partitioning and transmutation programme directed by MINATOM [116]. Most of activities relevant to accelerator-driven transmutation are carried out within the framework of ISTC projects.

Spanish activities

CIEMAT (Centro de Investigaciones Energeticas MedioAmbientales y Tecnologicas) started in 1997 a research programme to investigate the physics of accelerator driven transmutation systems. Also, a private enterprise, LAESA (Laboratorio del Amplificador de Energia), has been created in 1997 to build a laboratory for research, development and demonstration of the Energy Amplifier concept.

Swedish activities [117]

Research on partitioning and transmutation 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 groups has strong international cooperations and participate in projects supported by EU. The Svedberg Laboratory at Uppsala has recently started a project for cross-section measurements of interest for ADSs.

PSI activities, Switzerland

The PSI activities in the field of transmutation aim at investigating the potential ADSs with regard to reactor performance and radioactive waste management aspects and at supporting developments for the SINQ spallation neutron source. More specifically, the activities comprise high-current cyclotron development and conceptual design work [97], material technology development work for spallation targets, the experimental validation of models in nucleon-meson transport codes by means of the ATHENA irradiation experiments using actinide targets [118], reactor physics and safety analyses of ADSs, and comparison studies for different reactor types and fuel cycles with emphasis on the impact of alternative transmutation strategies on the long-term risk of the radioactive waste [119].

LANL project, USA

LANL is developing a linear accelerator with a beam power of hundreds of MW under the APT (Accelerator Production of Tritium) Programme. The project has had many independent reviews. Their conclusions were positive regarding the accelerator and target technologies, but pointed out the need for an appropriate R&D programme [120].

The LANL ATW programme [121,122] aims at reducing the amount and long time hazard of the spent fuel from US commercial nuclear reactors. The ATW-system would incinerate the TRU waste and transmute selected FPs, such as ^{99}Tc and perhaps ^{129}I. The ATW-system is a fast spectrum liquid lead-bismuth cooled device coupled to an APT-class linear proton accelerator. (The accelerator, providing a current of 100 mA and a proton energy of 1 GeV is developed for tritium production, also at LANL). The fuel fabrication and the fuel cycle relies on pyrometallurgical processing.

LANL is proposing an experimental programme called LIFT [123]. The experiments will be carried out using 1 MW proton beam at LANSCE on Pb-Bi loop and on a ~5 MWt integral target/blanket.

CERN EA project

CERN is working on the conceptual design of a so-called Energy Amplifier (EA) and this development may be of use to P&T. The design as originally suggested used a moderated system [124]. The present concept is an oxide fuel, Pb-cooled fast subcritical system driven by a 1 GeV-12.5 mA cyclotron [125]. This system was also proposed as a Pu burner [126]. The first demonstration plant is planned to run with a superconducting linac [127]. The CERN group has performed a high-energy physics experiment associated with the concept [128] on a natural U assembly irradiated by 1-3 GeV proton beam.

OECD/NEA programmes

The OECD/NEA Nuclear Science Committee (NSC) started activities related to ADS and published a report describing different transmutation systems [129]. This report was followed by an international benchmark exercise to compare a specific fuel scenario, where MAs were recycled either through a PWR, a fast reactor or an accelerator-driven system [130]. In October 1998, the NSC organized a workshop in Japan on the "Utilization and Reliability of High Power Proton Accelerators" [131].

The NSC is organizing a series of meetings on "Shielding Aspects of Accelerators and Irradiation Facilities (SATIF)". The fourth meeting was held at Knoxville, TN, USA, in September 1998 [132].

In addition, the NSC and the NEA Data Bank have activities of intercomparing the performance of computer codes [133-140] and have activities of evaluating intermediate energy nuclear data files [141,142], used for the modeling of ADS systems.

IAEA programme

IAEA published a status report [143] on accelerator-driven system to overview development activities and system concepts. A benchmark of sub-critical core (Stage 1) has been completed in the frame of the IAEA CRP on use of Th-based cycle in ADSs to incinerate Pu and to reduce long-term waste toxicities and results were reported to the Technical Committee Meeting on feasibility and Motivation for Hybrid Concepts for Nuclear Energy Generation and Transmutation, Madrid, Spain, 17-19 September 1997 [144].

European Commission projects

The impact of accelerator-based technologies on nuclear fission safety (IABAT) is being assessed by several research organisations of the European Union. The objectives of the project are to perform systems studies on ADS, to assess accelerator technology, to study the radiotoxicity of the fuel cycle and its non-proliferation aspects and to provide basic nuclear and material data useful for ADS [145].

2.3.5 Nuclear data of minor actinides and long-lived fission products

For the reactor transmutation studies, nuclear data libraries on the elements to be recycled were compiled from the JEF-1, then, more recently, the JEF-2 databanks. The JENDL Actinide File is being compiled in addition to the JENDL-3.2. It contains data on neutron-induced reaction for about 90 nuclides from ^{208}Tl to ^{255}Fm.

In the reactor transmutation studies on long-lived radioactive waste, nuclear data for MA nuclides and fission products are of primary importance. However, nuclear data for many MA nuclides are still not known with the desired accuracy. Accurate experimental data of neutron cross-sections for MAs are indispensable to establish MA transmutation technology by reactors. Accurate neutron cross-section data of RE nuclides become also necessary for designing the MA burning core. The data, however, are quite inadequate both in quality and in quantity.

As regards the fast spectra, results from experiments conducted in the Phénix reactor are available. They concern, on the one hand, irradiations of separated samples (PROFIL 1 and PROFIL 2 experiments [146]), and, on the other hand, an integrated experiment (SUPERFACT) during which fuel pins containing different neptunium and americium contents were irradiated [59].

As for the thermal and epithermal spectra, results from tests conducted on separated samples of actinides are likewise available (SHERWOOD and ICARE experiments conducted at the MELUSINE reactor). Furthermore, analyses of a number of experimental assemblies irradiated in power reactors are also available.

Critical experiments conducted in the MASURCA (for fast spectra), EOLE and MINERVE (for thermal spectra) reactors provided data on some minor actinide fission rates.

The fission products capture cross-sections were validated through oscillation experiments conducted in the MINERVE reactor [147]. Tc rods were irradiated in the HFR reactor in Petten for one year.

In the framework of the 1995-1998 Programme on Nuclear Fission Safety funded by the European Commission, six companies and research centres are comparing their MOX irradiation data banks with recalculations using modern methods and data, mainly from the JEF-2.2 file, so as to assess the accuracy of systems studies on actinide transmutation involving the use of MOX fuels. The experimental base comes from France (as detailed above), Belgium and Germany [148].

In fast reactor spectra, an irradiation of pure isotopic samples, similar to the PROFIL 1 and 2 experiments quoted above, was performed up to a maximum burn-up of 185 GWd/t in the KNK-II reactor at Karlsruhe. The results, after verification by JRC-ITU Karlsruhe, will be used to extend the data base.

For thermal spectra, while the CEA recalculates mass balances from MOX fuel irradiated in a 900-MWe PWR up to 46 GWd/tHM, the SCK•CEN Mol/Belgonucléaire group has compiled the results of many MOX irradiations in various PWRs and BWRs; the maximum burn-up reached was 82 GWd/tHM in the BR3 reactor. Cross-section libraries mainly based on the JEF-2.2 file are used for recalculations. Their checks and recalculations are backed by sensitivity analyses done in parallel at ECN Petten.

A complementary activity of ENEA Bologna, in relation with CEA, is to re-evaluate basic cross-section files for Pu and Am isotopes and to add photon production cross-sections.

The minor actinide nuclear data are measured for fission-neutron yields, delayed-neutron spectra, and fission yields at JAERI in collaboration with the Oak Ridge National Laboratory (ORNL) and Texas A&M University. Actinide nuclear data in the JENDL File are evaluated using the integral experiments at the fast critical facility, FCA [149].

Fission cross-section ratios of minor actinide nuclides (^{237}Np, ^{241}Am and ^{243}Am) relative to ^{235}U in the fast neutron energy region have been measured at YAYOI fast neutron source reactor [150].

Making use of back-to-back (BTB) fission chambers and a lead slowing-down spectrometer coupled to a 46 MeV electron linear accelerator at Kyoto university, the fission cross-sections of ^{237}Np, ^{241}Am, ^{242m}Am and ^{243}Am have been measured relative to that for ^{235}U(n,f) reaction in the energy range from 0.1 eV to 10 keV [151].

As a part of MA nuclear data evaluation, the analysis of a ^{237}Np sample irradiated in JOYO has been performed. Additional irradiation test of ^{237}Np, ^{241}Am, ^{243}Am and ^{244}Cm samples in JOYO was started in August, 1994.

Measurements of keV-neutron capture cross-sections of RE nuclides (^{147}Sm, ^{148}Sm, ^{150}Sm, ^{140}Ce, ^{141}Pr, ^{153}Eu, ^{143}Nd, ^{145}Nd) have been performed to evaluate the accuracy of the nuclear data libraries using the 3-MeV Pelletron accelerator of the Research Laboratory for Nuclear Reactors at the Tokyo Institute of Technology [152].

2.3.6 Interaction between plutonium incineration and minor actinide production

2.3.6.1 Present day PWRs

If present day PWRs could be licensed to accept 100% MOX containing depleted U, the balance of Pu consumption would be around 60 to 70 kg/TWhe during successive recyclings, but with concomitant production of minor actinides arising from 8 to 20 kg/TWhe. However, for high burn-ups in the range of 50 GWd/tHM, the number of plutonium recyclings in MOX assemblies with depleted uranium support is liable to be limited to two or three due to the degradation of certain safety-related coefficients, and particularly of the coolant void reactivity coefficient, which tends to become positive locally when the plutonium content substantially exceeds 12%.

2.3.6.2 Other alternatives

Other alternatives have been investigated, based on the use of PWRs, but with design changes intended to limit the Pu content of the MOX assemblies, and hence to restore the safety and reliability of multi-recycling.

High moderation reactors (HMRs)

High moderation reactors (HMRs) have a moderation ratio (moderator volume/fuel volume) of 3.

Compared with a standard PWR (in which the ratio is 2), the enhanced moderation helps to lower the Pu content of the MOX loaded with each recycle (one-third in the first recycle) and to improve Pu consumption per TWhe.

Thus the consumption of Pu would increase from about 70 kg/TWhe in the first recycle to nearly 110 kg/TWhe at equilibrium, but the production of minor actinides would rise from 10 kg/TWhe to nearly 34 kg/TWhe.

The Pu content would be stabilised around 20%, which is relatively high, requiring further physical feasibility studies for this type of reactor.

Plutonium recycling in MOX with enriched uranium support (MOX-EU)

In a standard PWR, self-recycle of all the Pu produced in the MOX-EU would help to limit and stabilise the Pu content at values in the region of 2%, which would require ^{235}U enrichment of about 3.5%, close to that of a standard enriched U fuel with the same performance. However, it must be observed that:

- The cost of the initial ^{235}U over-enrichment could be partially recovered through the higher residual enrichment in the reprocessed uranium (REU).

- The isotopic quality of the Pu deteriorates through accumulation of ^{238}Pu and more particularly ^{242}Pu increasing the conversion to MAs by a factor of 5.

Another alternative is MOX-EU with a higher constant Pu content (e.g. 8.7%, as in the first recycle) to reduce the overall number of MOX assemblies. In this case, Pu consumption would range from 75 to 60 kg/TWhe, and minor actinide production from 19 to 23 kg/TWhe for a 100% MOX refuelling in a standard PWR.

Yet, another alternative is an HMR using MOX-EU (see Table II.13).

Isotopic separation of ^{242}Pu before Pu recycling on depleted U support

The proportion of non-fissionable ^{242}Pu rises during successive recycles. Moreover, by neutron capture, it gives rise to the minor actinides ^{243}Am and ^{244}Cm. The hypothetical case of isotopic separation before recycling would hence prevent ^{242}Pu from accumulating and from increasing the production of minor actinides. Accordingly, in multi-recycling of Pu in standard MOX, the Pu content could be kept constant as a first approximation at a value close to that of the first recycle, reducing the production of minor actinides by a factor of 2 or 3.

2.3.6.3 FRs without blanket (FBuR)

In FBuRs without blanket, the consumption of plutonium depends on its initial content in the fuel: the higher the content, the more incinerating is the core.

At a content of 20% (EFR type without blanket), Pu consumption is about 20 kg/TWhe, at 30% (Phénix type without blanket) about 50 kg/TWhe, and at 100%, the theoretical limit, it could reach 110 kg/TWhe.

In the CAPRA type FBuR investigated by the CEA, the target of 75 kg/TWhe is reached with a Pu content of 42%, with a larger concomitant drop in reactivity, making it more difficult to control the reactivity and shortening the cycle.

The production of minor actinides remains approximately constant throughout the successive plutonium recycles. It increases with the initial Pu content of the core from 3 kg/TWhe (at 20% Pu) to 10 kg/TWhe (at 40% Pu).

Table II.13 **Pu and minor actinide balance of different types of reactors**

	PWR UO_2	PWR MOX (depleted U)	PWR MOX (2% enriched U)		HMR MOX (depleted U)	HMR MOX (enriched U)	FBR (EFR type)	FBR (Phenix type)	FBuR CAPRA
Pu content of MOX (%)		8.6 to > 13	2	8.7	5.9 to 20	5.9	20	30	42
Core exit Pu balance (kg/TWhe)	+30 to 35	-60 to -70	0	-75 to 60	-70 to -110	-67 to -45	-20	-50	-75
Exit MA balance (kg/TWhe)	+2.5	+8 to +17	+5	+20	+10 to +34	+12 to +18	+3	+6	+10

2.4 Transmutation of long-lived fission products

2.4.1 Transmutation of fission products in fission reactors

The incineration capacity for long-lived fission products in conventional reactors is very limited, and these neutron-absorbing substances tend to poison the core. The reactor neutron balance makes it conceivable to recycle some but certainly not all of them.

Table II.14 shows the production of long-lived fission products and of the corresponding chemical elements in a PWR with UO_2 fuel. One observes, for example, that ^{135}Cs, a long-lived isotope, accounts for only 10% of the mass of the chemical element caesium. Irradiation of the caesium would thus produce ^{135}Cs from the isotopes 133 and 134, which would severely compromise the desired objective.

Table II.14 **Production of long -lived fission products (PWR UO_2 50 GWd/t)**

Isotope	**Half-life (years)**	**Isotope quantity (kg/TWhe)**	**Element quantity (kg/TWhe)**
^{14}C(*)	5.73×10^3	0.0013	0.0013
^{79}Se	6.5×10^4	0.018	0.209
^{93}Zr	1.53×10^6	2.8	13.7
^{99}Tc	2.13×10^5	3.2	3.2
^{107}Pd	6.5×10^6	0.78	4.8
^{126}Sn	1.0×10^5	0.079	0.2
^{129}I	1.57×10^7	0.66	0.8
^{135}Cs	2.3×10^6	1.40	14.0
Total		9.0	37

(*) Activation product

2.4.1.1 Transmutation of ^{99}Tc and ^{129}I

In practice, ^{99}Tc and ^{129}I are the main FPs to be considered as candidates for transmutation in present reactors: only ^{99}Tc has been experimentally studied.

Transmutation of ^{99}Tc or ^{129}I to stable ^{100}Ru and ^{130}Xe, respectively, may be accomplished by neutron capture. Because no neutrons are produced in the transmutation process, introducing these nuclides into a fission reactor will lower the reactivity or shorten the cycle, unless one increases the fuel enrichment to compensate for the reactivity loss.

The neutron absorption cross-section of ^{99}Tc exhibits a strong resonance in the epithermal range, while ^{129}I is a 1/v neutron absorber (see glossary). When a nuclide with a spectrum-averaged one-group absorption cross-section σ is irradiated in a neutron flux φ, one may define the transmutation half-life:

$$T_{1/2} = \frac{\ln(2)}{s \cdot j}$$

This expression for the transmutation half-life will be used below (see Table II.15), as it characterizes the transmutation rate of the long-lived fission product in the targets. Because the fission product will also be produced in the reactor, one has to consider the net transmutation rate, subtracting the mass of the fission product produced in the fuel from the mass destroyed in the target.

The problems involved in the transmutation of ^{129}I are severe. Besides doubts about the stability of the chemical form (see Section 2.2.2.3), the formation of gaseous xenon requires the target to be vented, raising considerable safety issues.

2.4.1.2 Transmutation in fast reactors [153-155]

Transmutation of ^{99}Tc in fast reactors may be accomplished in several ways: in a special moderated sub-assembly loaded at the periphery of the core or in the inner core, or in a non-moderated subassembly loaded in the core. Moderation could be realised with a material like CaH_2. Attention has to be paid to the required fuel enrichment and to power peaking in the neighbouring fuel assemblies caused by moderation. Although the capture cross section of ^{99}Tc in a fast neutron spectrum is relatively low, transmutation in a fast reactor without moderation could be advantageous because of the very high neutron flux and the limited power peaking. The consequences of introducing FPs should be evaluated.

Typical values of transmutation rates and half-lives are given in Table II.15. A fast reactor with a power of 1 200 MWe could transmute the ^{99}Tc production of five 1-GWe-PWRs with moderation, or the production of four PWRs without it. But this would need a huge ^{99}Tc loading, leading to design problems and economic penalties.

To improve transmutation performance, a new concept of duplex pellet – a moderator annulus surrounding a central ^{99}Tc zone – is being studied [156]. The moderated target subassemblies would be loaded in the radial blanket region of the fast reactor. This concept seems promising, since a maximum ^{99}Tc transmutation rate was calculated to be more than double to reach about 10%/year.

The transmutation performance of ^{129}I has also been calculated [156]. ^{129}I was assumed to be loaded as NaI with an isotopic concentration of 76% ^{129}I. In the most effective case, the transmuted amount was 18 kg/year, which is about the output from three PWRs.

2.4.1.3 *Transmutation in light water reactors* [153,155]

The irradiation of ^{99}Tc and ^{129}I in standard PWRs has been considered in calculations with fission products located in special target pins without fuel, inserted into the guide tubes of the PWR assembly. ^{99}Tc was assumed in metallic form, at a density of 10.5 g/cm^3. Iodine was considered, as in [157], to be in the form of cerium iodide (CeI_3), with an iodine density of 4 g/cm^3 of which 76% is ^{129}I.

Absorption by the FP-containing clusters reduce reactivity in the core. To prevent this from interfering with operation, either they would be gradually drawn by a similar mechanism as for the control rods and the cycle would stay unaffected, or they would remain loaded, shortening the cycle.

Table II.15 gives for ^{99}Tc some calculated annual transmutation rates, together with the inventories and the transmutation half-lives. Transmutation is more effective in UO_2 fuel than in MOX fuel, owing to the softer neutron spectrum. The ^{99}Tc production of 1.6 reactors could be transmuted, which means that two on three PWRs should be loaded with such Tc targets to ensure equilibrium between production and consumption. For ^{129}I, the necessary ratio should be 2 out of 5 PWRs. PWRs are thus much less efficient than fast reactors, and would require special management of target pins.

2.4.1.4 *Transmutation in heavy water reactors* [153]

Deuterium as moderator has a lower absorption than hydrogen, so a high moderation ratio is tolerable with a low fissile content, giving a soft neutron spectrum particularly suitable for transmuting those fission products without epithermal resonances.

Several cases have been calculated with ^{99}Tc or ^{129}I in a HWR core, not only to determine their transmutation rates, but also to calculate the effects of these fission products on the reactivity coefficients, especially on the coolant void coefficient which could be positive for HWRs. The transmuted amounts of ^{99}Tc should be compared with the production of one 1-GWe LWR, which equals about 21 kg/year. In all cases, the ^{99}Tc loading equals about 3.8 t and additional enrichment is required. The most effective transmutation of ^{99}Tc is achieved when pins are placed in the moderator: the net ^{99}Tc transmutation rate equals about 81 kg/year, i.e. the production of four PWRs.

^{129}I and ^{99}Tc might be loaded in the centre pins of all fuel bundles. The net transmutation of ^{129}I would be 43 kg/year, about the production rate of nine PWRs. The coolant void coefficient would be strongly affected, demanding careful evaluation of the consequences. An increase in the fuel enrichment is unavoidable.

2.4.1.5 *Transmutation in thermal high flux reactors* [153,158]

Using of a thermal high flux reactor (HFR) may shorten transmutation half-lives. As a typical example of such a reactor, the Petten HFR was chosen for calculations; the conclusions are also valid for other thermal reactors with similar spectrum and flux level.

Calculations were done on a special subassembly containing three ^{99}Tc and six ^{129}I target pins. The transmutation half-lives were found to be about 8 years for ^{99}Tc and 5 years for ^{129}I. Because the power (40 MWt) and size of this HFR are small, no large amounts of ^{99}Tc or ^{129}I can be transmuted, but reactors with similar flux levels and higher power could perhaps be constructed in the future.

Similar calculations were made for ^{129}I and ^{99}Tc in the Belgian High Flux Reactor (BR2) which has a neutron flux of 3×10^{14} n/(cm^2·sec) [158]. For iodine the target was CaI_2 and for Tc the metal form. The calculated annual transmutation ranged between 6 and 9% for ^{129}I and 3 to 6% for ^{99}Tc. The major problem is the chemical stability of the CaI_2 target during irradiation and the very long transmutation half-life for both ^{129}I and ^{99}Tc of about 12 years or more.

2.4.1.6 Conclusion

A ranking of some reactor types with respect to transmutation rates and half-lives is given for ^{99}Tc in Table II.15. The transmutation rates should be compared with the production rate in a 1-GWe PWR, which is 21 kg/year or about 0.02 kg/MWe·year. Fast reactors with target pins loaded in a core sub-assembly (with or without moderation) seem best, as they would burn the ^{99}Tc production of up to five PWRs. The second best option would be to load targets in the moderator of a HWR. Transmutation rates in standard PWRs are much lower.

