

**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 $M \cdot A_n \cdot 2TBP$ (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).

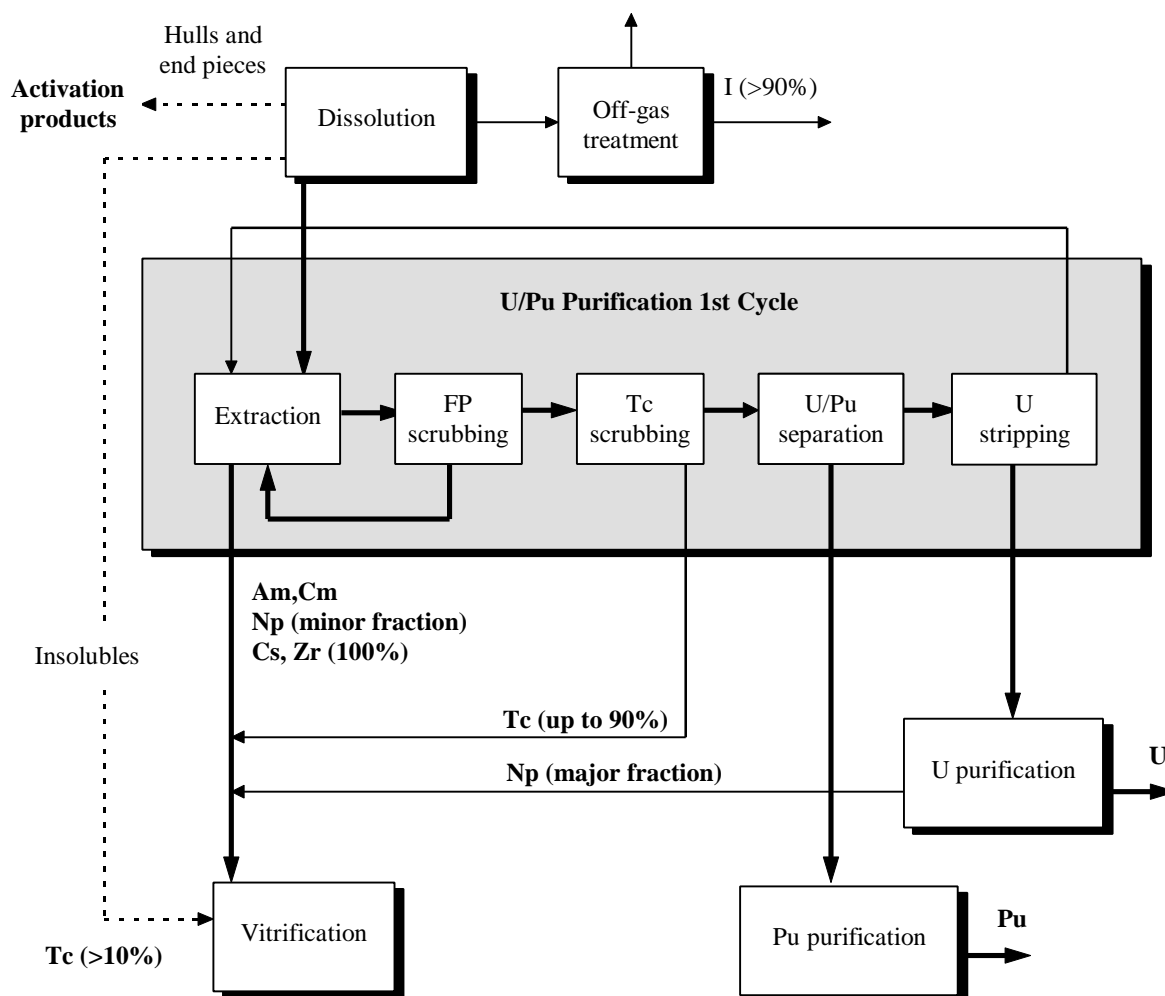
Technetium

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 UO₂ 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, TcO₄⁻. 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 ZrO²⁺ (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

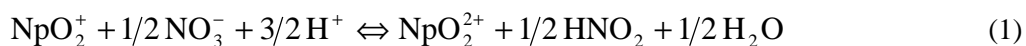
Neptunium is present in the irradiated fuel dissolution liquor (nitric acid medium) in oxidation states V and VI. The extractability of Np(V) by tributylphosphate (TBP) organic solution (the PUREX solvent) is rather poor whereas that of Np(VI) is good, approaching that of U(VI) and Pu(IV). Hence one 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:



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 HNO₃. 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 CO₂ 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 (Ba¹⁴CO₃) 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 $R = \text{Ln}/\text{An}$ is high (R is ~ 20 for 47.5 GWd/tHM burn-up uranium oxide fuels).

