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.

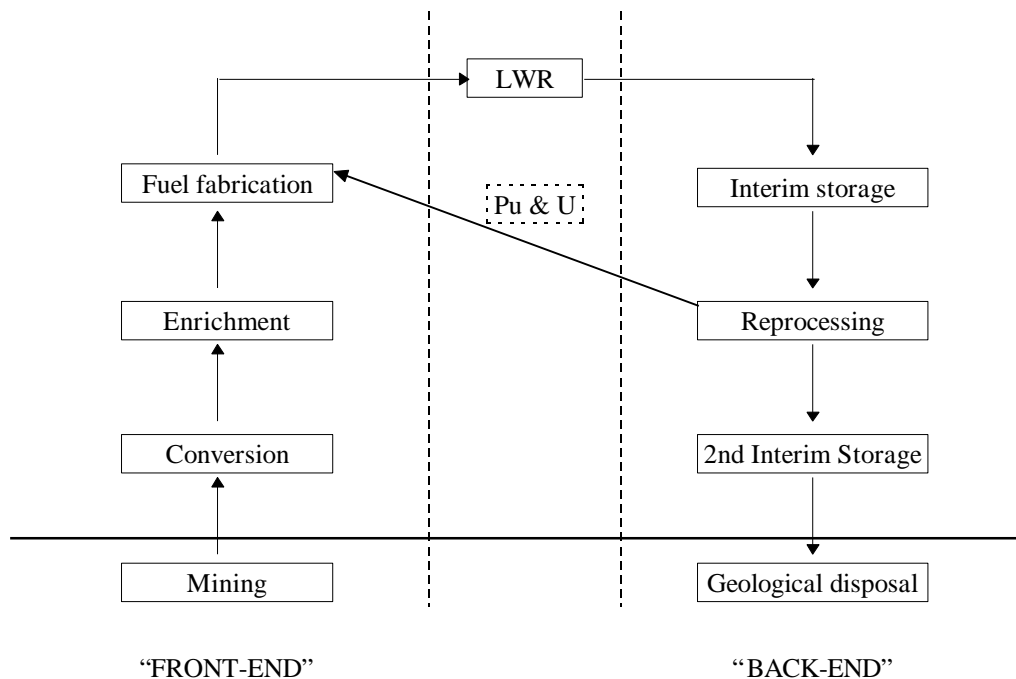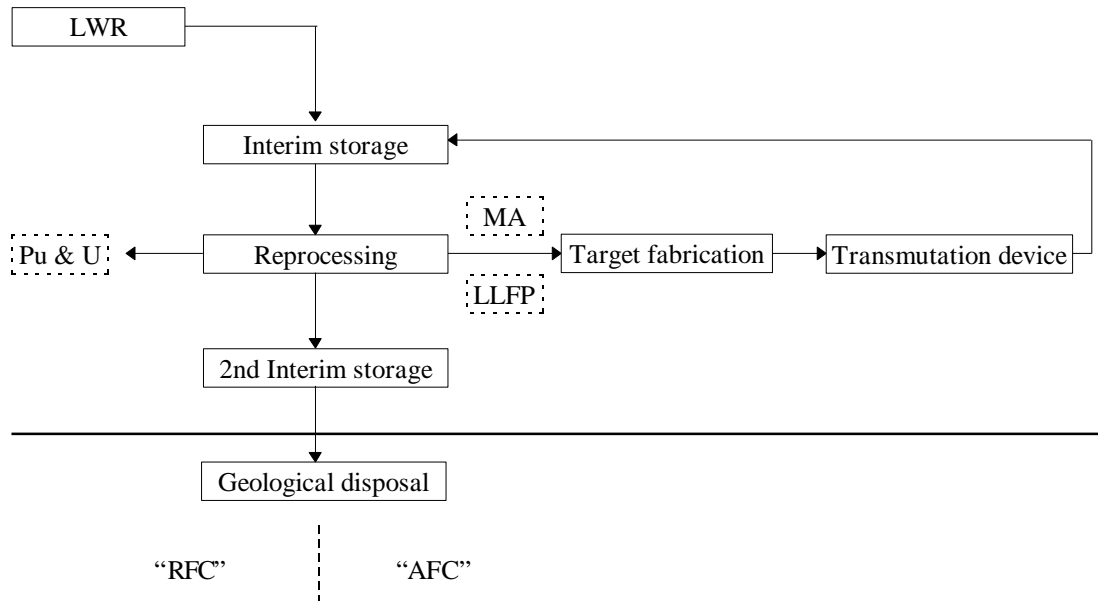Figure I.1 **A schematic diagram of the reprocessing fuel cycle for LWRs**

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

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┌────────┐     ┌──────────────┐                           │    Target    │  │  Transmutation       │
│ Pu & U │◄─── │ Reprocessing │──────────────────────────►│  fabrication │─►│     device           │
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     "RFC"              ┊            "AFC"
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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 $\left[(2\times5) - (1\times10) \right] \cong 0 \text{ kg}$ 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 $^{99}TcO_4^-$. 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.