Generally, transmutation of ^{99}Tc or ^{129}I in present reactors is not encouraging because of the long transmutation half-lives and the huge inventories of fission products required. Special-purpose high flux reactors could improve the prospects.

2.4.2 Nuclear data on fission products

The thermal neutron capture cross-section and resonance integral have been measured for ^{99}Rc, ^{129}I and ^{135}Cs using the TRIGA Mark-II reactor at Rikkyo university. For some nuclei, these data differ very much from previous values [159].

Table II.15 **Ranking of reactors with respect to ^{99}Tc transmutation capability**

Reactor	**Configuration**	**Inventory ^{99}Tc (kg)**	**Transmutation ^{99}Tc (kg/year)**	**Transmutation ^{99}Tc (kg/MWe-year)**	**Half life (year)**
FR	Moderated S/A in inner core	2 741	122	0.11	15
FR	Non-moderated S/A in inner core	2 662	101	0.09	18
LWR	Pin in guide tube UO_2 fuel	3 633	64	0.07	39
LWR	Pin in guide tube MOX fuel	1 907	17	0.02	77

3. DESCRIPTION OF CURRENT TRENDS IN P&T RESEARCH

3.1 Current R&D activities

Since the launching of the OMEGA programme in Japan in 1988, an interest in P&T option has been renewed throughout the world. France and Japan have set up comprehensive study and experimental programmes. In France the SPIN programme comprising the PURETEX and the ACTINEX phases is part of a global waste management strategy established by law. During the past decade, the OMEGA programme has steadily progressed in the field of partitioning, reactor- and accelerator-driven transmutation and will be reviewed in the near future by the Japanese authorities.

An extensive overview of the OMEGA and SPIN programmes is given in the Annexes B and C.

3.1.1 OMEGA programme

In 1973, the Japan Atomic Industry Forum published the report on the waste management of long-lived nuclides after two years of assessment studies by groups of scientists and engineers in Japan. The report entitled "A closed system for radioactivity" pointed out the importance of research and development for partitioning and transmutation (P&T) of long-lived radionuclides as a long-term strategy for waste management.

Japan Atomic Energy Research Institute (JAERI), the Japan Nuclear Cycle Development Institute (JNC) and the Central Research Institute of Electric Power Industry (CRIEPI) proposed to initiate a major R&D programme based on their studies. The development of the technology was deemed to be an interesting subject for ongoing investigation from the perspectives of possible long-term advances in radioactive waste management and potential utilisation of resources.

In 1987, Japan's Atomic Energy Commission (AEC) concluded that the potential benefits from the use of some elements among the fission products, and from recycling minor actinides for power generation, could be achieved provided that a well planned, efficient and effective R&D programme could be formulated. The AEC then submitted in October 1988 a report entitled "Long-Term Programme for Research and Development on Nuclide Partitioning and Transmutation Technology", which plots a course for technological development up to the year 2000. The programme is called "OMEGA" which is the acronym derived from Options Making Extra Gains from Actinides and fission products. The R&D programmes comprise effort by JAERI, JNC and CRIEPI.

In January 1989, the Japanese government (represented by Science and Technology Agency; STA) proposed an international co-operation for information exchange to cover the areas of nuclear physics, reactor physics, advanced process technologies and physico-chemical characterisation relevant to P&T technology under the auspices of the OECD Nuclear Energy Agency.

The OMEGA-programme is to be proceeded in two steps: the phase I was originally intended to cover a period up to about 1996, and the phase-II to about 2000. In general, the basic studies and researches are to be conducted in the phase-I to evaluate various concepts and to develop required technologies. In the phase-II, engineering tests of technological or demonstration of concepts are planned. After 2000, pilot facilities will be built to demonstrate the P&T technology. The first check and review of the phase-I of the programme was scheduled in late 1998.

Following items are being studied:

- physical and chemical properties of minor actinides and fission products;
- partitioning of high-level liquid waste from reprocessing process and recovery of useful metals;
- transmutation: nuclear and fuel property data of minor actinides, system design study, reactor fuel and accelerator target development, and development of a high power accelerator for transmutation.

JAERI has been developing technologies for a dedicated partitioning process and transmutation system based on the double strata fuel cycle concept. JNC has been devoting its major efforts to develop an advanced fuel cycle system with TRUEX process for U, Pu and MA co-extraction and with MOX-FBR for transmutation. CRIEPI has been developing an advanced recycle technology based on pyroprocess and on metallic-fuel FBR.

3.1.2 SPIN programme

The SPIN programme was launched by the CEA in 1992 following the law voted by the French Parliament in December 1991, concerning highly active and long-lived radioactive waste management. The law identifies three axis for further research in this field, in order to investigate and to assess before 2006 the different possible ways (P&T, geological disposal, long-term interim storage); the SPIN programme is the answer to the first axis of the law, concerning the investigation and the assessment of the possible routes for partitioning and transmutation of long-lived radionuclides.

The programme is supported by the French Government, and also by industrial operators (EDF and COGEMA mainly). The results obtained are annually submitted for examination to the French National Assessment Committee, instituted by the law.

The management of irradiated fuels in France was described by EDF at Global'95. It favours the recovery of plutonium from irradiated UO_2 fuels and its recycling in MOX for eventually twenty-eight 900 MWe-PWRs. After that, the resulting reprocessing wastes no longer contain significant amount of plutonium. The minor actinides (Am, Cm, Np) and the fission products, including long-lived (> 30 years), are currently vitrified and stored. The spent fuel in excess (mainly MOX) will also be temporarily stored.

The SPIN programme studied various technical methods aimed at modifying the composition of the wastes:

- the PURETEX programme for medium-active waste: the main goals are a best recovery of the plutonium, and waste volume reduction ;
- the ACTINEX programme for highly-active waste: this programme concerns partitioning and transmutation of, on one hand, minor actinides (which are the main contributors to the long-term radiotoxic inventory of such waste), and, on the other hand, some long-lived fission products (which are to be considered, owing to their relative greater potential mobility under storage or geological disposal conditions).

The main results obtained can be summarised as follows:

- a very significant reduction of the amount of medium-active waste has revealed feasible and been achieved by COGEMA at industrial scale in "La Hague" plants since their commissioning; hulls remain bulk contaminated by long-lived elements, prohibiting their surface storage;
- the PUREX process already separates U, Pu and I and could perhaps be extended to Zr, Tc and Np. Further research requiring complementary extraction steps, is needed for the separation of Am, Cm, Cs and of other long-lived fission products;
- the transmutation of MAs is feasible in fission reactors (critical or subcritical), in particular if fast neutron spectra are envisaged. The transmutation of long-lived fission products needs a relevant neutron excess, in particular if elements (and not isotopes) are considered for transmutation. Different modes of recycling can be envisaged (homogenous and heterogeneous). In the case of Am, a "once-through" irradiation of targets in moderated subassemblies is an option, which is presently under study. However, the combined management of Am and Cm could lead to reduce more significantly the source of potential radiotoxicity. Several options are examined in this respect:
  - separation of Cm, to be let to decay in a specific installation, in order to recover the resulting Pu, to be further recycled;
  - "once-through" irradiation of Am + Cm targets, with the objective of > 95 % cumulative fission (irradiation length > 20 years);
  - use of dedicated reactors, to be fuelled with some appropriate mixture of Am + Cm (and eventually some Pu); safety considerations (e.g. low β_{eff} with critical configurations) lead to the evaluation of ADS.

R&D activities are performed today in all these fields in particular in basic chemistry (thermodynamics ; molecular modelling …) and physics (e.g. nuclear data), and in more applied fields: separation processes design and hot tests in the ATALANTE facility; core concepts experimental validation, fuel studies … A significant irradiation programme has been drawn up, mostly performed or to be performed in Phénix, but also in the frame of collaborations (e.g. the European collaboration EFTTRA; collaboration with the Russian RIAR Institute at Dimitrovgrad, etc). As far as ADS, R&D activities are performed for the experimental subcritical neutronics validation, in the intense accelerator development field and in the material studies (related to the window and target). These activities are performed by CEA in close co-operation with the French National Research Institute CNRS, in the frame of a joint programme (GEDEON).

As a conclusion, the SPIN studies should shed light on the type and amount of wastes produced under the various partial or complete recycling options for plutonium and minor actinides in a power reactor park, to define the technical operations to be performed, and to evaluate their cost over uncertain time frame. This leads to the emergence of new concepts, as new extractants for partitioning, or innovative systems to transmute minor actinides and long-lived fission products.

3.2 Summary of current strategy studies

Strategic assessment studies of P&T have been undertaken in Europe and in Japan. The Japanese study was conducted by JNC and emphasised the role of FRs and Actinide Burner reactors. An important strategy study has been undertaken under the leadership of CEA in the framework of the 3rd European Union R&D programme on Nuclear Fission. This strategic assessment programme has been continued on an international basis within the European Union and expanded during the current R&D programme (1994-1998). These studies are summarised below.

3.2.1 European Union strategy study [77,160]

3.2.1.1 Reference and P&T scenario

Reference scenarios with and without conventional reprocessing, and scenarios using P&T are compared to assess their possibilities.

The three reference scenarios are considered:

- the Rl scenario covers the period from 2000 to 2100. The reactor population consists of PWRs supplied with UO_2 fuel at 4% ^{235}U enrichment and reaching a mean burn-up of 47.5 GWd/tHM. The installed capacity is 120 GWe, i.e. 80 reactors, with an annual electrical production of 740 TWh (roughly the present installed generating capacity in the European Union). The fuel cycle is open without reprocessing.
- scenarios R2 and R3 both include a plutonium recycling strategy but in different types of reactors. In the R2 scenario, plutonium is recycled as MOX fuel in PWRs. The fuel cycle is closed by PUREX reprocessing with the losses of 0.3% for U and 0.5% for Pu. In the R3 scenario it is recycled in fast reactors(FRs: 1 500 MWe) and the losses during FR fuel reprocessing are 0.9% for U and 0.25% for Pu. Recycling in PWRs is assumed to be applicable from the outset of the scenario (in the year 2000) while recycling in FRs is assumed not to begin before 2020, considering the lack of industrial maturity in this solution. The two scenarios therefore differ only after 2020.

Three scenarios are considered for partitioning and transmutation, two with available technologies, RP1-1 and RP1-2, and one with very advanced technologies, RP2:

- the RP1-1 scenario is compared with the R2 scenario. The transmutation of Np and Am starts from 2010 in PWRs in homogeneous or in heterogeneous mode. In homogeneous mode, neptunium or americium oxide is mixed with the UO_2 fuel to the extent of 1%. The losses during reprocessing are 0.3% for U, 0.5% for Pu, 5% for Np and Am and 100% for (Cm).

- the RP1-2 scenario is compared with the R3 scenario; as the minor actinide partitioning starts in 2010, Np and Am are stored before being recycled homogeneously or heterogeneously in FRs after 2020; in homogeneous mode, an amount of Np or Am representing 2.50% of the metal mass is mixed with the FR fuel.

- the RP2 scenario is similar to the RPl-2 scenario until 2030. CAPRA (Consommation Accrue de Plutonium en Réacteur Rapide) type FRs progressively start operation after 2030. They are still at the preliminary design phase and are dedicated 1500 MWe incinerators loaded with MOX containing 45% of Pu; they transmute Np in homogeneous mode and Am, Tc and I in heterogeneous mode. The fuel and targets are reprocessed with losses of 0.1% for U and Pu, 0.5% for Np, Am and Cm and 10% for Tc and I; Cm is placed in interim storage.

3.2.1.2 Potential radiotoxicity (Figure II.17)

Radiotoxicity has been chosen as a measure of the potential detriment of the waste resulting from the different scenarios analysed. The radiotoxicities are assessed for ingestion of all heavy radionuclides and three long-lived fission products (^{99}Tc, ^{129}I and ^{135}Cs).

Concerning the reference scenario, recycling in FRs (R3 scenario) leads to a larger decrease in radiotoxicity than recycling in PWRs (R2 scenario). The difference in reduction is by a factor larger than 5 for the R2 scenario between 10^4 and 10^5 years due to the recycling of plutonium. In the short period of $10 \sim 10^3$ years, the radiotoxicity is primarily due to ^{244}Cm, then to ^{241}Am. In the long period of $10^3 \sim 10^5$ years, the paramount contributions are from ^{243}Am, its daughter ^{239}Pu and from ^{240}Pu. In the very long period of more than 10^5 years, the radiotoxicity is dominated by ^{237}Np and the decay products of uranium.

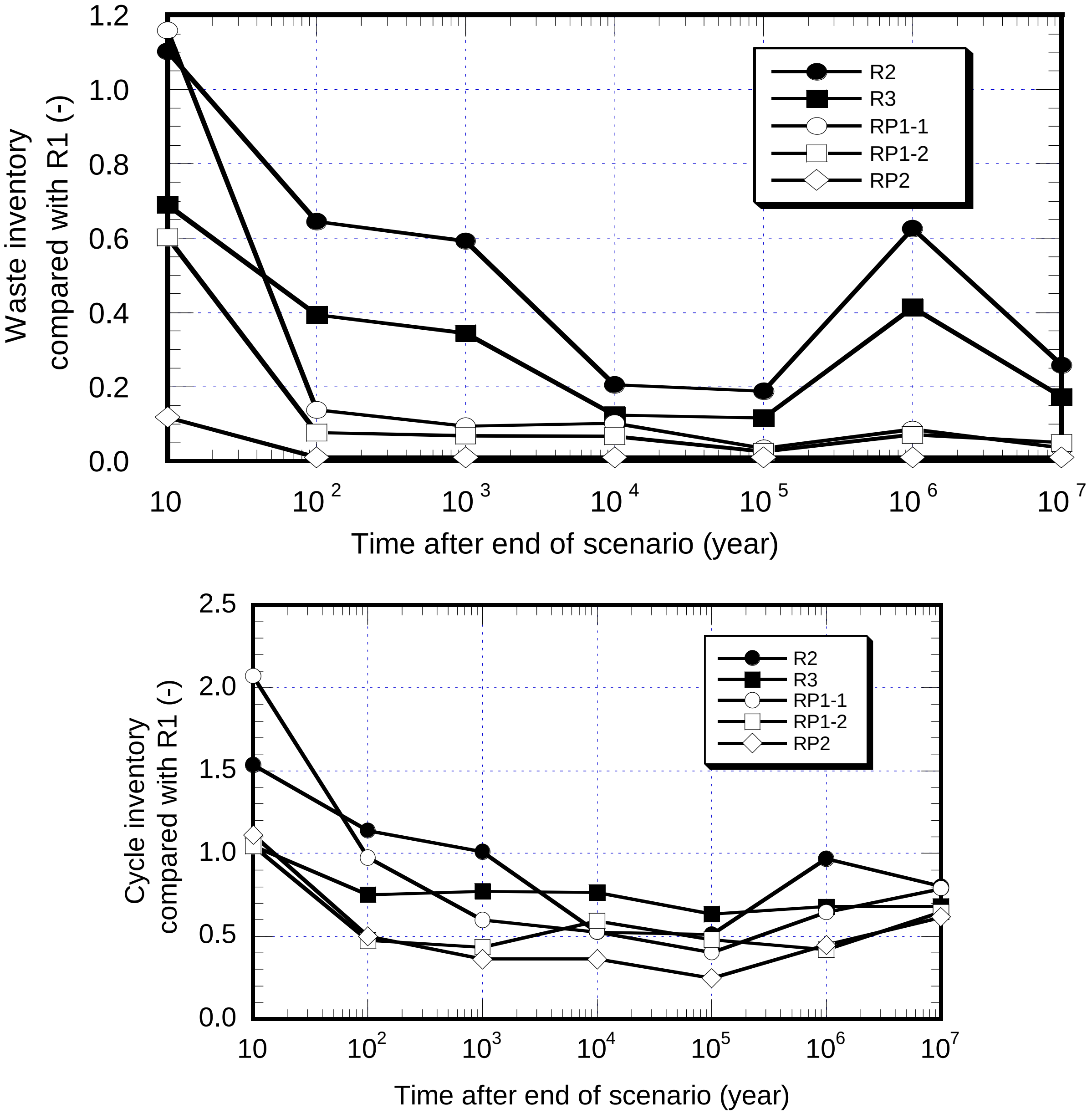
The reductions in radiotoxicity for the RP1-1 and RP1-2 scenarios are nearly the same as in the R2 and R3 scenarios respectively. There is an improvement by a factor of 6 between 10^2 and 10^3 years because of the decrease in Am inventory and similarly between 5×10^5 and 5×10^6 years due to the smaller Np content. For short time periods (<100 years), the gains are offset by the increased production of Cm. This effect appears to be more detrimental to homogeneous recycling, because the curium produced in the Am targets during heterogeneous recycling is allowed to decay to usable Pu and therefore not included to the waste.

For the RP2 scenario (not shown in the figures), the radiotoxicity is reduced by a factor of more than 10 with respect to the R2 scenario for time periods smaller than 100 years, because curium is put in interim storage. For the other time periods, the reduction factor for the radiotoxicity is between 10 and 30.

3.2.2 Studies in Japan

The analysis of MA mass balance was performed for two types of nuclear reactors namely a MOX-fuelled FBR and a specially designed actinide burner reactor for efficient burning of minor-actinides.

Figure II.17 **Radiotoxicity balances**



3.2.2.1 MOX-fuelled FBR [161]

Production of minor actinides

Minor actinides are produced in the cores of fission reactor (light water reactor and fast reactor) by the neutron absorption and decay reactions. In particular, almost all ^{241}Am is generated during spent fuel cooling by the β-decay of ^{241}Pu. A 1 000 MWe-LWR operating for a year discharges 24 tons of spent fuel containing 22.6 tons of uranium, 1 120 kg FPs, 250 kg Pu, and 20 kg MAs.

Mass balance

The MA mass balance, for instance in the advanced fuel recycle to reduce the accumulation of MAs and the MA concentration in FBR core fuel, was analysed according to the predicted nuclear energy production in Japan.

Plutonium and MAs are recovered from the LWR and Pu-thermal reactor, and recovered Pu and MAs are multiply recycled into fast reactors. Nuclear power generation is assumed in two separate cores to increase to 1 000 MWe/year or 1 500 MWe/year with the introduction of commercial fast reactors starting in the year 2030. New reactors are assumed to be totally FBR, and all spent fuel discharged from LWR and Pu-thermal reactors is assumed to be reprocessed.

In Case-1 (1 000 MWe/year), the total MAs transferred into the high level waste are calculated to be 310 tons from LWR, Pu-thermal LWR and FBR without recycling. In the case of recycling MAs into commercial FBRs after the year 2030, the MAs remaining in the fuel cycle in the year 2100 is reduced to about 60 tons, 80% less than without recycling.

3.2.2.2 Actinide burner reactor

The effect of introducing the transmutation system was investigated as a function of its initial MA loading, annual transmutation rate and initial introduction year (2010 or 2020). The conditions were:

- the amount of MAs annually produced from UO_2-LWRs should be equivalent in the year 2050 to that annually incinerated;
- out-of-pile storage capacity requirement for MAs should be zero in the year 2100.

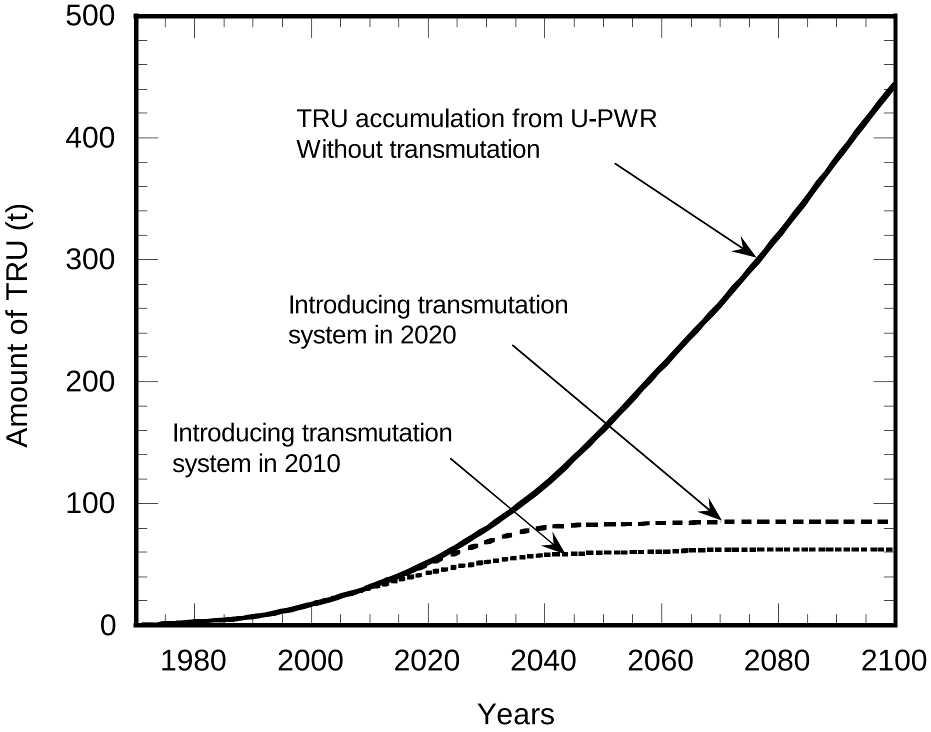
The calculated results show that the conditions can be satisfied by introducing realistic transmutation systems at the rate shown in Figure II.18.

A conceptual design study has been carried out on two types of MA transmutation systems which are specially designed for efficient burning of MAs.

The first is the “Actinide Burner Reactor” (ABR) which is a kind of fast reactor with a very hard neutron spectrum. The loading capacity of a 1 000 MWt ABR corresponds to less than 3 000 kg of MAs and the ABR transmutes more than 300 kg of MAs annually. The second is the proton accelerator-driven MA transmutation system composed of an intense proton accelerator and a subcritical fast reactor core. The MA inventory of the system is about 3 000 kg and its MA transmutation capability corresponds to about 300 kg/GWt-year.