Two alternatives are available to separate the An's 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

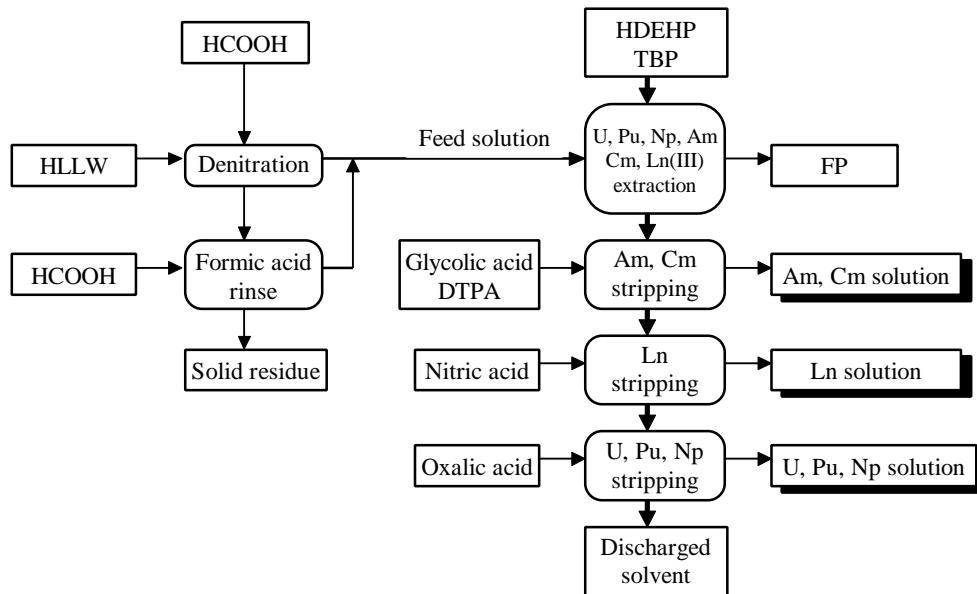
The principle of the minor actinide separation here is the co-extraction of the MAs with other elements present in the high active raffinate or in the HLLW, such as lanthanides, and then to partition the different extracted elements by selective stripping. Two processes are known in this category, both 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 ($\text{HNO}_3 = 0.5 \text{ M}$) 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.

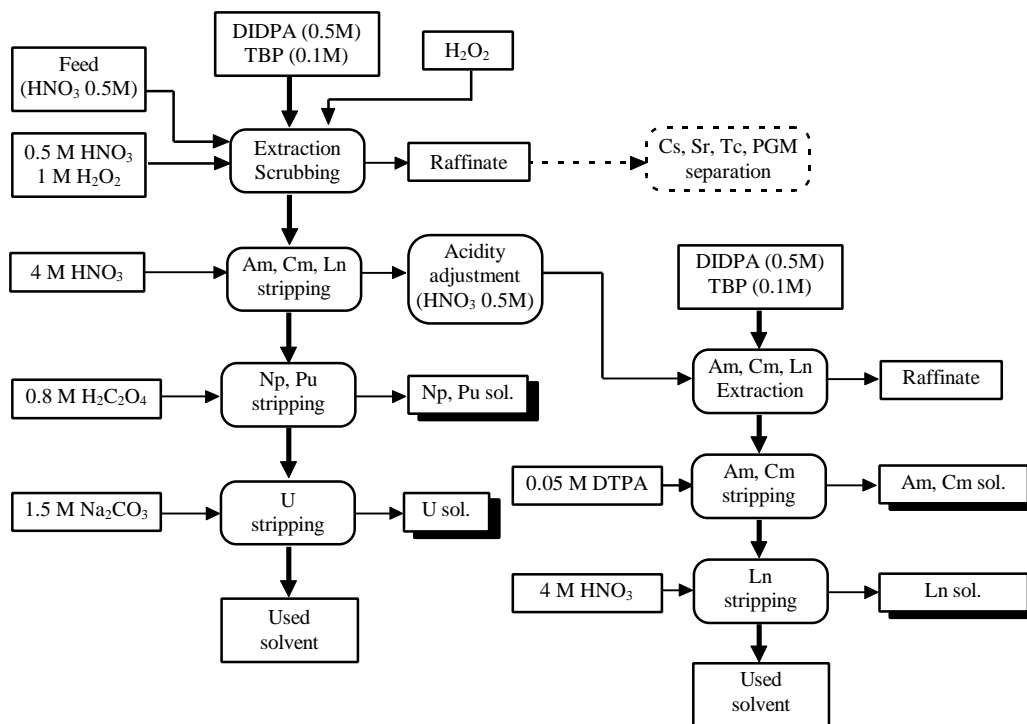
Figure II.2 TALSPEAK process



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 bidentate 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(III) 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.