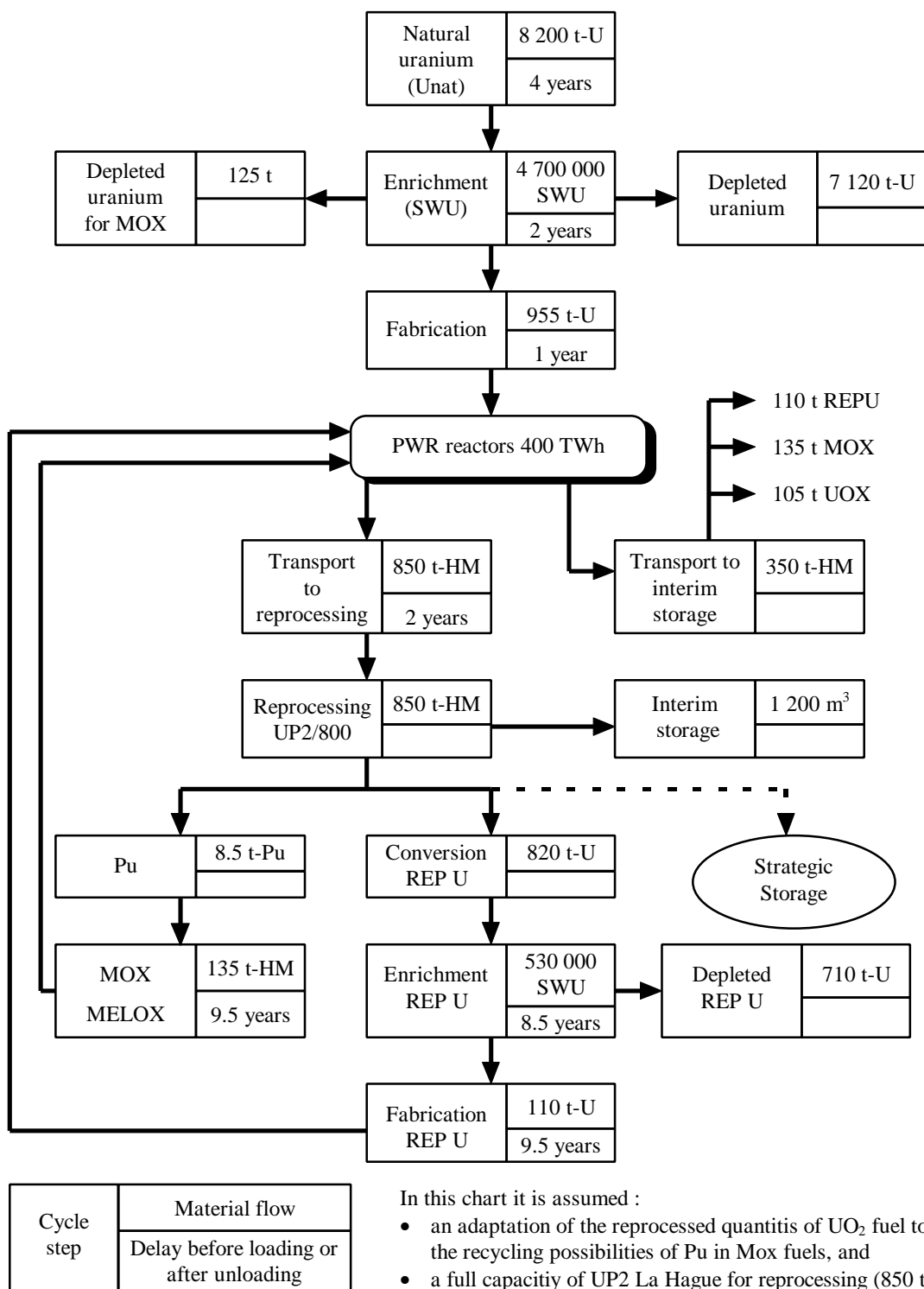Figure I.3 **A schematic diagram of the reprocessing fuel cycle for a 400-TWh LWR park (French case study)**

Natural uranium (Unat)	8 200 t-U
	4 years

Depleted uranium for MOX	125 t		Enrichment (SWU)	4 700 000 SWU		Depleted uranium	7 120 t-U
				2 years			

Fabrication	955 t-U
	1 year

PWR reactors 400 TWh

→ 110 t REPU
→ 135 t MOX
→ 105 t UOX

Transport to reprocessing	850 t-HM		Transport to interim storage	350 t-HM
	2 years			

Reprocessing UP2/800	850 t-HM		Interim storage	1 200 m^3

Pu	8.5 t-Pu		Conversion REP U	820 t-U		Strategic Storage

MOX MELOX	135 t-HM		Enrichment REP U	530 000 SWU		Depleted REP U	710 t-U
	9.5 years			8.5 years			

Fabrication REP U	110 t-U
	9.5 years

Cycle step	Material flow
	Delay before loading or after unloading

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.