Assuming that deployment of the transmutation systems begins in the year 2020, and the low-growth case is adopted as the nuclear power supply scenario, about fifteen 1 000 MWt-units of the proposed system are capable of compensating for MA generation from nuclear power plants and limiting their out-of-pile storage inventory to a very low level at an early stage.

Figure II.18 **Effect of transmutation on reduction of minor actinide accumulation**



4. IMPACT OF P&T ON RISK ASSESSMENT AND WASTE MANAGEMENT

4.1 Introduction and definitions of radiotoxicity and risk

4.1.1 Risks in the back-end of the fuel cycle

In speaking of radioactive waste management strategies, one frequently mentions (1) the risks associated with each strategy, and especially those inherent in it, (2) the extent of these risks, and (3) the radiotoxicity of the waste.

While the definition of the risks merely demands precision of language, risk assessment is very difficult. The term “radiotoxicity” is often qualified, without clarification, as “potential” or “residual”, and it is often confused with risk. These terms must, therefore, be explained.

According to the nomenclature of the World Health Organisation, risk is a quantified evaluation of the danger, and is expressed in terms of probability. Hazard is defined as the cause of a detriment, and is not quantifiable.

The danger that concerns us here arises from the radiation emitted by radionuclides and the effect of exposure to it on living matter.

The hazard posed by the chemical toxicity of the radioactive element is generally much less significant and seldom taken into consideration.

In a given set of conditions, exposure of matter (tissue or organ) to radiation delivers an absorbed dose, D (Gy) which is in principle measurable. The unit is joule per kg, and it is called as gray (1 Gy = 1 J /kg).

In the case of high exposure (several grays) delivered in a short time interval, the effects are known, and are somatic effects of a deterministic nature. These effects appear above a threshold and their gravity depends on the dose. In theory, the probability of their occurrence is one above the threshold and zero below it. However in practice the threshold for various deterministic effects is a broad range of values rather than a single number.

In the case of low exposure (i.e. smaller than doses at which deterministic effects appear) delivered over an appreciable time interval, cancers and genetic effects can be induced, although they may appear only after many years or even decades. These are probabilistic effects with a likelihood considered by the ICRP to be proportional to the dose, without a threshold.

At low doses, it is considered that the effect of ionising radiation on living matter depends upon the radiation type, R, and on whether the dose is to the whole body or to particular parts. This is incorporated in the notion of effective dose to the organism, E (Sv) which is related to the dose absorbed in a tissue or an organ exposed to radiation R, D_{TR} (Gy) by:

$$E_R = \sum_T W_T \sum_R W_R D_{TR}$$

where W_T is a weighting factor for tissue T, and W_R is a quality factor, which is nevertheless called a weighting factor, for radiation R. Values of W_T and W_R are recommended by the ICRP Publication 61 [164], The equivalent dose H_T is expressed as:

$$H_T = \sum_R W_R D_{TR}$$

and represents the threshold dose to a particular tissue. This weighted dose is expressed in J/kg, but this unit is given the name of sievert (Sv).

Thus the gray must be used in speaking of deterministic somatic effects, and the sievert must be used in speaking of stochastic effects.

In a given set of exposure conditions, each becquerel of a given radionuclide will cause an effective dose E_N (Sv/Bq) which depends on its nuclear properties (see some examples for internal exposure in Tables 2.16 and 2.17). The conversion factor from activity to dose is thus E_N (Sv/Bq).

Exposure due to a set of radionuclides of given activities, A_{RN}, thus leads to a value of the effective dose to the organism, E:

$$E(Sv) = \sum_{RN} E_{RN}\,(SvBq^{-1}) \times A_{RN}\,(Bq)$$

We shall return to these points in discussing incorporated radionuclides.

4.1.2 Exposure related risk

The risk associated with exposure can be called Radiological Risk, RR, (not to be confused with Relative Risk also sometimes abbreviated as RR), and can be expressed by a general formula of the type:

$$\begin{aligned} RR\ (\text{time}^{-1}) = \ & [\text{probability of a detriment per unit dose(dose}^{-1})] \\ & \times S\ [\text{probability of occurrence of an event } i\ (\text{time}^{-1})] \\ & \quad \times [\text{dose delivered by event } i\ (\text{dose})] \end{aligned}$$

Hence:

$$RR\ (\text{time}^{-1}) = \quad p\ (\text{dose}^{-1}) \times S\ P_i \times (\text{dose})_i$$

For deterministic effects, in the case of high exposure over a short-time interval, the doses are expressed in grays, p = 0 below the threshold, and p = 1 above it. The time taken into account can be the year or a shorter time interval depending on the time of appearance of the deterministic somatic effects. Hence:

$$RR = \sum P_i \times (dose)_i$$

For stochastic effects, in case of chronic low exposure, the doses are in *Sv* and p = 0.063 Sv^{-1} according to the ICRP recommendations (0.05 for a mortal cancer and 0.013 for a hereditary effect). The time is expressed in years, and the detriment in fact does not appear before a latency period of, say, fifteen-old years, hence:

$$RR\ (year^{-1}) = p\ (Sv^{-1}) \sum P_i\ (year^{-1})\ E_i\ (Sv)$$

4.1.3 Radiotoxic inventory of radionuclides and irradiated fuel

In order to give a quantitative meaning to the concept of radiotoxicity, the term "radiotoxic inventory" may be introduced. When a radionuclide enters the body by ingestion or inhalation, it is denoted FD_{RN}. The calculation of this value obviously depends on the physical properties of the radionuclide, but also, and above all, on its post-incorporation biokinetics. If exposure is assumed to take place over a fifty-year period, assigning the dose to the first year after contamination (referred to as the committed dose), the committed effective dose per unit intake is denoted FD_{RN} (Sv/Bq) and depends on the mode of intake (ingestion, inhalation). The radiotoxic inventory of any toxic radionuclide, $R_{RN}(Tx)$ can then be defined as:

$$R_{RN}(Tx) = A_{RN} \times FD_{RN} \times K$$

where K is a normalising factor depending on the terms in which the activity is expressed, for example in Bq per unit of quantity of matter (g, kg, t) containing the radionuclide, or in Bq resulting from the production of a given unit of energy (J, Wh, TWhe) or simply in Bq per unit of time (s, min, year). Thus it is expressed in Sv/t or in Sv/TWhe or even in Sv/year depending on the situation.

Given the use of FD_{RN}, it is clear that this definition is associated with exposure to low doses. Hence it is an excellent tool for expressing the radiotoxicity of the radionuclides which could be incorporated in one year, in small amounts, like those that could appear in the biosphere as a long-term result of radioactive waste storage.

Yet it is also commonly used to express the radiotoxic inventory of spent fuels. Thus, for a spent fuel, the radiotoxicity is:

$$R_{FE}(T_X) = \sum_{RN} R_{RN}(T_X)$$

It can be expressed in Sv/t if one considers the activity of the radionuclides which it contains in 1 t, or in Sv/TWhe if one considers the activity of the radionuclides which have been formed in the fuel to produce 1 TWhe (see Figures II.21-II.23 as examples).

This being said, prudence is the watchword concerning this definition and its general meaning. This emerges in considering the incorporated activities to be taken into account. If one assumes that these activities result from the incorporation of the fuel radionuclides by a single individual (which is obviously unrealistic), this leads to doses with a deterministic effect to which the Sv unit does not apply as we have seen. Thus the radiotoxic inventory of the fuel must be understood as an expression of the collective dose [162] that would be received by numerous individuals if they incorporated the activity of the fuel at rates which do not trigger deterministic effects. In this case the Sv unit can be used. Each expression for radiotoxic inventory corresponds to a specific need.

Expressed in Sv/t or in Sv/TWhe, it is an operating and management tool for the production of the waste, and, expressed in Sv/year, it is a management tool for a waste disposal project.

4.1.4 Assessment of the radiological risk

Evaluating the Radiological Risk, RR, requires associating the values of P_i and of the doses D_i or E_i whenever possible. This is where matters become complicated, particularly for events in the distant future.

In relation to the back-end of the cycle, many factors must be considered:

- the periods of time, because the activities which can cause exposure are or will be spread over long periods between the start of interim storage of the spent fuels and the disposal of the wastes;
- the strategy, which may be non-reprocessing or some form of processing for the spent fuels;
- the scenarios, normal and accidental, are based either on a sequence of natural events, or are influenced by man and dependent on the installation. Each of these scenarios is itself dependent on two types of parameters: physicochemical and sociological.

The evaluation of RR also depends on:

- the methodology used: deterministic with analysis of sensitivity to parameters, or probabilistic;
- the modelling (radionuclide transfers to the biosphere and calculation of impact on man);
- the quality of the “tools” and the data.

Once the RR has been evaluated, decisions must be taken.

4.1.5 Decision framework

Decisions are taken on the basis of the Safety Analysis. This consists of:

- comparing the RR values with a number of considerations including:
  - safety objectives: environment, present and future human health;
  - ethical considerations: principle of fairness, principle of precaution based on beliefs in the invariability of the characteristics of man, of society, and of the advancement of knowledge;

- accounting for a number of safety indicators which are not concerned with the individual but rather with society as a whole.

The assessment of the "risk proper" must also take account of factors such as:

- the estimation of the radioactive inventory and its dispersion: this point is important because the concepts of half-life and specific activity are important, and one cannot incorporate a large amount of activity of a long-lived radionuclide without raising a problem;

- the resources contaminated and the foreseeable contamination times;

- critical groups appropriate to each situation.

Decision making in this field is a complex trade between the conclusions of the experts in the different scientific disciplines and the sometimes subjective options taken by policy makers who are influenced by the sociological context of the moment.

4.1.6 Conclusion and recommendations

It is recommended to abandon the term "potential radiotoxicity", in speaking of nuclear wastes, because it gives the illusion of a management scenario for these wastes, whereas it is merely an inventory. It is preferable to express the inventories in Bq/t of fuel (or of heavy metal it contains) or in Bq/TWhe for each radionuclide. In the final analysis, what is measured is Bq and not Sv, and what is potential is the risk and not the radiotoxicity. Moreover, this would help to dispel the confusion among non-health physicists between "doses" (equivalent dose, effective dose and committed effective dose).

To visualise the comparison of the radionuclide inventories in terms of radiotoxicity, the use of the "radiotoxic inventory" would be preferable to that of "potential radiotoxicity". It would be easier to understand in so far as it preserves the correct notion of an inventory, while implicitly incorporating the weighting coefficients used in health physics. The evaluation of the risk in separation/transmutation must be based on safety analyses over time, which account for the different aspects: plant, interim storage and disposal. This risk must be compared with that of other strategies which are similarly evaluated.

4.2 Radiotoxic inventory of waste

The general strategy of introducing P&T as an additional waste management option is based on the radiological benefit which is expected from such an option. The ranking of the actinides and long-lived fission products can be made on the comparison of their intrinsic hazard (effective dose coefficients, Sv/Bq) coupled with their radioactive concentrations in spent fuel or HLW (Bq/tHM). The radioactive inventory (Bq) can also be related to drinking water standards [163] as it was defined initially, or to the more recent ALI (annual limits of intake) for comparison of their relative radiotoxicity [164,165].

The recent ICRP publication with a comprehensive data overview lists the effective dose coefficients FD_{RN} in Sv/Bq for workers [165] and the general public [166] in the nuclear field. These data are not of direct application to the long-term risk assessment but are the fundamental basis for the assessment of the radiotoxicity ranking of radionuclides.

Based on this criterion, the long-term radiotoxic inventory depends on the source term which is determined by the type of fuel (LWR-UO_2, LWR-MOX, FR-MOX), the burn-up and the storage time (up to a million years). These fuels contain the actinides and the long-lived fission products as major radiotoxic constituents.

In terms of hazard factors the following ranking can be made [167] for spent fuel seven years after discharge from the reactor.

$$^{238,239,240,241,242}Pu > ^{241,243}Am > ^{242,244,245,246}Cm >> ^{237}Np$$

However the short-term radiotoxic inventory of some fission products is comparable to that of the actinides within a time horizon up to 100 years. Beyond 300 years only the long-lived fission products remain radioactive (^{99}Tc, ^{93}Zr and ^{135}Cs) and constitute a radiotoxic inventory which is roughly 1 000 times smaller than that of the actinides. ^{129}I is in terms of effective dose coefficient (Sv/Bq) comparable with the actinides but its radiochemical concentration in the spent fuels, expressed in Bq/THM, is much lower. In vitrified HLW from reprocessing, the ^{129}I inventory is negligible.

Among the actinides the most important are Pu, Am and Cm, the effective dose coefficients [165,166] are given in the next Table II.16.

Table II.16. **Effective dose coefficients of actinides FD $_{RN}$ [165,166]**

Element	**Nuclide**	**Sv/Bq (ingestion)**
Uranium	^{235}U	4.6×10^{-8}
	^{238}U	4.4×10^{-8}
Neptunium	^{237}Np	1.1×10^{-7}
Plutonium	^{238}Pu	2.3×10^{-7}
	^{239}Pu, ^{240}Pu	2.5×10^{-7}
Americium	^{241}Am, ^{243}Am	2.0×10^{-7}
Curium	^{243}Cm	1.5×10^{-7}
	^{244}Cm	1.2×10^{-7}
	^{245}Cm, ^{246}Cm	2.1×10^{-7}

The long-lived fission products have toxicities which are very variable as shown in Table II.17.

In the case of the OTC all radionuclides contribute to the source term and the long-term radiotoxic inventory is mostly due to Pu, MAs and some LLFPs. However, the conditioning operations can provide artificial barriers which are potentially capable of confining the radionuclides within their package for thousands of years. After this time interval nothing can be predicted except that the solubility of the actinides (except Np) is generally very low whereas the long-lived fission products, particularly ^{135}Cs, ^{129}I and in some cases ^{99}Tc, display high mobilities in the geosphere.

Table II.17 **Effective dose coefficients of fission products FD $_{RN}$ [166]**

Element	Nuclide	Sv/Bq (ingestion)
Strontium	^{90}Sr	3.4×10^{-10}
Zirconium	^{93}Zr	8.6×10^{-10}
Technetium	^{99}Tc	6.4×10^{-10}
Iodine	^{129}I	1.1×10^{-7}
Caesium	^{135}Cs	2.0×10^{-9}
	^{137}Cs	1.3×10^{-8}

The general radioactive characteristics of the spent fuel as source term are calculated by computer programs (ORIGEN 2.1 or ORIGEN 2.S, KORIGEN and APOLLO). For a given standard burn-up (e.g. 40 or 50 GWd/tHM) the programs calculate for each element involved the mass, radioactivity, residual heat power, radiotoxicity etc. which are compiled in tables or graphs (see Annex E). Figure II.19 shows the typical evolution for each of the major components, actinides, fission products and activation products, as a function of time [168].

The total radioactivity of the spent fuel expressed per tHM 500 years after unloading will amount to about 200 TBq, which is 700 times lower than after one year cooling. During the first 200 years the radioactivity will be mainly due to the fission products and will drop from 1.4×10^{5} TBq after one year cooling to 200 TBq. After 200 years the actinides contribution (~300 TBq/tHM) becomes dominant and the radioactivity will decay very slowly. It will naturally decay to 100 TBq/tHM after 1 000 years and to 13 TBq/tHM after 25 000 years.

The residual heat will vary from about 2 kW/tHM one year after discharge to 65 W/tHM after 1 000 years. The radiotoxic inventory expressed as ingestion hazard (Sv/tHM) follows very closely the total alpha radioactivity inventory as a function of time. For UO_2 the curves for the individual elements are shown in Figure II.20. The plutonium isotopes and their decay products determine the radiotoxic inventory up to 1 000 000 years. In an extremely long time interval of several million years, Np and the daughter products of the uranium isotopes, determine the remaining radiotoxic inventory.

In the case of the RFC, the reprocessing operation and recycling of Pu as LWR-MOX significantly reduces the radiotoxic inventory of the HLW. In this case the long-term radiotoxicity is essentially determined by the minor actinides (Np, Am and Cm, see Figure II.21) and the long-lived fission products ^{99}Tc, ^{126}Sn, ^{79}Se and ^{135}Cs (see Figure II.22). An important long-lived fission product ^{129}I does not appear in this graph since it is discharged into the ocean as a consequence of the reprocessing operations. Between 100 and 10 000 years the most important radiotoxicity contribution in the HLW comes from Am and Cm isotopes. Beyond that period both isotopes have significantly decayed to ^{239}Pu, ^{240}Pu (daughters of ^{243}Cm, ^{244}Cm) and ^{237}Np (daughter of ^{241}Am) which become predominant.

The radiotoxic inventory of spent MOX fuel as a function of time is shown in Figure II.23. Since the Pu content of 7 spent UO_2 assemblies is put into one MOX fuel element, the actinide radiotoxic inventory of a spent MOX fuel element is much higher (about 8 times) than in spent UO_2 fuel elements. Beyond 1 000 years the radiotoxic inventory of actinides will decrease smoothly from 5×10^{8} Sv/tHM after 1 000 years to 1.2×10^{8} after 10 000 years and 5.0×10^{6} after 100 000 years.

Figure II.19 **Radioactivity of PWR type spent fuel (4.1% ^{235}U, 40 GWd/tHM)**

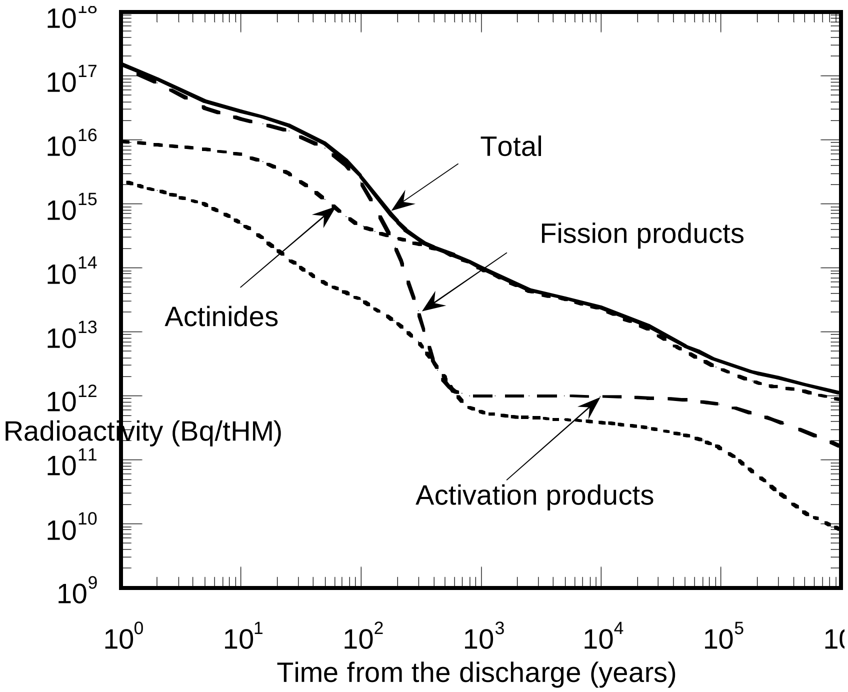
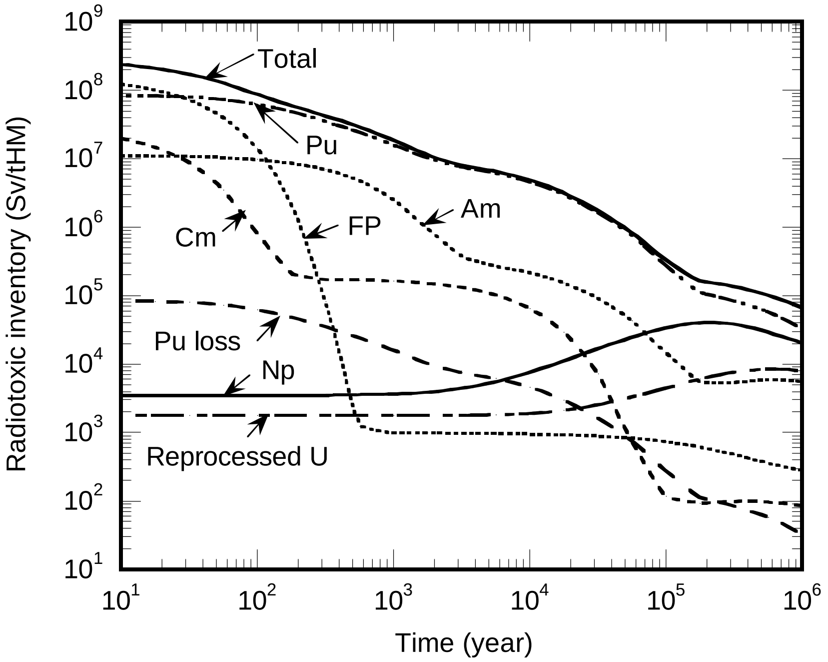


Figure II.20 **Radiotoxic inventory of UOX fuel as a function of time (3.7% ^{235}U, 45 GWd/tHM)**



In a long-term perspective of waste management the disposal of spent MOX fuel is a major factor in the overall assessment of the radiotoxic inventory in the RFC fuel cycle. Reprocessing of spent

MOX fuel and recycling of recovered plutonium are therefore important issues which have to be investigated in relation to radiotoxicity.

The waste management issues in case of an advanced fuel cycle (AFC) scenario are very different from the previous options. The impact of advanced reprocessing on the radiotoxic inventory of the HLW is quite striking, since the actinides (U, Pu, Np, Am and Cm) are ideally removed from the HLW with a high separation factor. During the first 200 years the fission products are dominant and the radiotoxicity of the actinides is of the same magnitude as that of the Pu losses (~0.1%).

Advanced reprocessing of MOX fuel with quantitative removal of Pu, Np and Am and Cm is also beneficial to reduce the long-term radiotoxic inventory of HLW glass (Sv/tHM_{EQUIV}), which will be essentially due to the fission products during the first 300 years, but in the long term the actinides losses will again be dominant although at a much lower absolute α-activity, and final radiotoxicity level will depend on the decontamination factor.