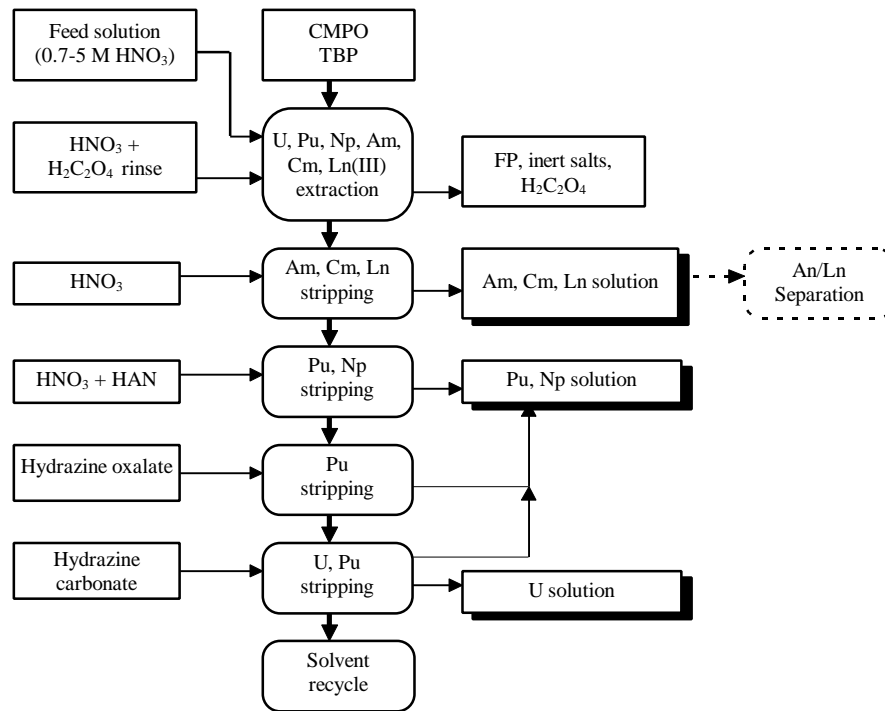
To cope with the third phase formation problem, the solvent contains a high concentration of TBP (1 to 1.4 M) used as a modifier. CMPO mixed with TBP allows the extraction of the most important actinides, except Np(V), and the process is directly applicable to HLLW solutions with a HNO₃ concentration of 0.7 to 5 M as it results from the conventional PUREX process. CMPO is such a powerful extractant (distribution ratio of 10⁴) 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-NaNO₃ 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 nitrophenyl-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)

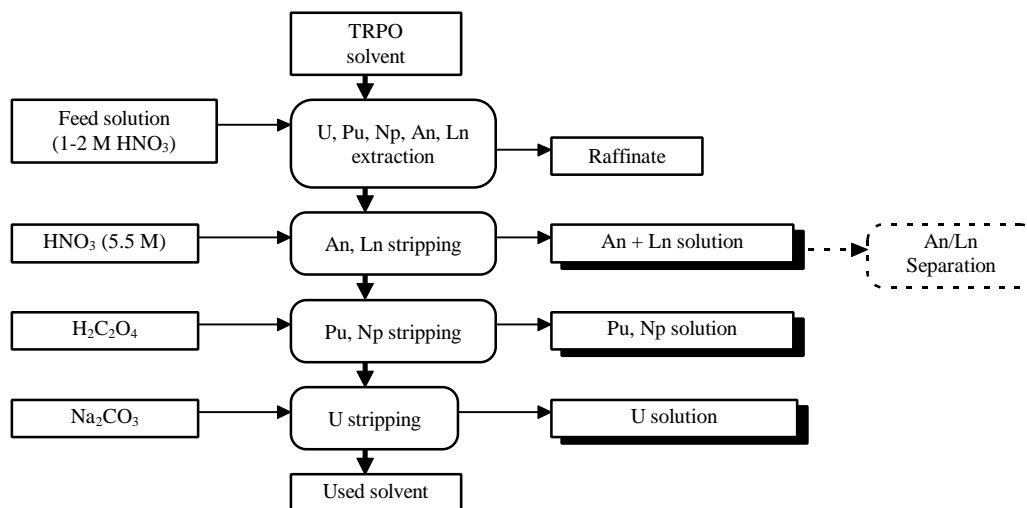
The TRPO process, developed by Zhu, Song *et al.* in Tsinghua University (China), is based on the use of liquid mixtures of TRPO soluble in aliphatic hydrocarbon diluent (kerosene) [14].

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 $\text{HNO}_3 = 5.5$ 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)