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

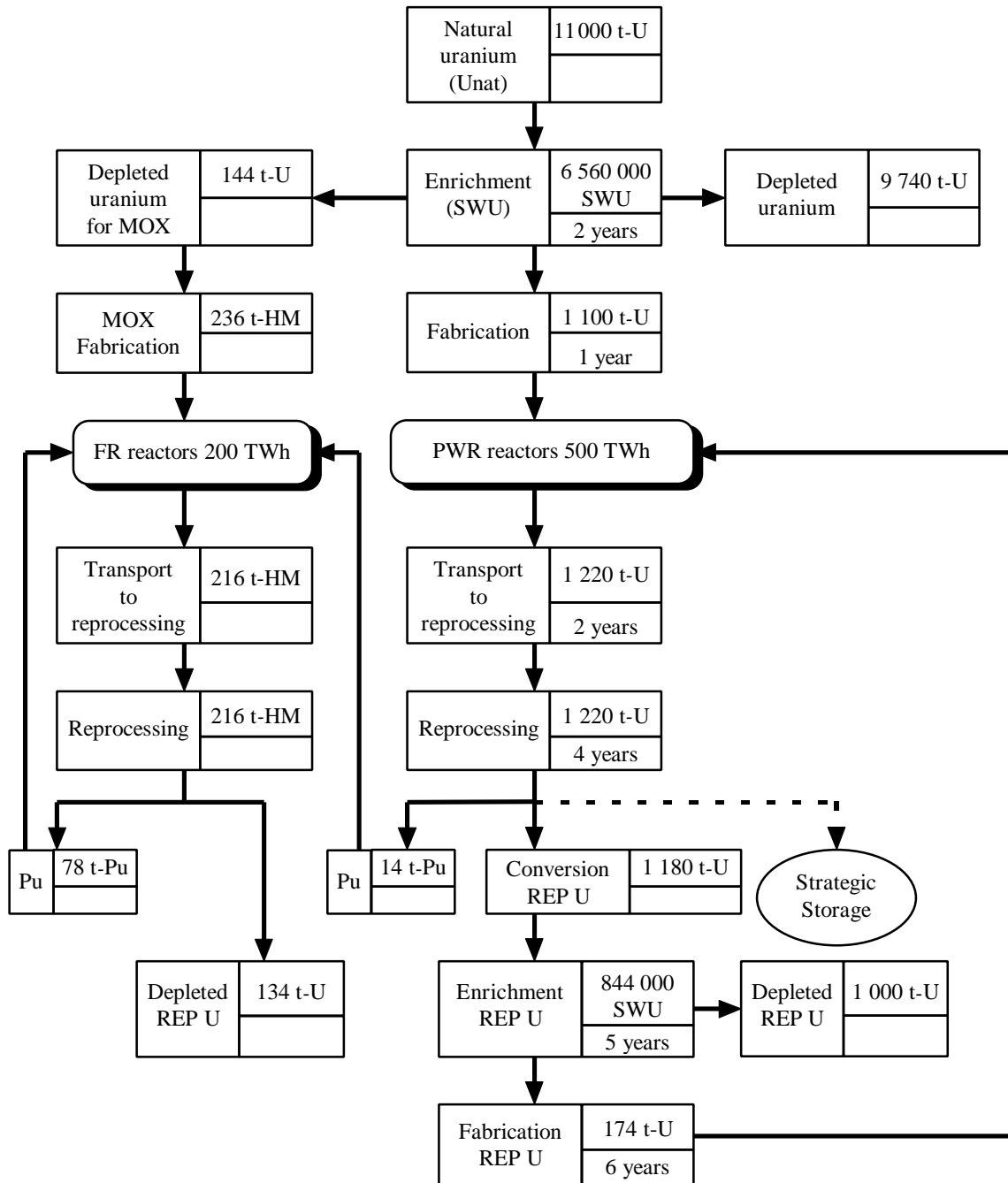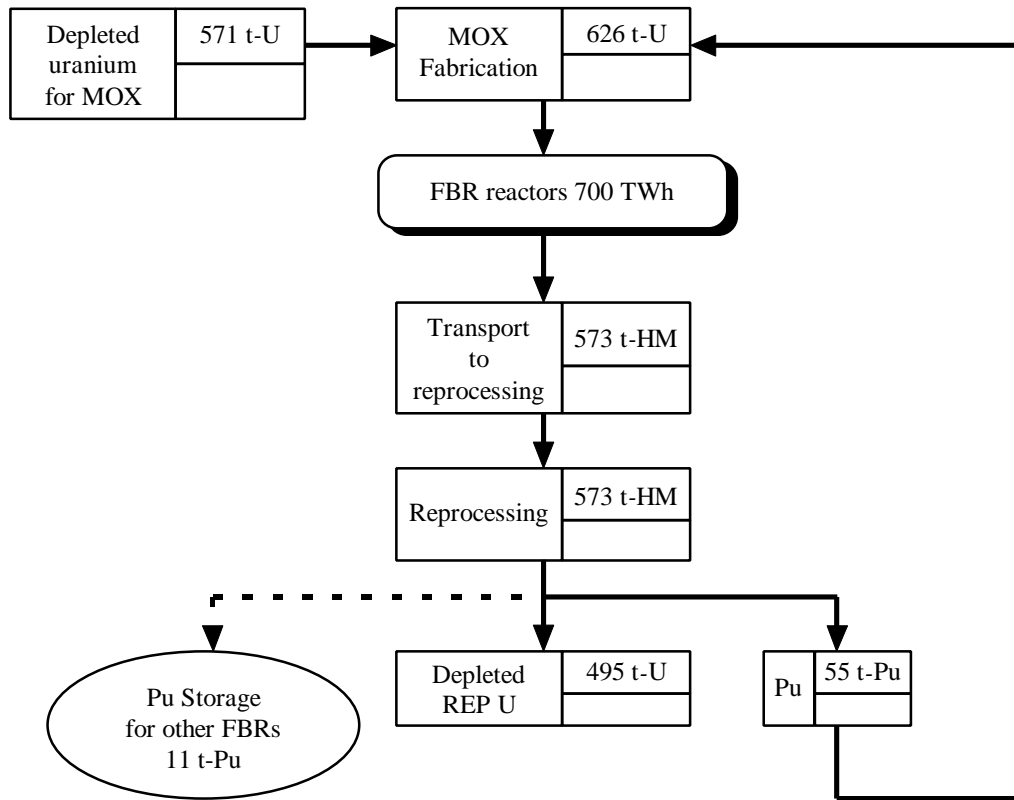Natural uranium (Unat) — 11 000 t-U