The main impact of the AFC strategy is a significant reduction in the radiotoxic inventory of the vitrified wastes with conversely a transfer of the long-lived actinides to the fuel cycle facilities and reactor core inventories. Comparing the radiotoxicity balances of the OTC and the AFC scenarios shows that the glass compositions become much more favourable. The radioactivity, although similar during the first few hundred years, decreases by factors of 10 and 50 after 300 and 1 000 years, respectively. The residual heat of the HLW becomes 3, 250 and 350 times lower after 100, 1 000 and 10 000 years. This is the main potential contribution of the AFC to waste management.

This long-term benefit has to be weighed against the short-term doses to workers, and the production of additional contaminated wastes, due to the increased complexity of the fuel cycle.

4.3 Possible impact o f P&T on TRU reduction

4.3.1 Necessary facilities to implement P&T

4.3.1.1 Inventories of minor actinides to be processed in a AFC scenario

The industrial reality involved in processing the MA quantities discharged [169] by a park of 100 GWe, i.e. the reprocessing of 2 000 – 2 700 tHM spent fuel with a final burn-up achieved of 40 to 55 GWd/tHM, is the treatment of the following annual inventories after 10 years discharge:

Element	g/tHM	kg/GWe-year	kg/TWhe	kg/100GWe-year
Neptunium	570 - 750	15.4	1.75	1 540
Americium	566 - 740	15.6	1.78	1 560
Curium	34 - 92	0.94 - 1.8	0.1 - 0.2	94 - 180

Figure II.21 **Potential radioactivity of actinides in the glasses from the standard reprocessing of PWR type spent fuel (4.1% ^{235}U, 40 GWd/tHM)**

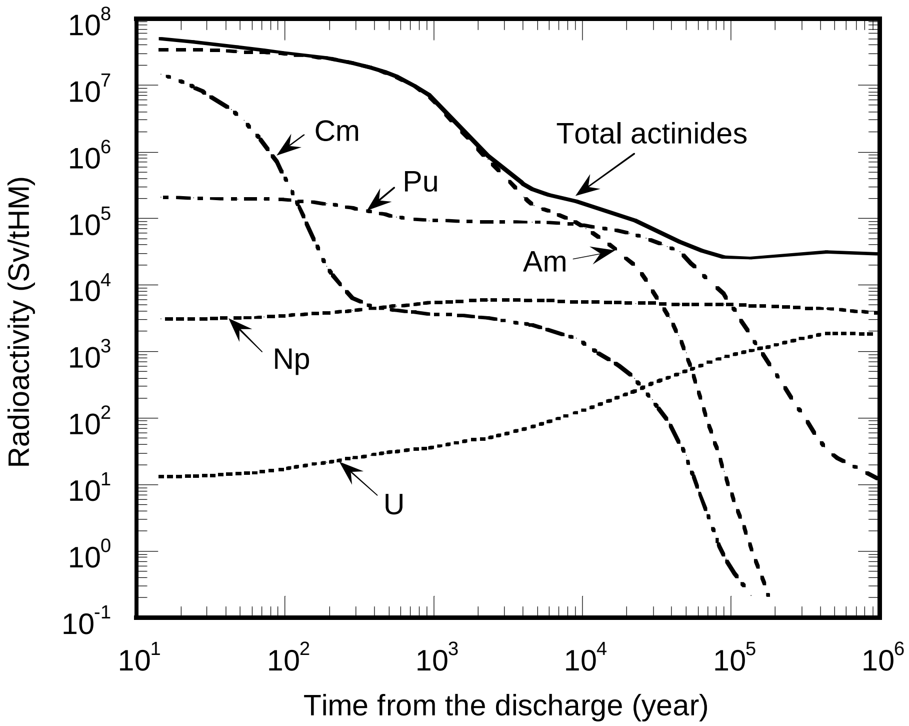


Figure II.22 **Potential radioactivity of main fission products in the glasses from the standard reprocessing of PWR type spent fuel (4.1% ^{235}U, 40 GWd/tHM)**

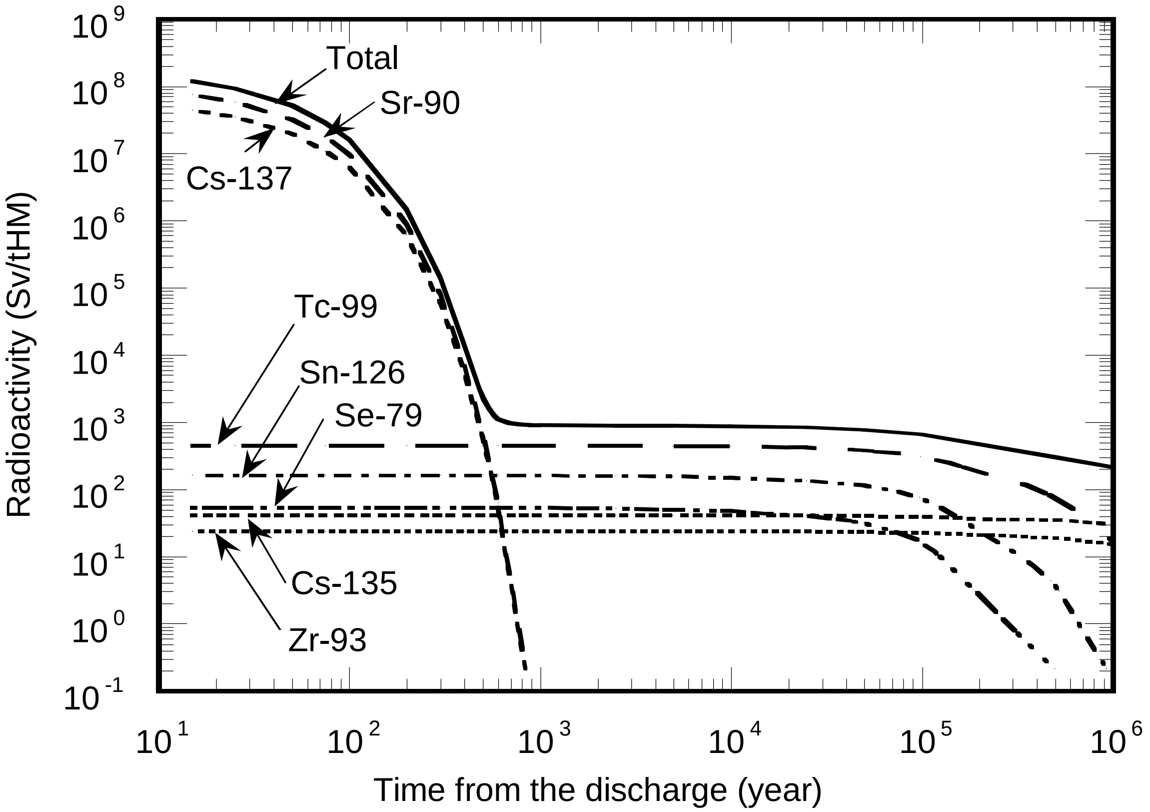
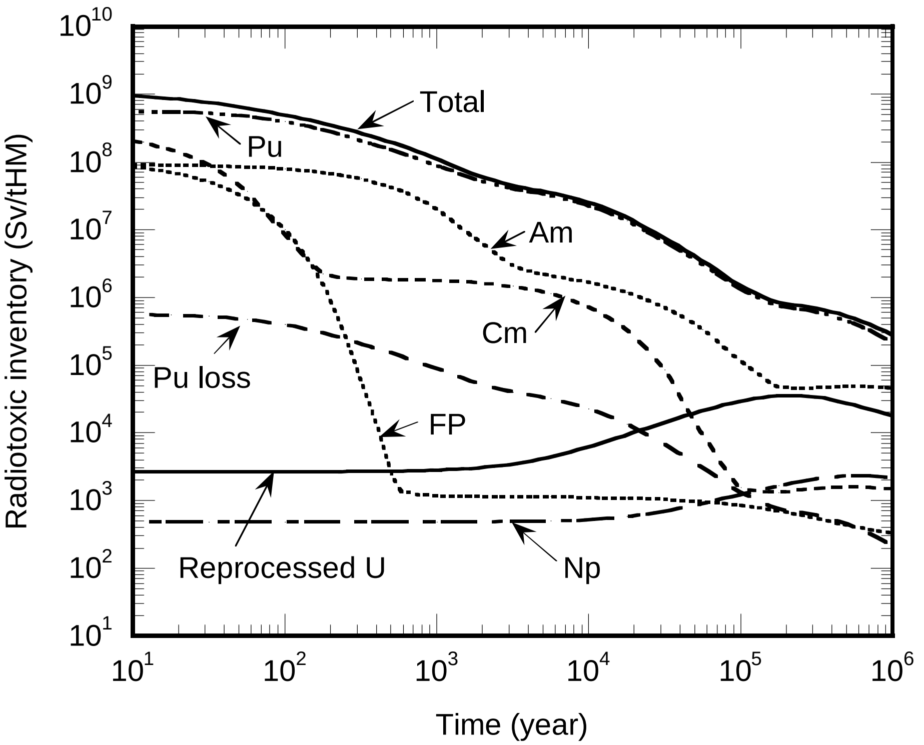


Figure II.23 **Radiotoxic inventory of MOX fuel as a function of time (8.28% Pu, 45 GWd/tHM)**



The facilities involved are plants to separate MAs from HLLW and fabricate fuel or targets, dedicated reactors for transmutation, and dedicated chemical or metallurgical process units for multiple-recycling.

In a first phase the necessary chemical facilities for separation of MAs from HLLW have to be designed and constructed. As can be deduced from the above table, a 100 GWe park will discharge annually 1.54 t of Np, 1.56 t of Am, and last but not least 94-180 kg of Cm. The corresponding fuel fabrication outputs are: 60 tHM U-Np fuel and 60 tHM U-Pu-Am fuel with 2.5% MA concentration. Provisional storage capacity would be needed for Cm covering e.g. 30 years production, i.e. 3 to 5 t Cm. All these facilities ought to be constructed on the reprocessing site in order to minimise transportation of these highly active concentrates. After conditioning and encapsulation, a transfer to a dedicated reactor site for transmutation should in principle be possible. However, if multi-recycling of the targets is required, either the irradiation facilities should be installed near the existing processing units or dedicated (e.g. pyrochemical) reprocessing facilities should be erected near the reactor buildings.

4.3.1.2 Recycling scenarios for power reactors [170,171]

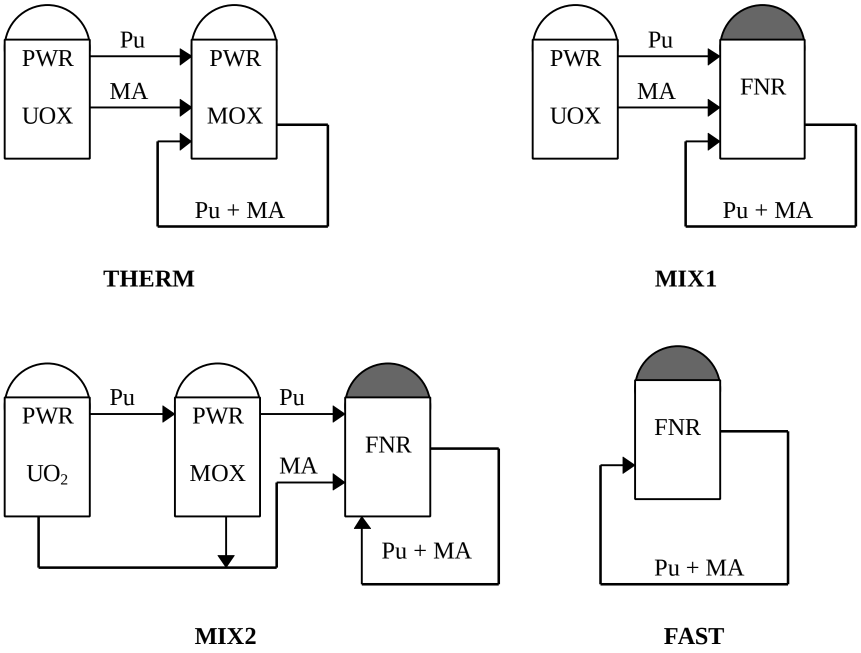
Research conducted in recent years has helped to clarify the feasibility of recycling actinides (mainly Am and Np) in the different types of reactor (FBR, PWR) in different recycle modes (homogeneous or heterogeneous).

Moreover, plutonium recycling and multi-recycling have been the subject of many studies. However, to reduce the radiotoxic inventory, it has been emphasised that any Pu multi-recycling strategy (whatever the aim of the strategy: Pu consumption, inventory stabilisation, etc.) must consider the essential goal of not increasing the production of minor actinides. Theoretically, the advantage of FRs (associated with lower σ_c/σ_f ratios in this type of spectrum) was illustrated in the physical approach described in the previous chapter.

According to present views and insights, a scenario with simultaneous management of plutonium and MAs seems possible in order to achieve equilibrium between production and consumption of Pu, Am, Np and Cm within the reactors. The source term of the radiotoxic inventory generated by these reactors would be significantly reduced. The overall losses in the different reprocessing and partitioning operations would represent the minimum radiotoxic inventory of the waste resulting from the nuclear electricity generation. Several reactor park compositions with balanced reactor types can be considered.

Figure II.24 shows a few examples.

Figure II.24 **Reactor park compositions**



THERM **MIX1**

MIX2 **FAST**

THERM	Reactor system consisting exclusively of PWRs, some of which are used for the multi-recycling of Pu and MAs.
MIX 1	A similar system, but the actinide multi-recycling function is performed by CAPRA type FRs.
MIX 2	A mixed system in which the Pu produced by PWR UO_2 is recycled once in PWR MOX. The residual Pu and all the MAs are recycled to CAPRA type FRs.
FAST	A reactor system consisting exclusively of EFR-type FRs.

Since these reactor systems all have the same installed capacity, they are said to be in equilibrium in two respects:

- the characteristics of these systems (proportions of the different reactors, actinide contents etc.) are determined so that equilibrium exists between actinide production and consumption. In the reactors, the actinide inventory remains unchanged. Only the losses in the different cycle operations (fabrication, reprocessing) represent the ultimate wastes intended for disposal;

- since the PWR MOX and FBuR incinerator reactors cannot consume all the actinides loaded in a single passage, this obviously implies a change in the isotopic composition of the fuel during multi-recycling. To avoid drawing hasty conclusions from the results obtained in the first recyclings alone, the calculations are iterated until isotopic equilibrium in the fuel is obtained.

Isotopic equilibrium is reached after about five to six cycles, and considering the inter-cycle times (cooling, reprocessing, fabrication), this corresponds to at least 50-70 years depending on the intercycle cooling time. Nevertheless, it must be emphasised that mass equilibrium between actinide production and consumption is always achieved in the first 5 cycles and before isotopic equilibrium is reached.

The reactor system consisting exclusively of PWRs (THERM) has the following features:

- in PWRs, it is not possible to recycle the MAs by mixing them only with Pu in MOX assemblies, because this would mean prohibitive Pu contents after the second recycle;
- the recycling of MAs must be therefore dissociated from that of Pu, with some of the PWR reactors recycling Pu in MOX and others dedicated to recycling the MAs. This scenario still needs further examination;
- in HMRs, the only possibility of jointly recycling Pu and MAs is to use an enriched U support while maintaining a constant Pu+MAs content. By way of example, with a Pu+MA content of about 8.4%, the masses of recycled Pu and MAs would theoretically be stabilised after twenty recycles. With a 55% HMR in the reactors, these masses would be around 962.5 t Pu and 245 t MAs, corresponding to annual flows of 78.75 t Pu and 19.25 t MAs in a system producing 700 TWhe/year.

The characteristics of the other types of reactor system considered are as follows:

- all the PWRs and FRs have an electrical capacity of 1 450 MWe. The burn-up of PWR UO_2 is 55 GWd/tHM.
- for the MIX 1 and MIX 2 scenarios, the FR cores correspond to the CAPRA reference oxide, with initial enrichment of 45% and core residence time of 855 EFPD.
- for the FAST scenario, the cores correspond to the reference EFR with zero regeneration gain (configuration with two axial blankets of 5 cm and one ring of radial fertile assemblies), with initial enrichment of about 20% and core residence time of 1 700 EFPD.

Neptunium is recycled homogeneously and mixed with the fuel in the core. Americium is recycled heterogeneously in the form of targets placed in the first peripheral ring of the core.

Curium is assumed to be stored for a century to allow the ^{243}Cm and ^{244}Cm to decay to Pu. This Pu is then recycled with the standard Pu flux. The residual curium (essentially ^{245}Cm) is recycled with americium in the targets.

Only the losses during reprocessing operations are considered with assumptions of 0.1% for Pu and 1% for MAs.

4.3.1.3 Results

In terms of installed capacity, the proportions of the different components of the reactor systems at equilibrium are as follows (Table II.18).

Table II.18 **Recycling mode with varying reactor park composition**

	PWR UO_2	**PWR MOX**	**FBR**
MIX 1	70%	–	30%
MIX 2	70%	10%	20%
FAST	–	–	100%

The radiotoxic inventory, expressed in Sv/TWhe, going to the wastes, and their changes over time are indicated for the different scenarios, and are compared with the radiotoxic inventory resulting from a reactor system with the same capacity consisting exclusively of PWR UO_2 managed in open cycle, i.e. with all the irradiated fuel discharged in the wastes. The reduction of radiotoxic inventory in the wastes are found to be similar for all the scenarios and close to a factor of 100 (Table II.19).

Table II.19 **Waste radiotoxic inventory reduction factor due to MA recycling as a function of disposal times**

Time (years)	**100**	**1 000**	**10 000**	**100 000**	**1 000 000**
MIX 1	74	72	77	130	82
MIX 2	71	75	84	140	80
FAST	110	100	100	150	150

One important result of all the scenarios examined is that the Pu+MA inventory in the cycle can be stabilised. For each of the systems considered, the mass inventory in the reactors is given in Table II.20.

Table II.20 **Reactor inventories in different systems (kg/GWe)**

	UO_2	MIX 1	MIX 2	FAST
^{237}Np	29	73	77	33
^{238}Pu	20	170	189	154
^{239}Pu	243	762	757	3 396
^{240}Pu	111	1 025	979	2 169
^{241}Pu	75	223	269	275
^{242}Pu	44	312	376	210
Total Pu	493	2 492	2 570	6 204
^{241}Am	13	252	246	259
^{242m}Am	0.05	19	17	21
^{243}Am	10	269	298	136
^{242}Cm	1.1	8.6	8.1	8.0
^{243}Cm	0.07	0.5	0.4	0.9
^{244}Cm	4.6	57	59	47
^{245}Cm	0.3	13	13	12
Total Cm	6.0	79	81	68
Total actinides	551	3 184	3 289	6 721

At equilibrium the reactor inventories increase by a factor of 6 to 10 when recycling is performed according to the MIX1, MIX2 and FAST scenarios.

4.3.1.4 Influence of non-recycling of curium

In the same scenarios, but with all the curium sent to the wastes, the reduction in radiotoxic inventory is significantly decreased.

Table II.21 **Radiotoxic inventory reduction factor as a function of recycling mode and disposal time without Cm recycling**

Time (years)	**100**	**1 000**	**10 000**	**100 000**	**1 000 000**
MIX 1	10	12	7	96	36
MIX 2	10	11	7	94	32
FAST	21	24	14	115	57

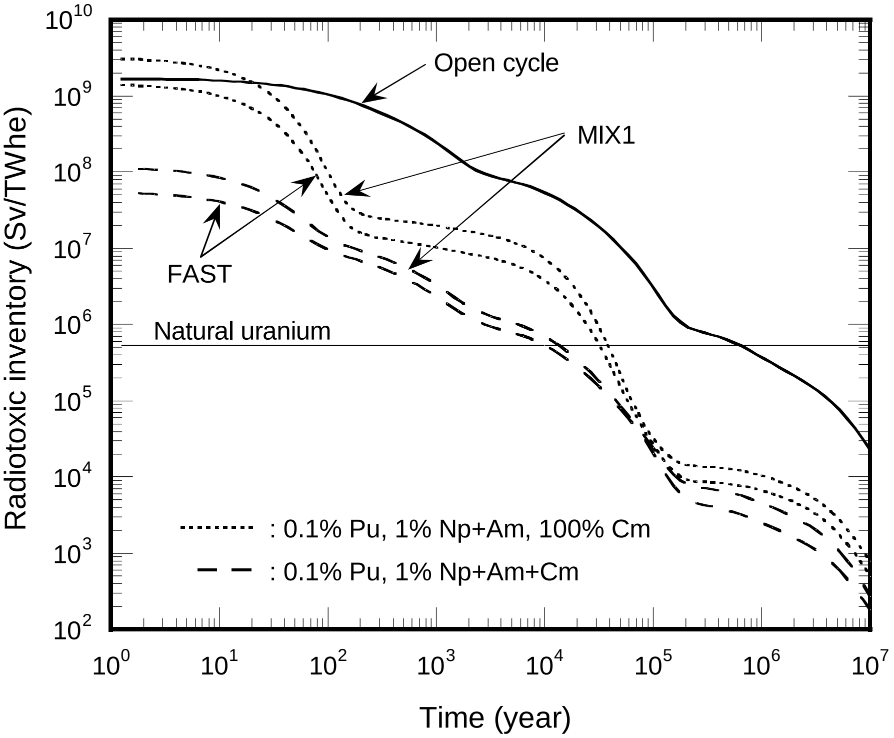
The comparison of the data of Table II.19, with the curium recycling option, to those of Table II.21 with curium discharge in the waste streams, shows that the radiotoxic inventory reduction factor is decreased from the range of 70-150 down to 7-115 depending on the time interval. This obviously has direct repercussions on the radiotoxicity levels in the conditioned high level wastes. Within the time frame of 10 000 years, curium recycling has a significant impact: the ratios between the reduction factors range from 5 to 10. Beyond 10 000 years the impact of curium recycling becomes negligible.