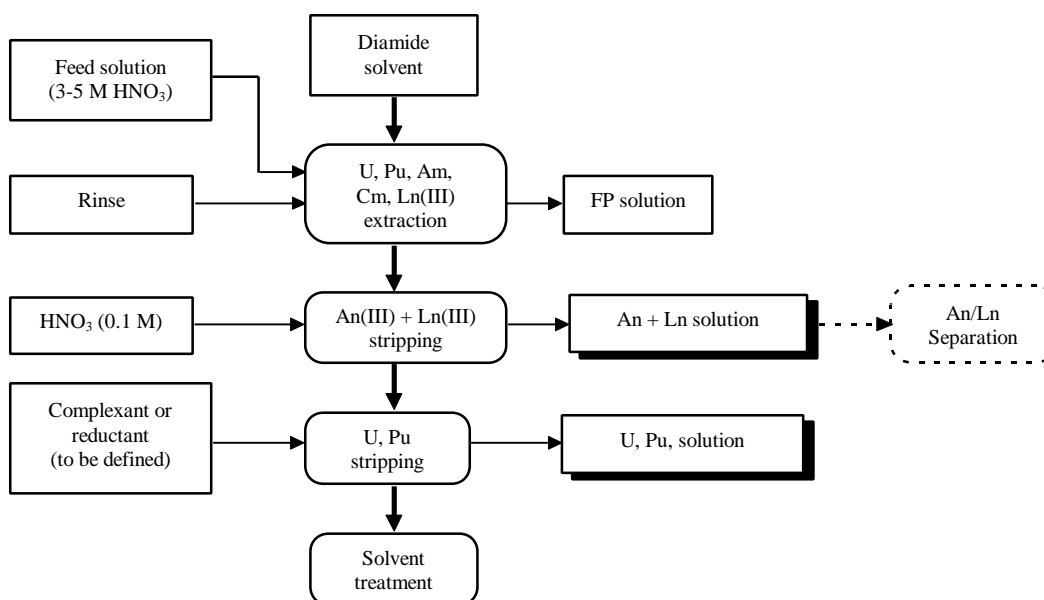
The DIAMEX (DIAMide EXtraction) process was first developed by Musikas *et al.* [15] at the CEA Fontenay-aux-Roses Research Centre (France) and by C. Madic and M.J. Hudson in a joint European research programme involving the CEA (Fontenay-aux-Roses) and the University of Reading (UK) [16]. This process 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 terdentate 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 ($pH \sim 4$) 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 $(Cl-C_6H_4)_2PS(SH)$ 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 ferricyanide 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 pertechnetetic 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).

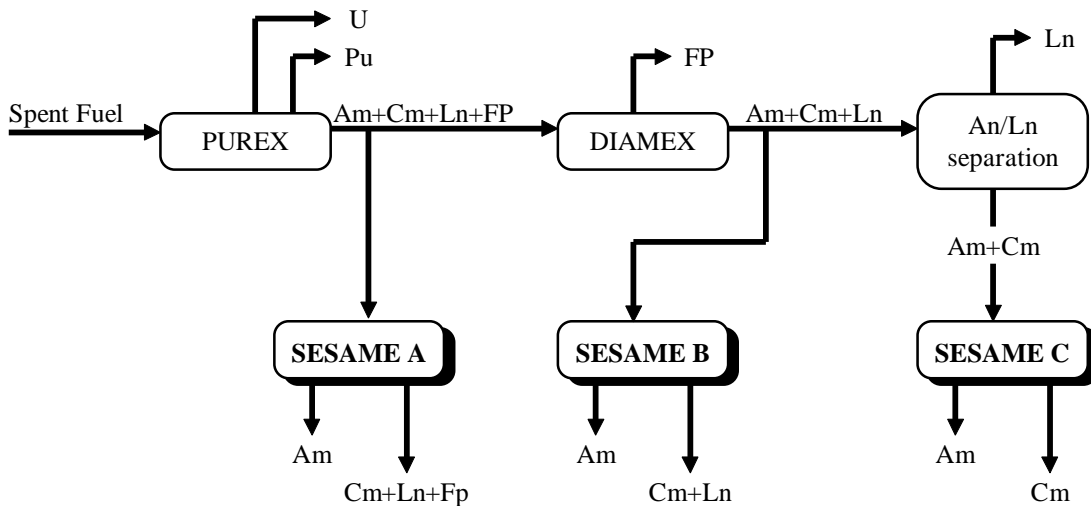
- Separation of americium from the high-level solution produced by the PUREX process (*SESAME A*). Technically speaking, this operation is complex because the solution contains many chemical elements that interfere with the oxidation and complexation of americium. These include ruthenium, which must 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, titanitic 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)		<input type="checkbox"/>	<input type="checkbox"/>	95% separation
(PUREX)		<input type="checkbox"/>		>95% separation
(DIDPA)		<input type="checkbox"/>		
(HDEHP)		<input type="checkbox"/>		
(TRUEX)		<input type="checkbox"/>		
Am + Cm separation: * based on An/Ln co-extraction (TALSPEAK)			<input type="checkbox"/>	
(DIDPA)		<input type="checkbox"/>		
(TRUEX)		<input type="checkbox"/>		
(TRPO)		<input type="checkbox"/>		
(DIAMEX)		<input type="checkbox"/>		
* based on An selective extraction (TPTZ)	<input type="checkbox"/>			
(Picolinamides)	<input type="checkbox"/>			
(CYANEX 301)	<input type="checkbox"/>			SF=5 900
* based on precipitation (Ferricyanide)	<input type="checkbox"/>			
Am separation in the oxidised state (SESAME)		<input type="checkbox"/>		Am/Cm separation
Tc separation (PUREX)			<input type="checkbox"/>	soluble Tc
(PUREX)	<input type="checkbox"/>			insoluble Tc
Tc - PGM separation (Denitration precipitation)		<input type="checkbox"/>		
(Active carbon adsorption)		<input type="checkbox"/>		
I separation (PUREX)			<input type="checkbox"/>	95% separation
Zr separation (PUREX)		<input type="checkbox"/>		
Cs separation (Calixarenes)		<input type="checkbox"/>		
(Zeolite)		<input type="checkbox"/>		
Sr separation (Titanic acid)		<input type="checkbox"/>		
Cs and Sr separation (Dicarbollides)			<input type="checkbox"/>	
Pd (PGM), Se, Ru separation (Electrolytic extraction)	<input type="checkbox"/>			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 $(\text{U}, \text{Pu})\text{O}_2$.