Enrichment (SWU) — 6 560 000 SWU — 2 years

Depleted uranium for MOX — 144 t-U

Depleted uranium — 9 740 t-U

MOX Fabrication — 236 t-HM

Fabrication — 1 100 t-U — 1 year

FR reactors 200 TWh

PWR reactors 500 TWh

Transport to reprocessing — 216 t-HM

Transport to reprocessing — 1 220 t-U — 2 years

Reprocessing — 216 t-HM

Reprocessing — 1 220 t-U — 4 years

Pu — 78 t-Pu

Pu — 14 t-Pu

Conversion REP U — 1 180 t-U

Strategic Storage

Depleted REP U — 134 t-U

Enrichment REP U — 844 000 SWU — 5 years

Depleted REP U — 1 000 t-U

Fabrication REP U — 174 t-U — 6 years

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

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┌─────────────┬──────────┐              ┌─────────────┬──────────┐
│ Depleted    │ 571 t-U  │─────────────▶│ MOX         │ 626 t-U  │◀──────────┐
│ uranium     │          │              │ Fabrication │          │           │
│ for MOX     │          │              │             │          │           │
└─────────────┴──────────┘              └─────────────┴──────────┘           │
                                              │                               │
                                              ▼                               │
                                  ╭───────────────────────╮                   │
                                  │  FBR reactors 700 TWh  │                   │
                                  ╰───────────────────────╯                   │
                                              │                               │
                                              ▼                               │
                                  ┌─────────────┬──────────┐                  │
                                  │ Transport   │ 573 t-HM │                  │
                                  │ to          │          │                  │
                                  │ reprocessing│          │                  │
                                  └─────────────┴──────────┘                  │
                                              │                               │
                                              ▼                               │
                                  ┌─────────────┬──────────┐                  │
                                  │ Reprocessing│ 573 t-HM │                  │
                                  └─────────────┴──────────┘                  │
                         ┌ ─ ─ ─ ─ ─ ─ ─ ─ ┴──────────┬──────────┐           │
                         ▼                  ▼          ▼                      │
                  ╭──────────────╮   ┌────────────┬─────────┐  ┌────┬────────┐│
                  │ Pu Storage   │   │ Depleted   │ 495 t-U │  │ Pu │ 55 t-Pu││
                  │ for other    │   │ REP U      │         │  └────┴────────┘│
                  │ FBRs 11 t-Pu │   └────────────┴─────────┘       │        │
                  ╰──────────────╯                                  └────────┘
```

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.

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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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