Figure II.25 shows the radiotoxicity evolution in Sv/TWhe as a function of time for the open cycle, the MIX1 and FAST scenarios with and without Cm removal.

For periods of time in which the radiotoxic inventory attributable to curium is high (around 10 000 years owing to the production of ^{240}Pu by α decay of ^{244}Cm), the reduction factors are similar to those resulting from a strategy of plutonium recycling only.

A realistic approach to reducing the potential radiotoxic inventory must therefore include separation/transmutation of curium as well as for the other actinides.

Figure II.25 **Evolution of radiotoxicity in wastes**



4.3.1.5 Separation performance objective

The analysis of the different scenarios examined, with assumed loss rates of 0.1% Pu and 1% for minor actinides, shows similar contributions by both to the radiotoxicity in the wastes. This is found directly in the mass balances of the wastes given in Table II.22.

Thus, any improvement in separation performance must apply to both plutonium and the minor actinides. With either alone it would achieve only moderate gains, e.g. a factor of 2 to 3 maximum in the radiotoxicity level due to a tenfold improvement in the separation factor. Thus, in the range of scenarios examined, plutonium should be separated 10 times more completely than the minor actinides in order to achieve a similar radiotoxicity reduction.

Table II.22 **Masses in wastes (kg/TWhe) ***

ISOTOPE	OPEN	MIX 1	MIX 2	FAST
^{237}Np	1.67	0.0300	0.0318	0.00806
^{238}Pu ^{239}Pu ^{240}Pu ^{241}Pu ^{242}Pu	1.12 14.1 6.44 3.44 2.54	0.00684 0.0329 0.0462 0.00903 0.0139	0.00656 0.0159 0.0372 0.00706 0.0140	0.00299 0.0857 0.0573 0.00713 0.00536
Total Pu	27.7	0.109	0.0807	0.159
^{241}Am ^{242m}Am ^{243}Am	1.09 0.00271 0.550	0.0508 6.0×10^{-4} 0.0553	0.0520 0.00285 0.0588	0.0330 0.00188 0.0158
Total Am	1.65	0.107	0.114	0.0507
^{242}Cm ^{243}Cm ^{244}Cm ^{245}Cm	2.63×10^{-5} 0.00325 0.220 0.0198	2.89×10^{-5} 0.00220 0.0557 0.00282	2.39×10^{-5} 0.00174 0.0535 0.00271	1.85×10^{-5} 0.00159 0.0236 0.00128
Total Cm	0.243	0.0608	0.0580	0.0264
Total actinides	31	0.31	0.29	0.24
Total actinides without Cm recycling	31	6.3	6.0	2.9

* 1 GWe-year = 8.76 TWh

4.3.1.6 Conclusions

Assuming industrial and economic feasibility, it would theoretically be possible to reduce substantially the output of radiotoxic heavy nuclei associated with the supply electricity from a mixed reactor park composed of LWR-UO$_2$, LWR-MOX and FRs.

This can be done by separating the heavy nuclei from the wastes during spent fuel reprocessing and recycling all of them (reprocessed U, Pu and MAs) to the reactor. Equilibrium between production and consumption of TRUs would theoretically require a period corresponding to at least 5 complete fuel cycles. Such a scenario requires the gradual build-up of a FR generating capacity of at least 20% i.e. 20 GWe, the equivalent of about 14 EFRs of 1 450 GWe each, in a 100 GWe reactor park.

The only "wastes" to be stored definitively would be the fission products and the residues due to losses in reprocessing and fuel fabrication operations. However, analyses of multi-recycling MAs show that they do not all offer the same potential for reducing the radiotoxic inventory:

- Neptunium is in fact easily separable and neptunium fuels would raise no fabrication problems in homogeneous mode. However, the irradiation of neptunium leads to the formation of Pu-238 which is detrimental to reprocessing. Moreover, no significant decrease is obtained in the radiotoxic inventory before 10^6 years. Thus there is no immediate advantage in recycling neptunium, except as part of a strategy of systematically incinerating all the MAs;

- If americium is recycled alone, only a moderate gain is achieved in comparison with plutonium multi-recycling owing to the accumulation of curium. Nevertheless, while Pu multi-recycling reduces the radiotoxic inventory of wastes by a factor of 5 to 10, this reduction would be increased to a factor of 30 by recycling americium, without curium;

- Curium must therefore ultimately be considered, if a maximum inventory reduction is intended. This could be achieved by fissioning the highly fissile ^{245}Cm. Yet, the very high activity of ^{244}Cm (half-life 18 years) makes it very difficult to handle such a target. An overall strategy could consider the separation of Cm and its interim storage for a century or so to allow the ^{244}Cm to decay to ^{240}Pu and then recycle the remaining mixture (^{240}Pu/^{245}Cm).

If only plutonium is multi-recycled, the equilibrium obtained involves the accumulation of 300 to 600 t of Pu in the cycle depending on the type of dedicated reactor. This stabilisation of the mass of Pu would be accompanied by a 2.5 to 10 fold increase in the mass of minor actinides produced. This mass would be discharged with the wastes and would commensurably reduce the anticipated gain in radiotoxicity to a factor of less than 10.

To make further headway, the MAs must be multi-recycled to a dedicated reactor, which would increase the mass of Pu and MAs in the cycle at equilibrium, but which would reduce the masses sent to the waste by a factor of nearly 100 in comparison with direct disposal.

However a residual mass of heavy nuclei would exist at equilibrium in the reactor and in the different fuel cycle plants.

This concept of equilibrium mass in a nuclear reactor system implies that, in the case of scheduled and progressive shutdown of the nuclear capability, it would theoretically be possible almost to eliminate the mass of heavy nuclei present in the cycle, by shutting down first the Pu-producing reactors, and then the Pu- and MA-consuming reactors as the inventory gradually decreases.

However, it must be understood that to implement such strategies would demand periods of several decades or even centuries.

For strategies using incinerating reactors and reactors fuelled with enriched uranium, the problem of natural uranium resources would arise within about fifty years.

A convertible reactor of the CAPRA type could perform equally well as a Pu consumer or a breeder, and would help to switch rapidly from a strategy of plutonium limitation or reduction to a strategy of plutonium use and regeneration or vice versa.

It must also be understood that physical studies of scenarios in no way imply their technical and technological feasibility, which are the subject of a major R&D support programme, including the development of an inert matrix for TRU targets, reactivity control in presence of high MA loadings.

4.3.2 Impact of separated nuclides

In the conventional fuel cycle, with U-Pu separation and vitrification of high level waste, the initial radionuclide content of the spent fuel is separated into different waste and product forms. For each of the product or waste streams industrial solutions for reuse, storage and disposal are available.

In the advanced fuel cycle, separated groups of nuclides or even individual nuclides have to be stored for a sometimes indefinite period and their intermediate conditioning form must be compatible with either transmutation or final disposal.

The MAs are the first group of elements to be considered. Storage of Np and Am in separate conditioned forms, or in a common matrix with very low solubility and potentially to be used as irradiation matrix, is a possible option which needs further investigation [78]. The volumes are low in comparison with the U-Pu fraction. Engineered storage of large quantities of these elements for long periods (25 to 100 years) will require specially designed and licensed facilities, situated preferably near the reprocessing plants.

The separate storage of Cm in order to let it decay to ^{240}Pu is a specific task which will require additional studies before its realisation. The heat and neutron emission from the Cm store and the time period involved, call for a thorough safety analysis before industrial implementation.

In the transmutation/incineration option the storage forms of Np, Am and Cm have to be compatible with neutron irradiation in reactors or hybrid facilities. In particular, the MA nuclide concentration in the target and the cladding materials are of utmost importance. If exhaustive (once-through) irradiation is performed the final product form will be very hot owing to fission products and residual higher actinides and special conditions for further intermediate storage and final disposal are necessary.

It is not obvious whether all the irradiation campaigns would be carried out in the same reactor or hybrid facility and these facilities will preferably, but not necessarily, be situated near the reprocessing plants. Transportation of hot targets would be possible in transport carriers designed to transport high burn-up spent fuel. The cladding materials are the most vulnerable components in high burn-up irradiations. Facilities for over-cladding irradiated targets have to be included in an overall transmutation/incineration strategy, in order to run very long-term irradiations or re-irradiations of discharged targets.

The waste management of separated MA fractions will have to be regulated and safeguarded as for fissile material. International control of the MA inventories must be envisaged for a long period of time.

Selected fission product concentrates, e.g. Tc, I, Cs and Zr could be conditioned in special chemical or metallurgical forms to be stored before irradiation or disposal. But many different packages will render the management of these sources much more complex than that of vitrified HLW, and perhaps more difficult to keep under control over extended periods of time.

Regionally, specialised repository structures could be developed to store the separated fractions under optimal safety conditions for these extremely long lived radionuclides. The management of individual isotopic sources in a geologic repository with guaranteed absence of contamination is the major issue to be investigated.

4.4 Risk and hazard assessment over time

While the radiotoxic inventory is a physical-biological concept intrinsically based on the laws of radioactive decay and the radiological damage due to a quantity of radioactivity incorporated in the human body, the risk and hazard concepts on the contrary rely on the extent of conditioning and packaging of waste streams, on the long-term behaviour of waste packages in geological media and on the routes which could be followed by radioactive releases on their return to the biosphere and to mankind.

- In a first phase one has to assess the direct radiological impact of additional fuel cycle operations.
- The second step is the radiological assessment of the waste types created by the AFC operations.
- Finally the hazard assessment over time is closely related to the repository design and location.

4.4.1 Pre-disposal waste management of RFC operations

The first positive impact is the expected decrease in uranium mining requirements. It may be estimated that recycling Pu in LWR-MOX reduces the uranium needs by 20%. If the MAs were also recycled a maximum benefit of 25% could be expected.

The present world-wide uranium requirements [172] are about 63 700 t natural uranium per year (in 1997) and this quantity might increase to values ranging between 62 500 and 82 800 t in 2015. The collective dose taken by about 250 000 workers world-wide is 1 300±300 man-Sv. The hypothetical reduction of uranium needs throughout the world by universal Pu recycling would decrease the mining requirements by 11 000 to 13 000 t based on the present needs, and 13 000 to 16 000 t based on future extrapolated needs. According to UNSCEAR data [173,174] the average effective dose for underground workers is 5 to 10 mSv/year depending on the type of mining[a)].

The uranium requirements can also be expressed in t/GWe-year (load factor = 0.8). In the present conditions this corresponds to 183 t/GWe-year or 26 t/TWhe. The average dose to workers expressed in amount of uranium extracted is 23 ± 3 man-mSv/t natural uranium. World-wide recycling of Pu as LWR-MOX would consequently reduce the collective dose by 20 to 25% or 260~375 man-Sv to about 1 000 man-Sv. But this is a hypothesis which cannot be taken for granted since it would imply a drastic change in the national fuel cycle and reprocessing policies of some major countries.

However, we may deduce the specific dose rate saving per GWe-year or TWhe in order to compare these “savings” with the other contributions in the fuel cycle. The uranium requirements can be reduced from 183 tHM to 138-146 t/GWe-year or 20.8-19.5 t/TWh in case of Pu and actinide recycling in LWRs.

[a)] A recent report indicated overestimation of the dose from U-mining activities by a factor of 200-400, see *Long-Term Population Dose Due to Radon (Rn-222) Released From Uranium Mill Tailings*, SENES Consultants Limited, Canada, April (1998)

Some gains can also be made in the uranium conversion and enrichment services but these dose reductions are very small and may be neglected.

In the field of uranium fuel fabrication, the mean occupational dose of the 24 000 workers world-wide is 0.45 mSv/year with maximum values of 1.7 mSv/year. The normalised collective dose is 11 man-Sv world-wide and 0.07 man-Sv/GW-year for LWR fuel, with the lowest values in the recent period (1985-1990).

MOX fuel fabrication induces mean individual doses of 7 mSv/year and collective doses of 1.5 man-Sv/GWe-year. The higher exposure from MOX fabrication results from the presence of Am-241 in standard LWR-Plutonium. Introducing ALARA principles reduced the collective radiation dose to 40 mSv/tHM MOX fuel [175].

Reprocessing is a key technology for P&T and has to be examined in more detail. The large industrial reprocessing plants have very well established statistics on the collective effective dose equivalent generated by their operation on the workers and on the environment. The modern La Hague UP2 and UP3 plants have been able, due to ALARA practices, to reduce the dose to their 4 740 workers to 0.26 mSv/year leading to a collective occupational dose of about 1.23 man-Sv/year. From 1991 the collective occupational dose has been further reduced to less than 0.1 man-Sv/GWe-year.

The increase in reprocessing capacity (UP2 and UP3) to 1 600 tHM/year corresponding to 80 GWe-year will ask for additional separation and purification units to keep the environmental impact at the current levels.

The collective dose to the Sellafield workers is about 20 man-Sv/year but substantial improvements are to be expected with the operation of the THORP plant.

Reactor operations do not influence the overall risk analysis since it may be expected that in the future, new reactor types will replace older ones with the same or improved radiological impact, but that, at least in the OECD Member countries, the reactor park will not grow substantially. The radiological impact of PWRs throughout the world in the period 1985-1989 was about 4.3 man-Sv/GWe-year which is at about the same level as uranium mining. However, it may be expected that important dose reductions will occur in the future.

As a conclusion, one might suggest that compared with the RFC, the AFC with P&T would moderately increase the collective dose to the workers in the fuel cycle, and particularly to those in fuel and target fabrication. However, appropriate measures must be taken to reinforce shielding, especially against neutrons, throughout the entire recycling facilities, and this will significantly increase the overall investment cost.

Since the only short-term radiological benefit of the RFC lies in a decrease in uranium mining and milling requirements (due to the substitution of recycled plutonium for ^{235}U), the additional recycling of MAs will not significantly modify this picture. However, an appropriate management strategy for ^{244}Cm is a prerequisite for such a conclusion. When the dose to man in the different fuel cycle options is expressed as man-Sv/TWh, or if a double-strata approach is assumed to be industrially realised, small changes in dose to workers are to be expected.

4.4.2 Radiological impact of waste discharges

As with doses to the workers, the main environmental impact from the nuclear fuel cycle is due equally to uranium mining and milling, and to the operation of the nuclear power plants. The enrichment and fuel fabrication plants have a minimal impact on the environment.

The nuclear reprocessing operations do not have world-wide a very large impact because only a fraction of the spent fuel is being reprocessed for Pu recycling, furthermore many technical improvements have been introduced to decrease the discharges into the ocean, particularly those of ^{90}Sr, ^{99}Tc, ^{137}Cs and the actinides.

Assessing the influence of P&T on the waste discharges needs more specific data on the chemical methods used for MA separation. But a global assessment can be made through the discharges from the present reprocessing plants.

The UNSCEAR 1993 [174] and the NEA 1993 [175] report provide overview and comparative data for the period 1980-1985 and for the period 1985-1990, respectively. More recent data about releases are available in the proceedings of the RECOD'94 conference [176]. Additional efforts have been made in the 1985-1995 period to reduce the radioactive releases, especially α emitting nuclides.

The data are summarised in Table II.23 through a compilation and intercomparison of release rates from different international sources, providing a coherent overview of their relative impact.

Table II.23 **Radionuclides discharged from reprocessing plants (compiled from UNSCEAR, NEA and RECOD'94 [173-176])**

Airborne effluents

	La Hague			**Sellafield**		
Year	1980-85	1986-91	1990-95	1980-85	1986-88	1990-95
Throughput (GWe-year)	35.7	83.2	143	16.3	8	20.35
Tritium (TBq/GW-year)	0.91	1.44	1.84	120	54	173.4
^{14}C (TBq/GW-year)	–	–	–	3.5	2.2	1.05
^{85}Kr (TBq/GW-year)	11 500	11 000	5 476	14 000	15 800	14 779
^{129}I (TBq/GW-year)	0.0049	0.0198	8.04×10^{-4}	0.0037	0.009	0.006

a) Excludes tritium.
b) Cs and Sr.

Table II.23 **Radionuclides discharged from reprocessing plants (compiled from UNSCEAR, NEA and RECOD'94** [173-176]) **(Continued)**

Aqueous discharge

	La Hague			**Sellafield**			**Tokai**
Year	1980-85	1986-91	1990-95	1980-85	1986-91	1990-95	–
Throughput (GWe-year)	35.7	83.2	143	16.3	8	20.35	–
Tritium (TBq/GW-year)	186	234	242	579	656	557.7	240
Total beta[a)] (TBq/GW-year)	174	43.5	4.92	969	96	29.6	10^{-5}
Fission product[b)] (TBq/GW-year)	20.3	3.5	–	784	28	9.6	10^{-4}
Total Alpha (TBq/GW-year)	0.1	0.027	6.3×10^{-3}	8	2.9	0.5	10^{-5}

a) Excludes tritium.
b) Cs and Sr.

The collective dose to the environment has been significantly reduced by installing separation plants for ^{137}Cs and ^{90}Sr and by improving the α decontamination factor in the effluents.

The impact of P&T on these release figures would in a first period only affect the α emitters (Np, Am and Cm) which would be separated from HLLW. The α-waste discharge due to partitioning operations [177] is assumed to increase proportionally to the inventory of MAs in HLLW and represents the Pu+MA process losses in the waste streams. The MA/Pu ratio depends on the burn-up of the spent fuel, the cooling time and the degree of separation [178]. If the same separation efficiency were obtained for the MAs as for Pu (99.9%), the increase in α discharge rate, from an advanced reprocessing operation of LWR fuel at 47 GWd/tHM, would amount to a factor of 2, essentially due to ^{241}Am and ^{244}Cm after 10 years cooling time. From this preliminary analysis may be deduced that the environmental impact of P&T can be limited if the appropriate separation plants are installed on the same sites as the large plants for LWR fuel reprocessing. In order to compare the different contributions of each of the fuel cycle operations, the normalised collective effective dose equivalent commitments, for local and regional populations, are given in man-Sv/GW-year.

From these UNSCEAR 1993 data [174] it appears that uranium mining and milling and LWR reactor operations are mostly responsible for the local and regional collective doses. Reprocessing and recycling of U, Pu lower the local and regional collective doses from U mining and milling, but contribute in their turn to a slight increase in the marine contamination. P&T operations on MAs are expected to further slightly decrease the uranium needs but not to influence this picture drastically.

A recent study carried out by Cogéma under sponsorship of the European Commission [180] and presented at GLOBAL'97 [181] provides new data on dose rates to the public resulting from the fuel cycle operations. The data are expressed in TWhe for the entire fuel cycle including the waste disposal and transportation doses. For the OTC the total fuel cycle dose amounts to

0.191 man-Sv/TWhe and the RFC with MOX fuel recycle 0.201 man-Sv/TWhe which corresponds to about 1.33 to 1.41 man-Sv/GWe-year. This recent set of data is in absolute value roughly 40% lower than the dose rate derived from the earlier UNSCEAR data. This decrease results from the reduced discharges into the biosphere by the major fuel cycle operations. In both studies, the relative contributions of uranium mining and milling on the one hand and the reactor operations on the other hand are equally the main contributors to the environmental dose rate. Recent studies by the Uranium Institute [179] have confirmed the overestimation made in the earlier UNSCEAR data for uranium mining and milling. The new study takes into account a more realistic set of data resulting from on site measurements of the radon emission rate, the surface of tailings, the population density and the local air dispersion factors. The estimated long-term population dose is according to this study 0.96 man-Sv/GWe-year.

In the OTC scenario, uranium mining and milling constitutes 90% of the total regional dose. In the RFC, the impact of reprocessing on the regional dose amounts to 10% in the UNSCEAR study and 28% in the Cogéma study. If the AFC should become an industrial reality in those countries having a reprocessing infrastructure, the contribution of the MAs and FPs separation and conditioning operations would slightly increase the overall radiological impact on the environment but not modify the overall picture as shown in Table II.24.

Table II.24 **Local and regional collective doses to the public** [174,179]

Operation		Dose (man-Sv/GWe-year)
Uranium mining and milling		0.96-1.15
Uranium fuel fabrication		0.003
Reactor operation	atmospheric releases	1.3
	liquid discharges	0.043
Reprocessing	atmospheric releases	0.05
	marine releases	0.2
Total		2.55-2.74

4.4.3 Impact of P&T operations on waste management of AFC [78]

Separation of ^{237}Np is technically feasible and could be implemented on industrial scale as soon as a waste management policy for this long-lived nuclide required a separate treatment. However the long-term storage of that element in a separate package has to be co-ordinated with the management of ^{241}Am which decays to ^{237}Np. Conditioning the mixture of Am and Np together in one matrix is therefore preferable to separate treatment of both radionuclides.

The most probable scenario for Am consists of a group separation with some rare earth contamination (1 to 10%) followed by conditioning in a thermodynamically stable matrix (SYNROC, Zr-silicate, ceramic matrix, etc.) which can be stored in engineered facilities and retrievably disposed in a repository structure. The mixture of Am and Np leads in the long term (after 4 000 years) to a single Np concentrate.

The Cm issue is more complex since its separation from Am is a very difficult task. Assuming that Cm can be effectively separated from Am and the REs, its engineered storage is not an easy task because of the high heat dissipation (120 W/g pure ^{244}Cm), important neutron emission (10^7 neutron/s per *g* of pure ^{244}Cm) and its decay to ^{240}Pu. The matrix in which it would be embedded must have a high heat transfer coefficient and be crystallographically compatible with the Pu daughter.

Alternatively, it might be preferable to develop a partitioning technology in which the three MAs (Np, Am and Cm) were brought together in one matrix, e.g. a SYNROC type of mineral in which Cm would be the limiting radionuclide in terms of specific activity. The matrix could be stored in a retrievable form to use it at a later stage as nuclear technology progresses. If this matrix could also be used as a radiation target for eventual transmutation later generations could recover the matrix and "incinerate" it before final disposal as fission products.