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 UO₂ and PuO₂. 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 PuO₂ recovered will be mixed with UO₂ 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 pyroprocessing 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 UO₂ or co-precipitate with PuO₂ by supplementary electrorefining for UO₂-PuO₂ recovery. That behaviour was demonstrated during the BOR-60 spent MOX fuel reprocessing [40]. The process for Np pyrochemical co-deposition with UO₂ and UO₂-PuO₂ 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 UO₂-PuO₂ 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)

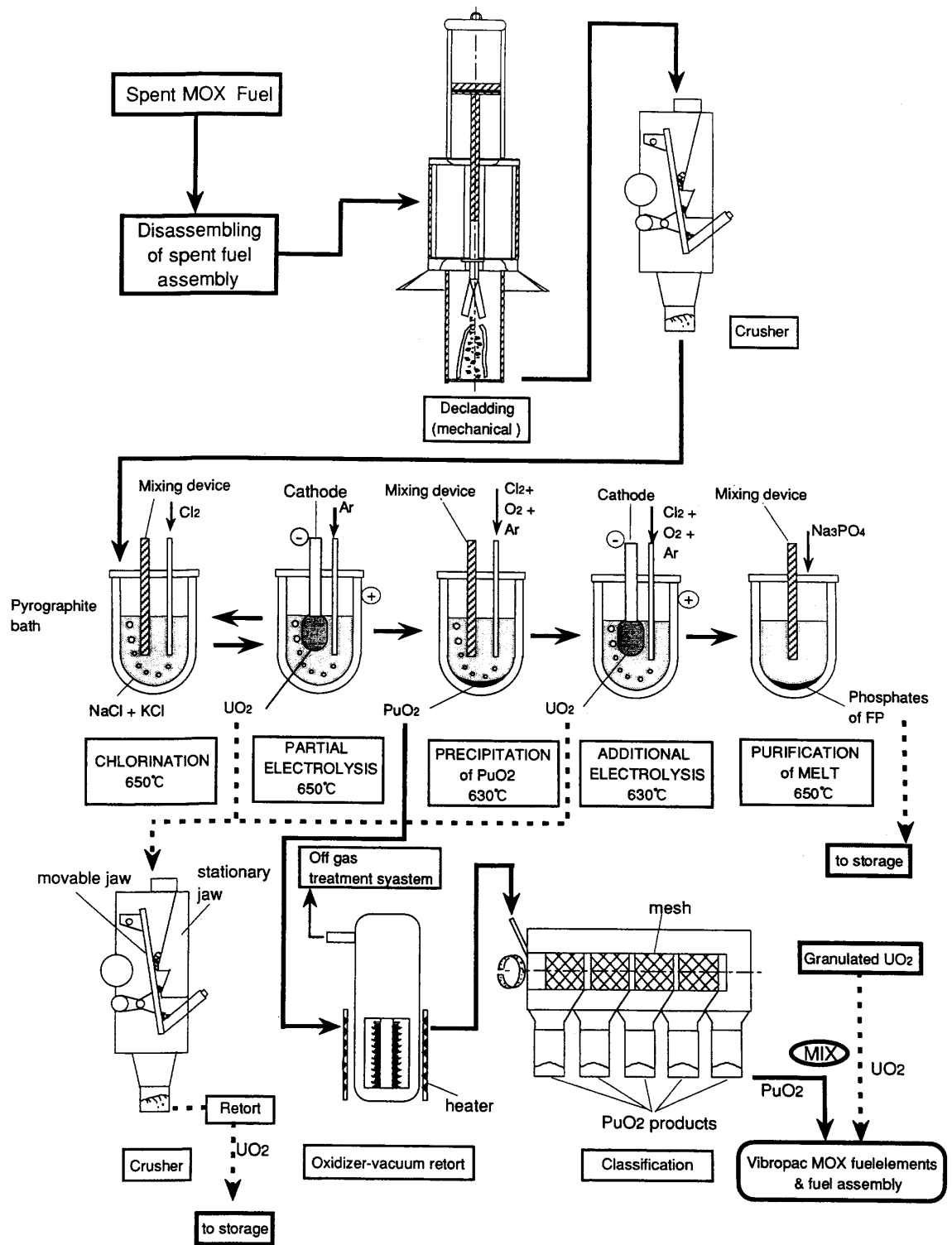
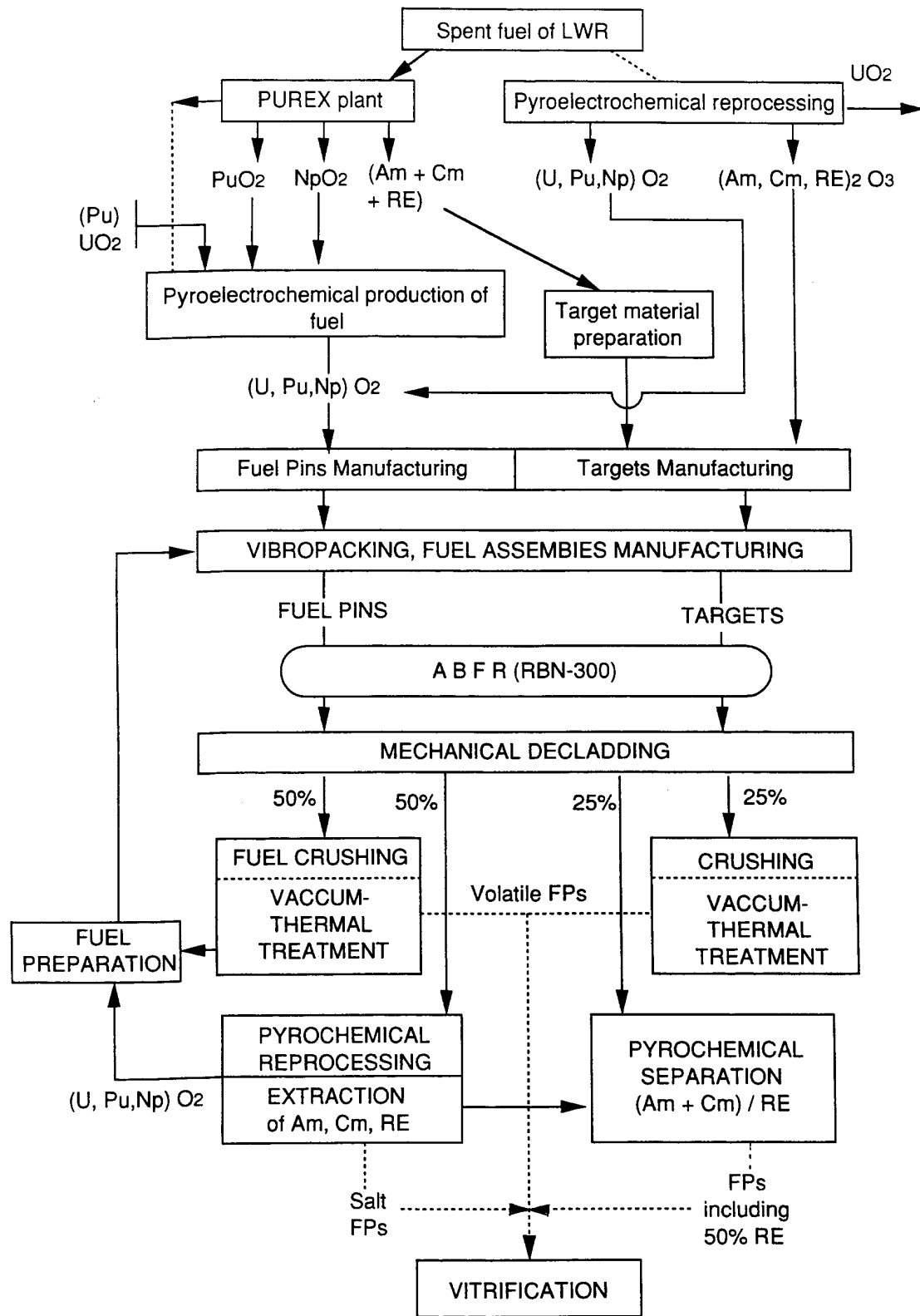
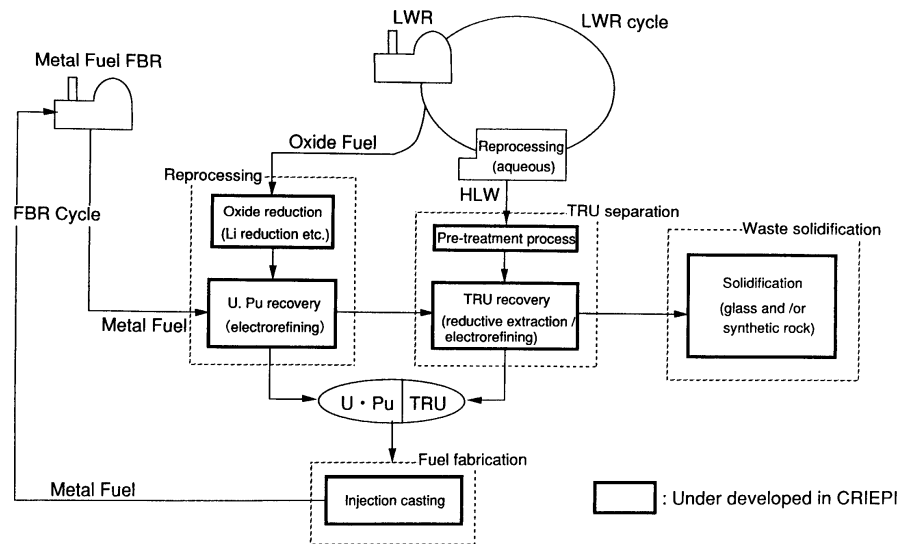