4.4.4 Global fuel cycle situation for waste management decisions [182,183]

A NPP park of 100 GWe-year produces annually 700 TWh electricity and generates as derived from the values in Figure 1.3 an equilibrium mixture, depending on the burn-up, of 1 600 to 2 100 tHM spent fuel types which are partially reprocessed and partially stored. Storage is an essential item in the overall nuclear strategy and needs further consideration of the long-term implications. Recycling of Pu from LWR-UO_2 reduces the fresh uranium requirements to 14 000 tHM/year instead of 17 500 tHM/year. The spent fuel is in the given scenario reprocessed in the available facilities (1 600 tHM/year in La Hague, 900 tHM/year in Sellafield, 90 tHM/year in Tokai, and in the future, 800 tHM/year in Rokkasho-mura). The European and Japanese spent fuel arisings can be reprocessed and the resulting HLLW wastes can be stored till vitrification.

The other countries, except Russia and India, do not have the necessary facilities to reprocess the spent LWR-UO_2 fuel. In terms of generating capacity, the countries dedicated to reprocessing and those to direct disposal are equal in power output. If the P&T strategy intends to improve the radiotoxic inventory situation on a world-wide scale, additional industrial reprocessing units for a total of about 3 500 tHM/year should be built and operated. This is a strategic decision, not very probable in the present economic and political climate. Long-term and possibly retrievable storage of large spent fuel masses will be the rule rather than the exception.

The consequences are that with a continuous nuclear power production of 320 GWe, additional large repositories will be necessary. If the maximum content of a repository is 70 000 tHM spent fuel like that of Yucca Mountain in USA, then without any reprocessing, every 10 years a new repository of that size or equivalent will have to be constructed. With partial reprocessing (3 900 tHM/year), the interval is increased from 10 to 25 years. However, as long as there are no regional or international conventions on waste transfer to other countries, the optimum size of each repository will depend on the size of the NPP park in each individual country and vary with time and location.

The *impact of reprocessing* on the strategic decisions in the fuel cycle is multiple:

- without reprocessing, a total amount of 50 to 60 t of plutonium and 7 to 8 t of MAs have to be disposed of annually world-wide;
- reprocessing of spent fuel is from a long-term radiotoxic point of view a preferable strategy since it decreases the amount of Pu in HLW;

- conventional reprocessing does not remove MAs from the HLLW, and Np, Am and Cm constitute the main long-term hazard;
- the RFC implies the construction and operation of Pu handling facilities for LWR-MOX and in the future for FR-fuel types;
- the reprocessing of LWR-MOX fuel is an issue which has not received a full industrial answer and which would have to be investigated in a long-term waste management programme;
- the long-term hazard of the MAs is, beyond 40 000 years, determined by ^{237}Np;
- the construction of a repository for MA-free HLW glass, during the initial phase, is of standard design and construction.

The impact of P&T is an improvement of the long-term hazard but it requires additional actinide handling facilities and does not eliminate the necessity of geological disposal:

- without reprocessing, P&T cannot be implemented;
- partitioning of MAs from HLLW is possibly a first step in the gradual decrease of the radiotoxic inventory of vitrified HLW;
- long-term storage of partitioned MAs waste fractions and long-lived fission products will be necessary since special reactors have to be developed for transmutation;
- partially “incinerated” or “deactivated” separated actinide or fission product targets will need special preparation and conditioning before disposal;
- the risk of contaminating the geosphere will be decreased if the conditioning of the toxic radionuclides is improved (e.g. by using ceramic matrices or improved glass compositions for the separated MAs);
- the fraction of radionuclides involved in fuel cycle and waste management activities will shift from mainly disposed materials to mostly stored inventories as shown in Table II.25;
- compared to these reactor and facility inventories, the tonnages of waste discharged annually in a reactor park of 100 GWe-0.8 year (= 700 TWhe) are given in Table II.26.

Table II.25 **Mass of transuranic elements in reactor park (tHM/100 GWe)** [184]

Elements	**UO_2**	**MIX1**	**MIX2**	**FAST**
Np	2.9	7.3	7.7	3.3
Pu	52.2	249.2	257	620
Am	2.3	54	56.1	41.6
Cm	0.6	7.9	8.1	6.8
Total	58	318.4	328.9	671.7

Table II.26 **Annual discharge of TRU wastes (tHM/100 GWe-0.8 year)** [184]

Elements	UO_2	MIX1	MIX2	FAST
Np	1.155	0.021	0.022	0.0056
Pu	22.78	0.076	0.056	0.111
Am	1.152	0.075	0.0798	0.0354
Cm	0.126	0.042	0.040	0.018
Total/year	25.21	0.214	0.198	0.17
Total/30 year	756	6.42	5.94	5.1

On comparing the OTC data in terms of inventory point of view, it is obvious that the amount of spent fuel as "waste" represents world-wide 13 years of operation of a 70 000 tHM repository for 320 GWe park. In the MIX1, MIX2 and FAST options, the equilibrium TRU inventory of the reactor and nuclear facilities amounts to 98% of the total amount of actinides involved in the fuel cycle, and the waste discharged over 30 years becomes very small (<1%). However, this strategic option implies the replacement of 20% or more of the present LWRs by FRs, and a complete recycle of recovered plutonium.

Apart from the construction of dedicated transmutation and incineration facilities, this option also implies the construction of specialised fuel fabrication facilities capable of processing 1 490 tHM UO_2 fuel, 236 tHM LWR-MOX fuel, and 20 FRs with an actinide throughput of about 100 tHM per year.

The options for continuous nuclear energy production during several centuries are of course debatable but it takes several decades to evolve from a pure LWR-UO_2 park to a mixed NPP park with LWR-MOX and FRs.

The nuclear fuel cycle during such evolution undergoes gradual changes which were taken into account in the assessment of the nuclide quantities occurring annually. However, this option like any scenario has an end when other energy production systems will one day make nuclear energy in its present and foreseeable forms redundant or obsolete. The question then arises what will be the ultimate nuclear fuel inventory and how will this be managed.

One can assume that the conventional LWR-UO_2 reactors which require a regular input of fresh uranium will be the first to be replaced by new energy-producing facilities. This is already the case with LWR-(30%)MOX reactors. In a shrinking nuclear park, spent nuclear fuel and waste materials must be appropriately managed. The LWR-(100%)MOX reactors will be in competition with the FRs to incinerate Pu from the LWR park. It is very probable that the quality of produced plutonium (due to the increase of ^{238}Pu, ^{240}Pu and ^{242}Pu) in this composite nuclear production park will gradually become unsuitable for thermal reactors. At this point in evolution, FRs, FBuRs and the ADS are the only alternatives to burn the residual Pu and MAs.

Multiple recycling of FR spent fuel is a very difficult issue because of the highly radioactive mixture (^{238}Pu, ^{241}Am and ^{244}Cm) which has to be processed [185]. At this stage of development, pyrochemical reprocessing will probably become indispensable to recycle TRU materials.

The maximum inventory of reactor cores to be disposed of is the equilibrium MIX2 case and the minimum NPP core capacity is the FAST case. The total amounts of TRU to be managed are given in Table II.25. The number of reactor cores is the same in both cases but the FAST case has an overall TRU inventory which is much higher than the MIX2 case (672 tHM compared with 329 tHM).

4.4.5 Evolution of long-term radiotoxic inventory [77,170,186]

Unless a comprehensive TRU incineration programme is set up e.g. by accelerator driven transmutation, the residual cores from LWR-MOX, FBuR-MOX or FR-metal will have to be disposed of. The composition of residual core fuel is different from one type of reactor to the other. The main parameter to assess the radiotoxicity is the α-radioactivity.

Table II.27 **Alpha activity of TRU nuclides in spent fuel types in TBq/tHM** [185, 187]

	UO_2, 40 GWd/tHM	**MOX, 40 GWd/tHM**	**FR-MOX, 150 GWd/tHM**
^{238}Pu	128.76	588.3	6 138.3
^{237}Np	0.0148	0.00592	0.03996
^{241}Am	55.5	324.12	2 223.7
^{244}Cm	99.9	1 276.5	14 282
Total α	323.75	2 382.8	23 791

Cooling time: 7 years

The most striking feature of this table is that Np, in terms of radiotoxicity, is negligible compared to the other TRUs. However, its very long half time (2×10^6 years) is of the order of magnitude of geological phenomena. As time goes on the radiological significance of Np increases but remains in absolute value very low with respect to the initial radiotoxic inventory.

Unless very long engineered storage times are envisaged the difficulty of disposing of spent fuel increases progressively from LWR-UO_2 to LWR-MOX and finally to FR-fuel. The major problem to be solved is the decay-heat dissipation which increases proportionally with burn-up and TRU content.

The LWR-UO_2 fuel irradiated to 40 GWd/tHM has a thermal output of 1.43 kW/tHM 10 years after discharge and this decay heat, mainly due to fission products, drops to about 100 W/tHM after 100 years. The decay-heat from TRUs drops from 50 W/tHM after 100 years to 0.75 W/tHM at 10 000 years. A large NPP park of 100 GWe produces between 1 600 and 2 100 tHM/year which constitutes a diffuse heat source declining from about 3 000 kW after 10 years to 25 kW after 1 000 years. A large repository of 70 000 tHM filled in 30 years will have to dissipate a mean thermal output of 380 (FP)+69 (TRU) W/tHM or 31 400 kW. In order not to disturb the natural hydrogeological and geomechanic properties of the host rock, long cooling times in surface engineered structures or forced underground cooling are to be investigated and assessed.

At equilibrium, a LWR-UO_2 NPP park of 100 GWe producing 700 TWh/year discharges annually 1 760 tHM LWR-UO_2 and 440 tHM LWR-MOX. Recycling plutonium from LWR-MOX in LWRs is not useful because of the degraded Pu isotopic composition in the fuel. The LWR-MOX will have to be stored in engineered facilities till disposal. The disposal of such fuel on an industrial scale will have to accommodate an 8.6-fold increase in radiotoxic inventory with associated heat and criticality problems. The decay heat amounts to 2.22 (FPs)+1.17 (TRU) = 3.39 kW/tHM which is twice as much as for LWR-UO_2. The TRU content of spent LWR-MOX is 42.3 kg/tHM compared with

11.4 kg/tHM for UO_2 fuel (factor 3.7). Very long cooling times (more than 50 years) may be needed before disposal operations could start. The spent MOX fuel contains 16.7 kg fissile isotopes per tHM after the decay of ^{241}Pu.

Delayed reprocessing of spent LWR-MOX fuel is probably the best but perhaps not the cheapest solution. Transfer of the TRU content to a FR for multiple recycling is one of the alternatives to disposal. At this stage in nuclear development partitioning could play an essential role in the reduction of the radiotoxic inventory of HLW glass before disposal.

Ultimately, managing the spent fuel discharged from the residual core of FRs is undoubtedly the most difficult issue in the back-end of the fuel cycle [185]. The fuel, containing between 30 and 40% TRUs, would have been irradiated to 120 – 150 GWd/tHM. The decay heat ranges from 20 to 25 kW/tHM. If a NPP park had been gradually reduced during several decades, the last fraction which remained would be of the FR type and in the hypothesis of this report amount to 30% of the initial NPP park, i.e. about 30 GWe. The "last residual" cores would contain about 6 tHM TRUs each, bringing the total for disposal to 180 tHM very hot fuel. Three alternatives are possible: to "dilute" the spent fuel assemblies in a large conventional repository with previously cooled HLW glass or spent UO_2 fuel, to keep the hot spent fuel in permanent bunkers for hundreds of years, or to transmute this TRU mass in a accelerator driven transmutation system.

The radiotoxicity of this type of fuel is very high (74 times that of UO_2 fuel) owing mainly to ^{244}Cm, ^{241}Am and ^{238}Pu. The radiotoxic inventory will decrease with the half-life of ^{244}Cm (18 years) and then with the half life of ^{238}Pu (87.7 years). The total α-activity per tHM would decrease to 5 217 TBq alpha after 100 years and to 1 110 TBq α after 500 years. The long-term radiotoxic inventory of this kind of waste material is very high and needs special conditioning methods to allow for safe decay in geological repositories.

The last alternative is to submit this ultimate residue from the nuclear fission era to transmutation reactions in ADSs.

In any event, and despite whatever nuclear developments may be expected in the future, there is no alternative to disposal. For very concentrated TRU waste, P&T options should be kept open in order to allow for further technological progress which would reduce as much as possible the radiotoxic inventory. However, the development of repository concepts and structures adapted to the disposal of hot TRU waste (i.e. host rocks with capabilities to safely confine concentrated nuclear heat sources such as spent fuel, HLW, or isotopic targets) remains a priority in the development of the nuclear fuel cycle.

4.4.6 Role and impact of cut-off period

The lower cut-off half-life is the minimum below which there is no sense in transmutation to nuclides with shorter half-lives. Most of the fission products have half lives ranging from days to one or two years. It is obvious that there is no benefit in nuclear processes to influence their radioactive properties. There are a few medium-term exceptions for which the half lives are long compared with the life-span of a technology, notably ^{137}Cs ($t_{1/2}$ = 30 years) and ^{90}Sr ($t_{1/2}$ = 28 years), their disappearance by natural decay takes about 300 years. Partitioning of ^{137}Cs and ^{90}Sr received for several decades very much R&D attention because of its impact on waste management. Removal of these nuclides from nuclear streams has a direct impact on treatment, conditioning and disposal, and should therefore be pursued. Since ^{137}Cs and ^{90}Sr are the principal sources of decay heat in vitrified HLW, separation from HLLW merits further attention.

However transmutation cross-sections of ^{137}Cs and ^{90}Sr in LWR and FR neutron spectra are so small, and accelerator driven transmutation systems without accompanying fission reactions are so expensive, that it is doubtful whether R&D efforts in transmutation of these nuclides are justified. The development of specific nuclear reactions by for example "Adiabatic Resonance Crossing" might be a new route which ought to be explored.

Other fission products with half-lives shorter or equal to about 30 years need not be considered in a comprehensive P&T strategy.

For TRU nuclides the situation is different, since most of the "heavy metals" are linked to each other in the radioactive series (4n, 4n+1, 4n+2, 4n+3). Important nuclides are ^{241}Pu, ^{243}Cm, and ^{244}Cm which are mother isotopes respectively of ^{241}Am, ^{239}Pu and ^{240}Pu, and these are very important contributors to the total radiotoxic inventory. Despite their relatively short half-lives, these nuclides may not be disregarded in a P&T scenario. Incineration or transmutation of these nuclides can greatly modify the long-term radiotoxic inventory.

At the other extreme of the time scale lie the very long-lived fission products (^{135}Cs, ^{99}Tc, ^{129}I), plus ^{237}Np and the natural actinides (U series) with extremely long half-lives. The partitioning of the long-lived fission products is difficult, while some are accompanied by other isotopes of the same elements, and isotopic separation of fission products cannot presently be considered.

In the case of ^{135}Cs the presence of natural ^{133}Cs might interfere with transmutation-depletion reactions. A similar interference from natural ^{127}I should be investigated in case of ^{129}I transmutation. Transmutation of ^{99}Tc can proceed without interferences since it is the only long-lived isotope without any natural equivalent.

Most TRUs have long half-lives which are significant compared with the span of a human life, a civilisation or even a geological period. In principle every P&T action which might decrease the long-term impact of man-made actinides is beneficial, but there are limitations beyond the grasp of mankind.

Three time periods can be considered as cut-off limits for further hazard assessment:

- 10 000 years, as administrative limit for a spent fuel repository (USA);
- 100 000 years as the time when uranium daughter products, ^{226}Ra and others, start to dominate the radiotoxicity of the spent fuel;
- 1 000 000 years; the period significant in the evolution of the earth crust.

Ten thousand years is perhaps short from both scientific and technical points of view, but one million years is definitely beyond any anthropological horizon. A cut-off period of 100 000 years seems a suitable "logarithmic" compromise for the assessment of radiological effects.

Except for ^{237}Np, there are no significant TRU inventories which will survive the million-year term, and beyond that period the natural radioactivity of uranium and its decay products become the dominant radiotoxic inventory [188]. Not only natural uranium but also depleted and reprocessed uranium need to be considered when comparing the long-term radiotoxicity of the different man-made TRUs.

The choice of the long-term cut-off time strongly influences the collective dose [189] which would result from waste discharges and from waste disposal. But taking into account the uncertainty in the population growth, its distribution and its localisation, it is difficult to draw final conclusions on the validity of this approach for assessing waste management scenarios.

4.5 Waste management concepts

4.5.1 OTC concept and direct disposal

4.5.1.1 The Spanish repository concept [186]

Spanish policy for management of spent fuel, stated in the General Radioactive Wastes Plan, foresees direct disposal in deep geological formations after an adequate period of interim storage. Currently, three host rock options (clay, granite and salt) are being considered. The Spanish nuclear park consists of nine LWRs with an overall installed capacity of 7.1 GWe. On the basis of a 40 year lifetime, Spain will produce about 20 000 spent fuel assemblies (11 600 of PWR and 8 400 of BWR type) or about 7 000 t of uranium.

The repository concept envisages final disposal of spent fuel and other high level wastes in unshielded canisters, placed centrally along a system of horizontal disposal drifts and surrounded by a suitable backfilling material.

A carbon steel cylindrical disposal canister 4.54 m long, 0.90 m in diameter with a wall thickness of 10 cm, is currently being considered. It could be loaded with either four intact PWR or 12 BWR spent fuel assemblies or three vitrified waste canisters, i.e. a total weight of about 15 t. Remaining open space could be filled with glass beads. The disposal canister will be designed to provide substantially complete containment over a period of at least 1 000 years. A total of 3 600 canisters will be needed. For handling and transport, the disposal canister will be supplemented with a shielding overpack.

The surface facilities, see Figure II.26, have been designed to receive unconditioned spent fuel, providing the capacity to handle and encapsulate the expected waste production over a period of 30 years. An area of 480 000 m^2 is required (including 200 000 m^2 for the barren rock dump). Four openings connect the surface facilities with the underground repository and ensure independent ventilation systems for the waste emplacement and construction areas.

The disposal concept contemplates a mined-type repository excavated at a depth of 250 m in clay, 500 m in granite, and 600 m in salt. As shown in Figure II.27, the repository layout consists of an array of full-face drilled drifts for spent fuel disposal and a completely separate repository area (consisting of drifts in clay, a silo in granite, and caverns in salt) for ILW disposal. The disposal canisters will be emplaced horizontally in circular disposal drifts with a diameter of 2.40 m. A layer about 0.75 m thick of an appropriate material (bentonite for clay and granite options, and salt briquettes for the salt option) will act as a buffer element, completely surrounding the canister.

Figure II.26 **Surface facilities. General layout**

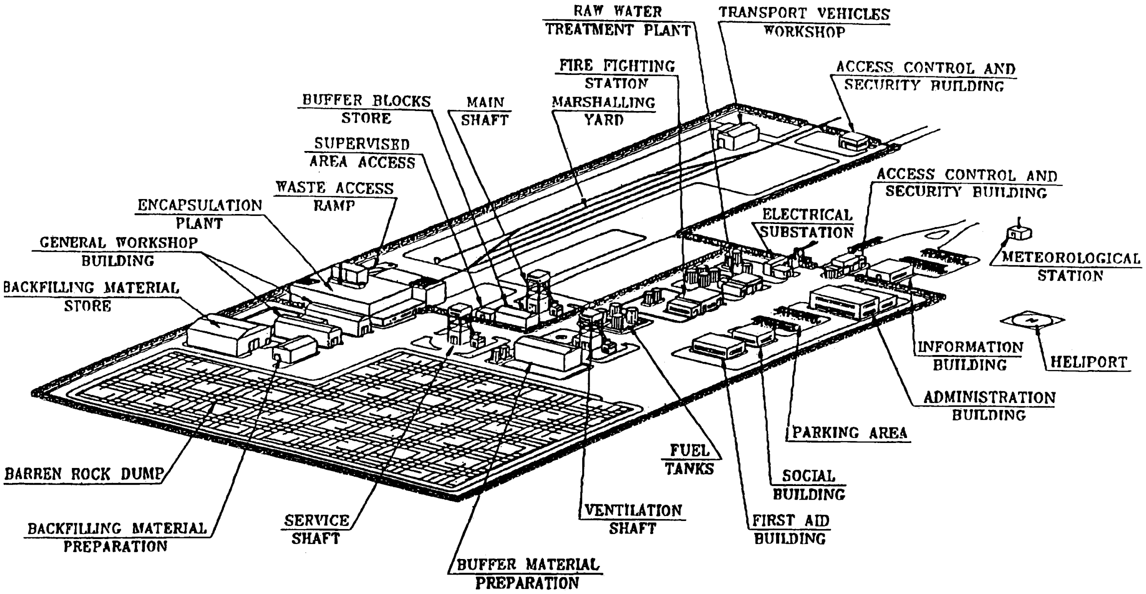
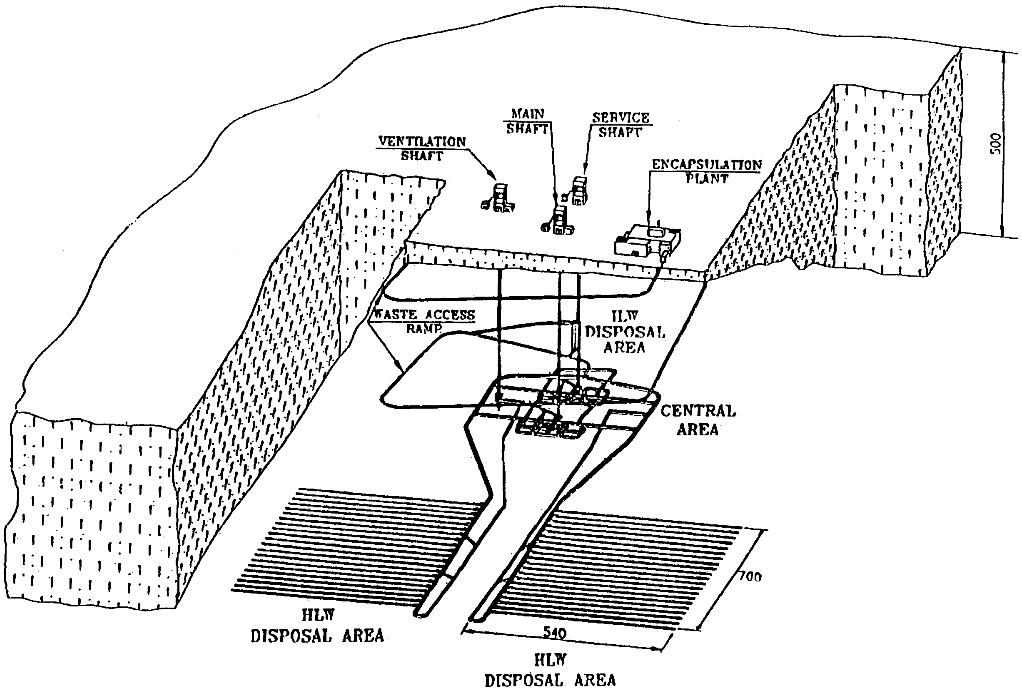


Figure II.27 **Underground facilities. General layout**



The repository area needed is 2.58 km^2 for clay, 4.1 km^2 for granite and 1.2 km^2 for salt, and the total excavated rock volume amounts to about 765 000 m^3 for clay, 569 000 m^3 for granite and 579 000 m^3 for salt.

The disposal geometry to be selected for a specific repository site must be compatible with the different thermal requirements imposed by the specific medium. The thermal analysis performed recommended a spacing between emplacement drifts of 50 to 100 m for clay, 35 m for granite and 23 m for salt. The length of the waste emplacement drift will be limited to 500 m for practical reasons, with a cross-section of about 5 m^2 and capacity for up to 87 disposal canisters. For adequate buffering, the space between canisters has been fixed at 1 m for granite and salt options, and 2.5 m for a clay option.

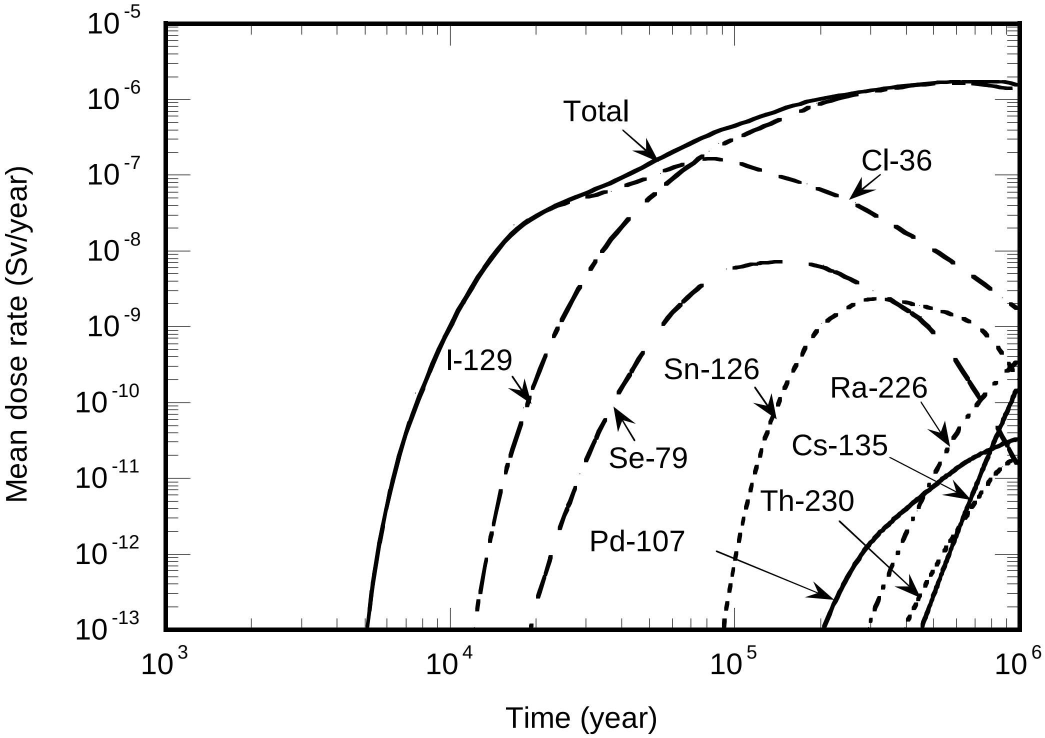
Once the emplacement activities are finished, all open space inside the repository mine must be backfilled and sealed. Different backfilling materials will be used (in-situ compacted reconstituted clay for the clay option, in-situ compacted mixture of bentonite and sand for the granite option, and in-situ compacted crushed salt for the salt option). Filled regions will be isolated from the rest of the emplacement area by means of dedicated seals. The repository area will be isolated by seals in the access shafts.

A probabilistic performance assessment for a generic site in a granitic host rock formation has been completed. It has permitted evaluation of the relative importance and performance of the various components of the repository total system as well as a sensitivity analysis of the various parameters.

A normal evolution scenario, with appropriate performance ascribed to the designed engineering barriers, has been considered as reference scenario. A hydrogeological regime based on present-day conditions and a reference biosphere has been assumed.

Figure II.28 shows the mean dose rates obtained for the normal evolution scenario averaged over 100 simulation runs. A peak dose of 2.36 *m*Sv/year, well below the regulatory limit of 100 μSv/year, is reached 600 000 years after waste disposal, with ^{129}I responsible for 99% of it.

Figure II.28 **Evolution of mean dose rates in the case of disposal of 40 GWd/tHM UOX spent fuel**



Doses are controlled by ^{36}Cl up to 70 000 years and by ^{129}I afterwards. Both radionuclides have high solubility values and are weakly sorbed in the bentonitic buffer and in the granitic host rock. Since chlorine is less sorbed than iodine, the release of ^{36}Cl to the biosphere begins earlier. ^{79}Se and ^{126}Sn have low solubility and are moderately sorbed onto the buffer material and the host rock.

Releases to the biosphere of strongly sorbed radionuclides, such as ^{107}Pd, ^{135}Cs and actinides and their daughters begin to be relevant after several hundred thousands of years.

4.5.1.2 The Swedish repository concept [190]

The plans for a deep repository in Sweden are as follows: the safety of a deep repository for spent nuclear fuel is achieved by the application of three principles:

- level 1 – Isolation
  - Isolation enables the radionuclides to decay without coming into contact with man and his environment.
- level 2 – Retardation and retention
  - If the isolation is broken, the quantity of radionuclides that can be leached and reach the biosphere is limited by:
    - very slow dissolution of the spent fuel;
    - sorption and very slow transport of radionuclides in the near field – engineered and natural barriers;
    - sorption and slow transport of radionuclides in the bedrock.
- level 3 – Recipient conditions
  - The transport pathways along which any released radionuclides can reach man are controlled to a great extent by the conditions where the deep groundwater first reaches the biosphere (dilution, water use, land use and other exploitation of natural resources). A favourable recipient means that these conditions limit the radiation dose to man and the environment. The recipient and the transport pathways are, however, influenced by natural changes in the biosphere.

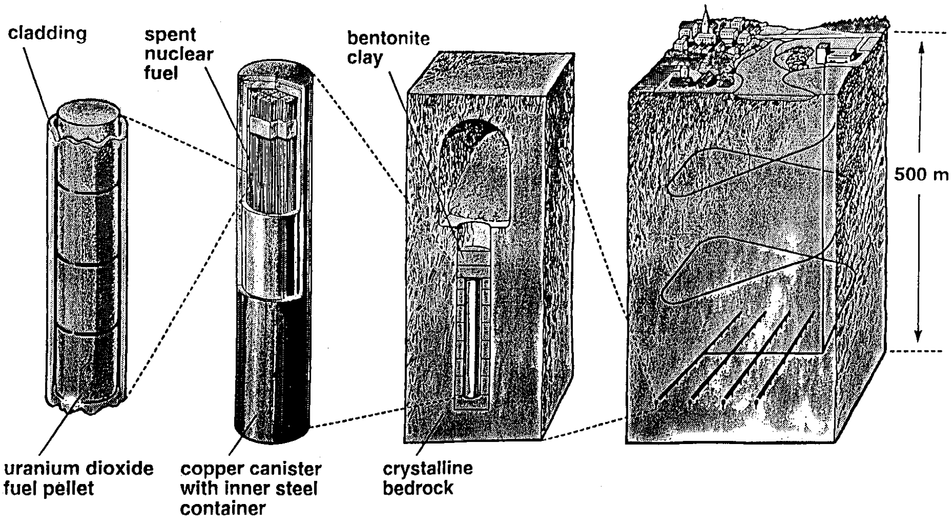
The safety functions at levels 1 and 2 are respectively the most important and the next-most important. They are achieved by means of requirements on the properties and performance of both engineered and natural barriers and on the design of the deep repository. Within existing constraints, a good safety function at level 3 is also sought through suitable placement and configuration of the deep repository.

Deep repository

The isolation of the spent nuclear fuel from the biosphere is achieved by encapsulating the fuel in a canister with good mechanical strength and very durable resistance to corrosion. The conceptual design adopted is a copper canister with a steel insert. The copper provides a very good corrosion resistance in the geochemical environment foreseen in a deep repository in Sweden. The steel insert provides the mechanical protection needed. Each canister contains about 2 t of spent fuel. The canisters are placed in deposition holes drilled below the floors of tunnels at about 500 m depth in the crystalline, granitic bedrock (Figure II.29). Each canister is surrounded by blocks of compressed bentonite. When the bentonite absorbs water from the surrounding bedrock it will exert an intense swelling pressure and completely fill all void space in the near vicinity of the canister. The clay barrier will contribute to the isolation by preventing or delaying access to the canister by dissolved corrosive species that may exist in minor amounts in the ground water. The clay will also provide some mechanical protection for the canister. The tunnels will eventually be backfilled by some material like a mixture of crushed rock and bentonite.

For a repository to dispose of all spent fuel from the present Swedish programme (about 8 000 t or 4 000 canisters), the underground facilities will need some 30~40 km of tunnels and cover an area of about 1 km^2.

Figure II.29 **Swedish repository concept**

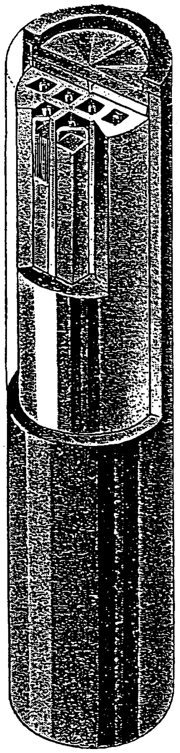


Encapsulation of spent nuclear fuel

A necessary facility is a plant for encapsulating the spent nuclear fuel. The plant would take fuel assemblies from interim storage and transfer them to canisters made of copper with a steel insert, change the atmosphere to inert gas, put lids on the canister and seal the lids by electron beam welding. The quality of the filled and sealed canisters would be checked by non-destructive examination (NDE) methods – ultrasonic and radiographic – before shipping to the repository.

Each canister can hold 12 BWR fuel assemblies or 4 PWR assemblies. The copper thickness will be about 50 mm and the steel thickness as a minimum about 50 mm (Figure II.30). The copper must be thick enough to prevent corrosion from penetrating the canister as long as the spent fuel radiotoxicity substantially exceeds that in a rich uranium ore. The combined thickness of steel and copper should be enough to prevent any significant radiolysis of water outside the canister after deposition in wet bentonite clay. The steel insert is designed to withstand the normal mechanical loads that will prevail in the repository, such as hydrostatic pressure and the bentonite swelling pressure. The total weight of a canister with fuel will be about 25 t.

Figure II.30 **Overview of the canister**



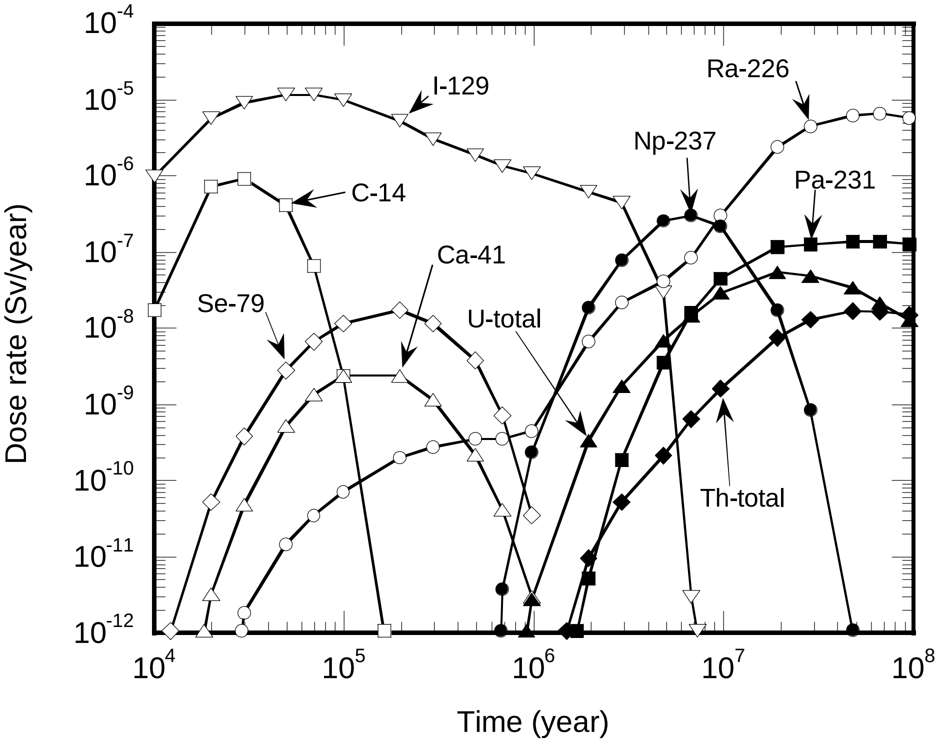
The steel insert is cast with thick steel walls between fuel assemblies. This gives a good mechanical stability besides providing adequate protection against criticality in the unlikely event that the canister at some unspecified future time should be filled with water.

The fabrication of copper canisters of the size needed is by no means an industrially available technology. The seal welding technology has been demonstrated on a laboratory scale in work sponsored by SKB at The Welding Institute in UK. Full-size canisters have also been fabricated on a trial scale. In order to develop the key technology, SKB operates a laboratory for encapsulation technology at Oskarshamn. This laboratory will be ready in 1998 and will primarily be devoted to further development of the seal-welding process and of the NDE-methods.

4.5.1.3 Performance assessment of spent fuel disposal in clay [190]

Taking into account the uncertainty about the choice of reprocessing as a fuel cycle step, a study was recently (1996) undertaken by the nuclear sector in Belgium to determine the impact of 5000 tHM spent fuel (consisting of a mixture of UO_2 and MOX fuel with burn-ups ranging from 33 to 45 GWd/tHM) on the Boom Clay repository environment (see next paragraphs describing the RFC scenarios) in Belgium. The result of the calculation shows that ^{129}I is the most important contaminant giving rise to about 10 *m*Sv/year from 20 000 to 200 000 years after disposal. The actinide dose is several orders of magnitude below that figure during this period (see Figure II.31), and crosses the ^{237}Np curve at around 3 000 000 years. Beyond that “geologic” period the decay products of U and Np become predominant. The very long-term dose on a geological time scale is determined by ^{226}Ra and ^{231}Pa. This dose is of the same order of magnitude as the initial ^{129}I dose, i.e. 10 *m*Sv/year. As a conclusion, ^{129}I dominates the dose rate between 10 000 and 2 000 000 years; later, the actinides and their decay products determine the ultimate dose.

Figure II.31 **Evolution of the expectation value of the dose rates in the case of disposal of 45 GWd/tHM MOX spent fuel**



In P&T studies, attention has always been concentrated on the residual radiotoxicity, and from that perspective the separation of actinides is the most important issue. However, when approaching the problem through a "dose to man" perspective in a normal or "upwelling" scenario, the long-lived fission products (^{129}I and ^{99}Tc) are the most important radionuclides to be eliminated since they are particularly mobile in clay and tuff media, respectively. Their elimination by transmutation without prior reprocessing is a very problematic issue both technically (very long irradiation times) and economically (no fission energy). Quantitative transmutation of these nuclides seems, in the present state of technology, very difficult to achieve within a reasonable time frame.

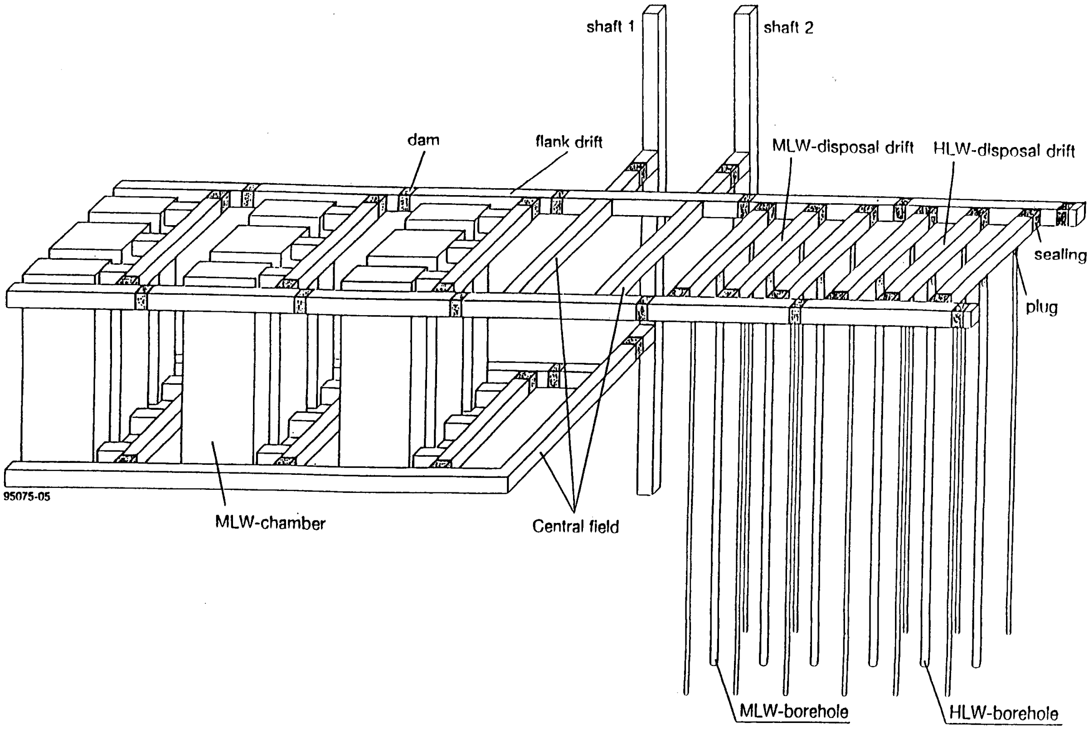
4.5.2 RFC concepts for disposal

In the case of the RFC, attention in waste management and disposal is focused on the conditioning of the High Level Waste (HLW) and Medium Level Waste (MLW) forms produced during the reprocessing operations and their disposal in geological formations. Two case studies are briefly described in this report: the disposal of HLW in salt domes, as proposed by Germany, and disposal in clay layers as investigated in Belgium. In both case-studies the source term is made up of the waste canisters produced by the Cogéma reprocessing facilities of La Hague in France. The sizes of the nuclear programs are obviously different (29 GWe and 5.7 GWe).

4.5.2.1 Disposal in salt formations [193]

The reference repository in salt formations is the Gorleben salt dome situated in northern Germany which has a horizontal extension of 14×4 km^2. The base of the repository is 800 m below the surface. The reference repository is situated 540 m below the cap-rock. A diagram of the repository structure for HLW and MLW is shown in Figure II.32. After closure, the repository structure will be filled-up with brine contained by dams with permeabilities of 7×10^{-16} m^2. In the German concept of radioactive waste disposal, heat producing waste will be disposed of in bore holes, and non-heating wastes in chambers. About 58 000 canisters HLW (10 454 m^3) corresponding to a nominal initial quantity of 73 000 tHM spent fuel will be disposed of. Feed sludges and cladding wastes totalling 80 000 m^3 will accompany the HLW in the repository structure. The most important safety feature is the convergence of the salt towards the cavities created for the emplacement of the waste.

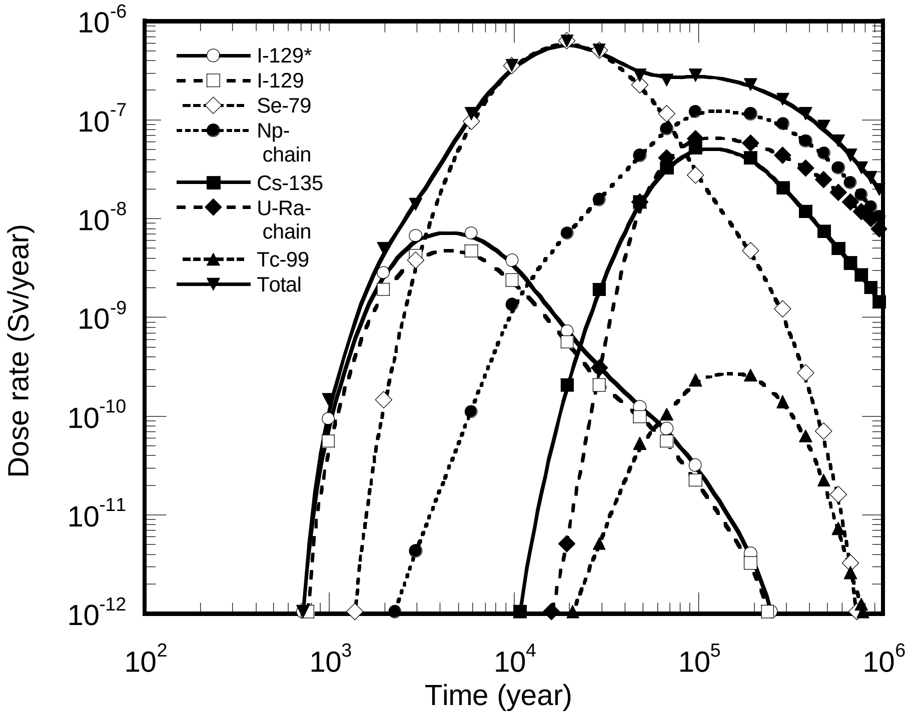
Figure II.32 **Reference repository in salt formations in the Gorleben salt dome**



In the framework of the EVEREST project [194], several scenarios have been investigated: the convection-diffusion scenario, the cavern convection scenario and the uplift scenario.