Figure II.9 Fuel cycle actinide burner reactor (DOVITA Fuel Cycle)



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



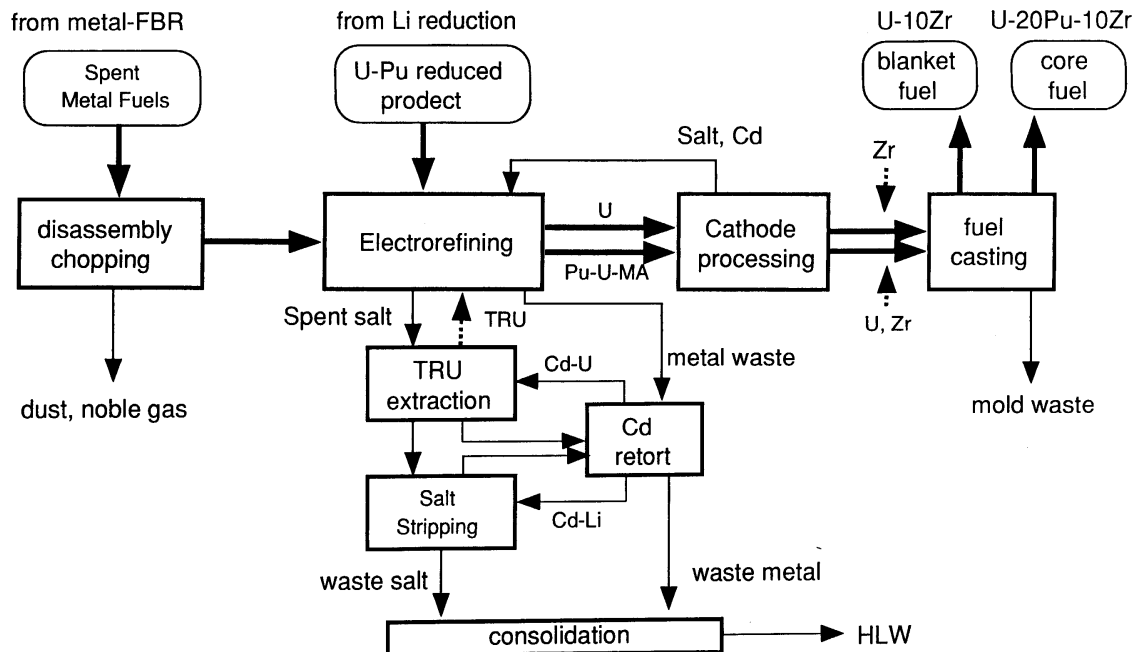
1.2.2.1 Pyrochemical reprocessing of metal and oxide fuels

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

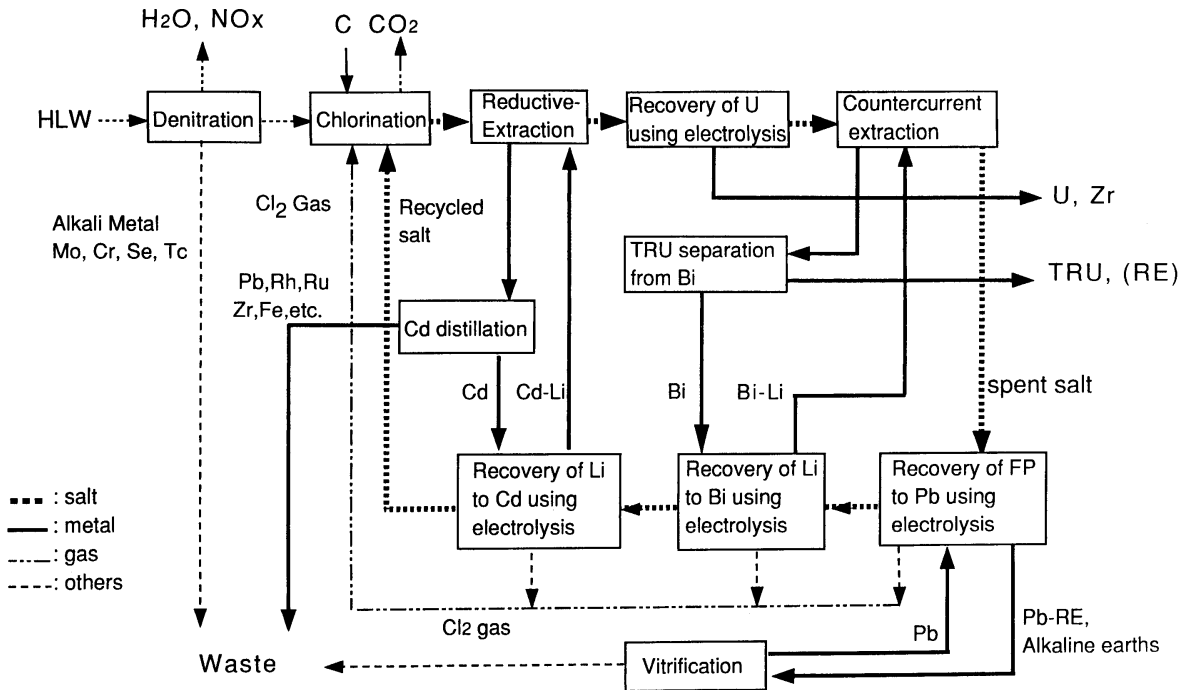
The process investigated at CRIEPI to recover transuranic elements from high-level PUREX wastes consists of denitration to oxides, chlorination, reductive extraction and electrorefining in a LiCl-KCl/Cd or LiCl-KCl/Bi system. After denitration and leaching with water to remove the soluble alkali-metal 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.