The calculations show that the maximum dose generated according to the convection-diffusion scenario does not exceed 0.7 *m*Sv/year after 20 000 years. The following nuclides are responsible for the bulk of the radiological burden: ^{79}Se, ^{129}I, ^{135}Cs, ^{237}Np and its daughter products. The peak in annual dose ranges from 6 000 years for ^{129}I through 30 000 years for ^{79}Se to 100 000 years for ^{237}Np and finally to 200 000 years for ^{99}Tc and ^{135}Cs. The highest annual dose due to ^{237}Np amounts to 0.12 *m*Sv/year at the peak release. The parameter uncertainties involved in the calculations are predominantly related to the geophysical conditions (sorption, permeability-porosity) prevailing during the elapsed time period. Figure II.33 shows the evolution of the dose rates for the most significant radionuclides.

Figure II.33 **Evolution of the individual dose rate for the most important nuclides (I-129*: refers to full ^{129}I inventory in spent fuel)**



In the event of human intrusion occurring between 500~1 000 years after closure (cavern-convection scenario), the peak annual dose is one order of magnitude higher (1 *m*Sv/year) than in the convection-diffusion case, but the time of arrival of the peak concentrations is much delayed. The most sensitive parameter being the location of the intrusion with respect to the repository zone. Figure II.34 shows the dose rate evolution for the cavern-convection scenario.

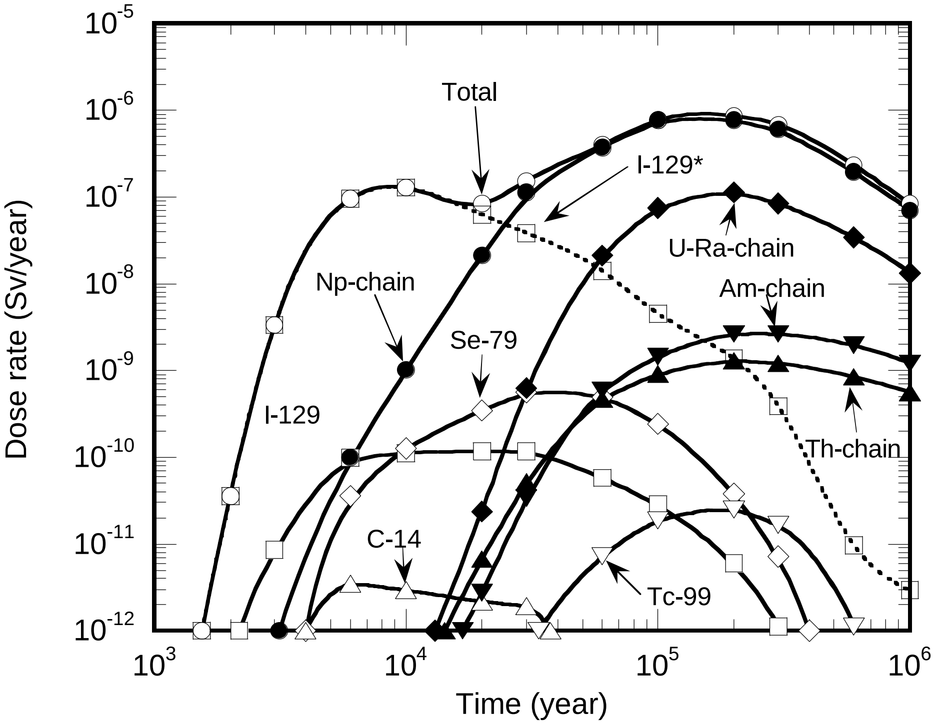
In the uplift scenario the doses depend essentially upon the uplift rate. The maximum dose rate ranges from 0.6 *m*Sv/year for an uplift rate of 3.3×10^{-5} m/year to 100 *m*Sv/year at an uplift rate of 2×10^{-4} m/year. However, the time of arrival of the peak concentrations is extremely long i.e. 2.6 to 16 million years.

4.5.2.2 Disposal in clay formations [193,194]

The Boom clay formation at the Mol site in Belgium was chosen as the reference case for the performance studies on repositories in argillaceous formations. There are however many similar clay formations throughout the European continent and the United Kingdom. A schematic view of a national repository in the Boom Clay is shown in Figure II.35.

The Boom Clay layer is about 100 m thick and situated at a mean depth of 220 m. It consists of about 80% of illite and vermiculite clay minerals with a very fine grain size of 2 to 20 *m*m. The overall permeability of the clay layer is below 10^{-10} m/s which reduces migration to diffusion. An underground laboratory has been installed since 1982 and no adverse hydrological behaviour has been observed for the last 15 years. In situ and laboratory migration data have been obtained for a series of FPs and actinides. Owing to the strong sorption capacity of the clay, the migration velocity is so low that most of the radionuclides embedded in vitrified HLW canisters cannot leave the clay body as radioactive species except for ^{237}Np, ^{99}Tc, ^{135}Cs, ^{231}Pa and long-term daughter products of Np.

Figure II.34 **Cavern-convection scenario, evolution of individual dose rate for the most important nuclides (I-129*: refers to full ^{129}I inventory in spent fuel)**



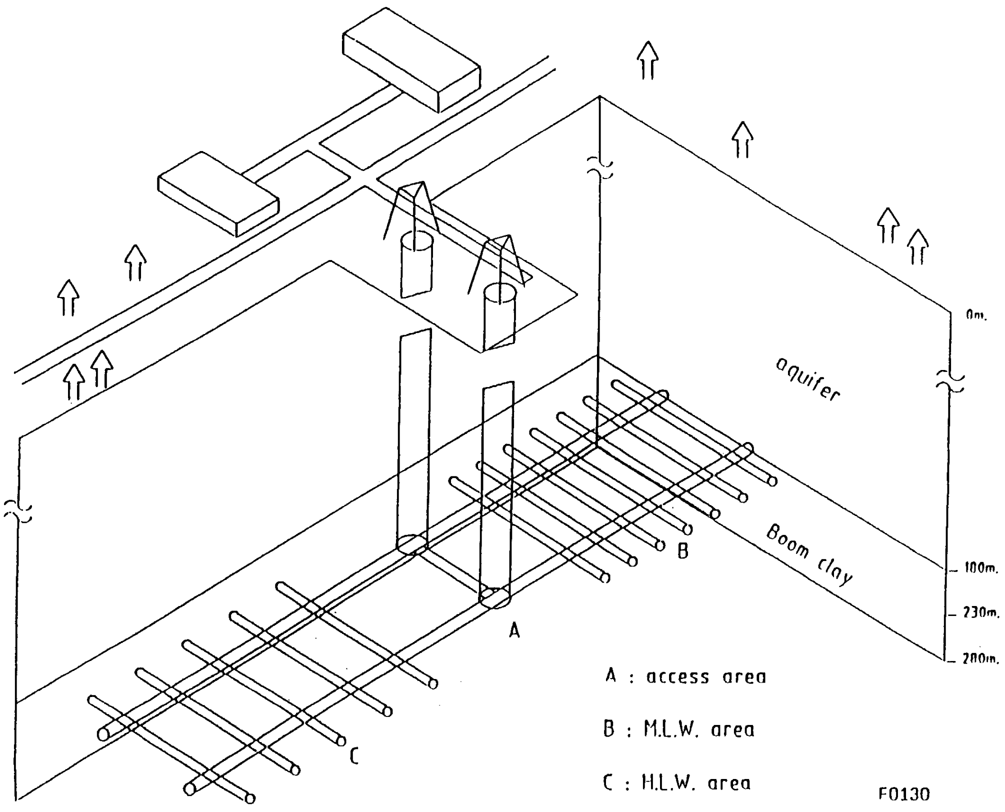
In 1988, a comparative performance study (PAGIS) was undertaken within the framework of the European Commission in order to assess the long-term impact of the disposal of HLW resulting from a conceptual 10 GWe reactor park operated for 30 years. The equivalent of 8 180 tHM glass canisters (about 900 m^3 vitrified HLW) were considered as the radioactive source term (fission products, minor actinides and corrosion products). The calculations were refined in the PACOMA [193] and EVEREST [194] projects.

The actinides Am and Cm and the daughter product ^{240}Pu decay completely within the first 10 m of the clay layer. This is not the case with ^{237}Np or with uranium if it were disposed of. The maximum annual dose due to the presence of ^{237}Np is about 0.02 *m*Sv/year when the use of drinking water from a well in the upper aquifer near the clay layer is considered as the main pathway. According to the same scenario, the doses due to ^{135}Cs and ^{99}Tc amount to 0.015 *m*Sv/year and 0.15 *m*Sv/year, respectively. Figure II.36 shows the contribution of the different nuclides.

In a climate change scenario, where water underlying the clay layer might be used for drinking purposes, the dose to man due to ^{129}I would reach the tolerance level of 0.2 mSv/year if all the iodine waste (as AgI) recovered from the effluents of the reprocessing plant were stored in the clay repository.

Since 95 to 99% of ^{129}I is separated from the HLW during the reprocessing operations and discharged in the ocean, only 1 to 5% is supposed to be associated with the cladding materials, so the dose resulting from the leaching and migration of ^{129}I in the near field comes from MLW and would amount to 30 *m*Sv/year. Partitioning of ^{129}I during reprocessing and washing of the hulls in order to minimise the residual quantities of iodine is therefore very important. Conventional transmutation of iodine waste by n-γ reaction in LWRs has proved not to be very effective. Selective transmutation reactions by alternative irradiation facilities (ADS or other types of reactor) capable of transforming this nuclide into an inactive species should therefore be investigated.

Figure II.35 **A schematic view of a repository in the Boom Clay**



Scheme of the repository

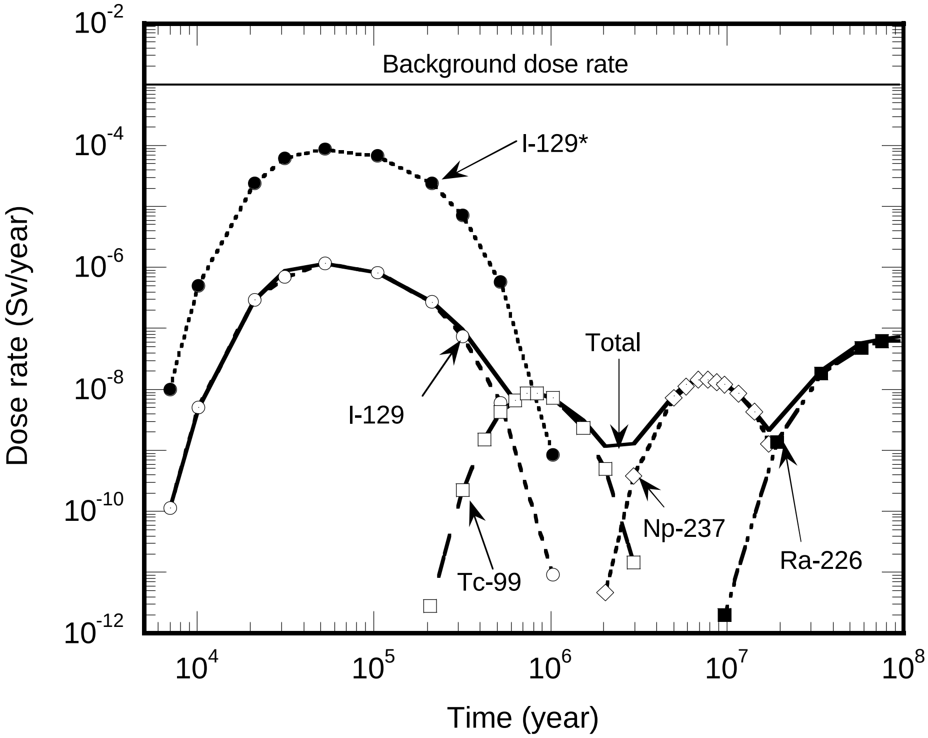
4.5.3 AFC-specific waste management issues (targets, reactor cores)

It is premature to make forecasts about the waste management of irradiated targets and residual reactor cores since it will depend on the future evolution of nuclear energy production.

However, the immediate potential benefit of the AFC scenario would be the quantitative reduction of actinides in the vitrified HLW by a factor of 10 to 100. This effect will essentially improve the hazard perception of the vitrified HLW but will not reduce the needs for repository construction which are determined by the heat emission of the waste and not by the radiotoxic inventory. The underground gallery space necessary for the disposal of vitrified waste is independent of its MA content during the first 200 years period during which the repositories will have to be built and operated.

In order to reduce the waste disposal costs, the heat producing nuclides (^{137}Cs and ^{90}Sr) have to be separated. The removal of these nuclides from HLLW will have a beneficial effect on the volume of the repository and as a consequence on its overall cost. But it is not very plausible to imagine a surface storage of these radioisotope concentrates for hundreds of years. However, the separated nuclides have, under this hypothesis, to be stored for a period of about 300 years in engineered facilities.

Figure II.36 **Normal evolution scenario; calculated total individual dose rate for the water pathway (^{129}I*: refers to ^{129}I in MLW)**



The separation and transmutation of some very long-lived fission products, e.g. ^{129}I, ^{99}Tc and some other nuclides depending on the type of waste or repository, would give rise to larger volumes of medium-and low-level waste than presently encountered. But the total radiotoxic inventory of these FPs will be lower if the irradiation process has been effective. The residual FP targets will be considered as MLW rather than as HLW.

The major potential benefits of an AFC scenario are as follows:

- the further reduction of plutonium and MA inventories in vitrified HLW and geological disposal;
- a general reduction of TRU inventories in all waste fractions and a decrease in the residual spent fuel inventory through systematic recycling into fission reactor cores (LWR and FR), and later into hybrid reactor systems that can more exhaustively deplete TRU inventory could before disposal;
- the use of potentially more efficient types of conditioning which could be adapted to each separated nuclide or depleted target, so reducing the radiological risk associated with the individual radionuclide source;
- the possibility to complement current MA and FP destruction techniques by future developments. Existing nuclear destruction techniques for long-lived fission products are unsuited to industrial application. The ^{90}Sr, ^{99}Tc and ^{129}I targets could be specific examples for which future neutronic developments might bring unexpected solutions.

4.6 Criticality safety

Typical LWR fuel contains some 3 – 4% of fissile material (^{235}U) before irradiation and about 1.5% fissile material (^{235}U plus ^{239}Pu and ^{241}Pu) after a burn-up of 40 GWd/tHM. Fuel from other thermal power reactors has similar fissile contents. One exception is natural uranium fuel (e.g. from HWR) where the fissile content of spent fuel is of the order 0.5% or less.

The used fuel is proposed to be encapsulated in canisters with a content of a few tonnes heavy metal before disposal. This means that one canister with spent enriched nuclear fuel will contain more fissile material than the theoretical minimum critical mass. A whole repository will contain many times more. The disposal of spent nuclear fuel thus means that the potential for an unintended critical configuration of the fissile material has to be addressed in the safety analysis.

Two main cases have to be considered:

- potential risk for criticality of the fuel as deposited in the canister;
- reconfiguration of the fissile material to a critical configuration, e.g. by selective leakage and precipitation of fissile elements.

The fuel will (in many proposed cases) be encapsulated in the same geometrical configuration as used in the reactor, (i.e. close to optimum from the reactor physics point of view), the temperature in the repository is ambient (much lower than in a reactor), and the short-lived strongly neutron-absorbing radionuclides such as ^{135}Xe have decayed. This means that criticality is conceivable with only a limited number of fuel assemblies if a moderating material is added. A spent fuel canister must therefore be designed in such a way that criticality is not achieved even if the canister is filled with fuel that for some reason has not reached full burn-up and starts taking in groundwater from the surrounding repository formation. In practice this is achieved by rearranging the fuel in a non-critical configuration, by having strict administrative control of the fuel burn-up and other important parameters in the encapsulation plant, and/or by mixing the fuel with some neutron-absorbing material, e.g. an insert with walls between each fuel assembly [195]. In the design and safety analysis several real or hypothetical phenomena must be accounted for, such as long-term reactivity changes, any dissolution of neutron absorbing material or any reconfiguration of fuel rods.

The issue of reconfiguration of the fissile material was addressed in the 1970s [196] and has recently been revisited [197,198]. The issue has two aspects: (1) the probability that some process will rearrange the fissile material into a configuration that might develop and sustain a neutron chain reaction, and (2) the probable or possible consequences of such a chain reaction if it should occur. The early studies of a repository in granitic bedrock [196] concluded that the reconfiguration of plutonium from several canisters is an extremely unlikely event owing to the very slow chemical processes in the prevailing geochemical environment as compared with the half-lives of fissile plutonium. Reconfiguration of uranium from LWR-fuel is also very unlikely because (owing to the low ^{235}U content) several tonnes of uranium must be assembled in proper concentration and configuration. Criticality with uranium could, for geometrical reasons, only occur in the backfilled excavations of the repository, where re-concentration by absorption is possible, and would require the movement of uranium from several canisters to the same spot during a very long time. The consequence of a hypothetical criticality in plutonium or uranium from spent LWR fuel was furthermore judged to be very limited.

Some analyses [197] point out that fissile material in certain concentrations and configurations could pose a risk of divergent neutron chain reactions (self-sustained criticality) with considerable energy release as a result. The probability of creating such a configuration has not however been properly assessed. Other analyses [198] challenge this scenario and point out a number of factors that will make it very unlikely indeed for a spent fuel repository. A recent study [199] for the proposed Yucca mountain repository concludes that the concentrations reached for ^{239}Pu (or later for ^{235}U) from vitrified weapons-grade plutonium are too low for self-sustained criticality. In conclusion, conceivable ways of creating a risk of unintentional criticality have to be addressed for any repository for large amounts of fissile material. The assessment has to be done for each specific geological environment and repository design. There are however means to avoid such a risk and to make the criticality scenarios very unlikely indeed.

5. COMMENTARY ON EXISTING P&T SYSTEMS STUDIES

5.1 First generation systems study reports

Partitioning and transmutation as a new waste management issue was initiated in the United States during the 1970s and investigated from a theoretical and assessment perspective at the Oak Ridge National Laboratory. Claiborne [200] demonstrated in 1972 the neutron-physical feasibility of transmuting "by-product actinides" in LWRs. Simultaneously the German Research Centre of Karlsruhe, the CEA in France and European Commission at the Joint Research Centre of Ispra started a comprehensive theoretical and experimental R&D programme. In France, the Castaing Commission 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 partitioning-transmutation approach aiming at the reduction of the radiotoxic inventory by recycling long lived nuclides into fission reactors.

Four major final "assessment" reports were published in the early 1980s [201-205] which led to the following conclusions:

- the ORNL studies conclude that there are no cost or safety incentives for partitioning and transmutation of actinides 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 currently available LWRs;
- the conclusions of the EC programmes on P&T were that the impossibility of total actinide recycling and the impact of the process flowsheets' complexity on waste streams are 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 ($<5\times10^{-4}$);
- transmutation of MAs is theoretically feasible from the point of view of neutron physics and fuel cycle technology but it is 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 concludes that it is 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 IAEA assessment report on P&T investigated the technical feasibility and the radiological impact. 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 fission products 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 period of active investigation on P&T starting in 1972 was terminated around 1982-83 as no international consensus was obtained on the benefits of P&T as an alternative or complementary waste management option.

5.2 Second generation reports

During the eighties, a growing awareness of the inherent difficulties in creating and licensing large nuclear repositories, and growing delays in the 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.

In 1988, the Japanese government launched the ambitious OMEGA R&D programme and invited the international community, through the OECD/NEA agency, 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.

Important experimental programmes were conducted in the United States at the Argonne National Laboratory (ANL) and large scale R&D programmes are still being conducted in Japan at JAERI and JNC, and in France at the CEA in co-operation with several European countries under sponsorship of the European Commission.

As a result of this sudden explosive growth in R&D programmes, the need was felt to re-examine the validity of the P&T option in the light of the recent results. A series of American reports has been published in the meantime.

On the basis of a ORNL retrospective assessment of P&T [206], the Electric Power Research Institute (EPRI) started a detailed evaluation programme [207] on the concept of transuranic burning using liquid metal reactors 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 [208].

The main conclusions of these US reports are:

- 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 spent fuels from LMRs would be reprocessed in a smaller scale pyrochemical unit which has still to be developed;

- 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 make the acceptance of P&T as a waste management scenario very improbable under the present economic conditions;

- the radiotoxicity of a repository is not accepted as a scientific argument in the assessment of the risk except in the case of human intrusion.

Only the combined use of ^{137}Cs – ^{90}Sr separation and actinide burning followed by a long (300 years) surface storage would alleviate the repository heat problem.

The most recent and most comprehensive assessment report on P&T was issued by the National Academy of Science of the US under the chairmanship of N.C. Rasmussen [209]. The report is entitled "Nuclear Wastes: Technologies for Separations and Transmutation" and 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.

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 [210-212] 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).

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