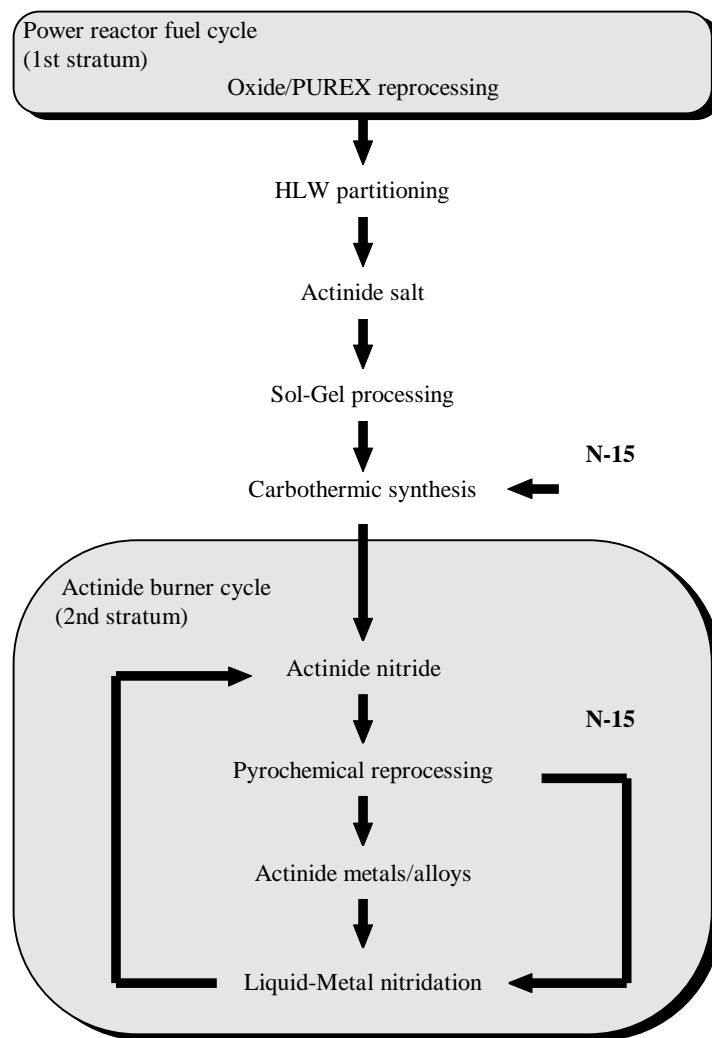
The metathetical reaction between Li_3N and actinide chlorides (AnCl_3) yields AnN and LiCl in the salt. Since the stability of actinide and lanthanide mononitrides varies relatively little from element to element, the nitrides are formed in the reverse order of stability in the corresponding halides. 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 $(\text{U}, \text{Pu})\text{O}_2$. 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 $^{236}\text{Pu} - ^{232}\text{U} - ^{208}\text{Tl}$ 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 $(\text{PuO}_2\text{-UO}_2)$ 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 $(\text{Pu}+\text{Am})$ 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, PuO₂ powders from the reprocessors are taken from storage. Subsequently, these powders are introduced into a glove box to be milled and blended together with UO₂ 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 PuO₂ 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 μ Sv/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 ²³⁸Pu by (α ,n) reactions. This favourable result is caused by the dilution of the plutonium from the MOX fuel with the plutonium from UO₂ 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)O₂ 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 AmO₂ 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 $^{14}\text{N}(\text{n,p})^{14}\text{C}$ 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 $\text{N}_2\text{-H}_2$ 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 AmO₂ 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: Am₂O₃, AmO₂, AmN and Am₂C₃ 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, Y₂O₃, Al₂O₃, MgAl₂O₄, and Y₃Al₅O₁₂ 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. MgAl₂O₄-AmO_{2-x} pellets containing 10% ²⁴¹Am by weight have been fabricated by the impregnation method by ITU. A reaction between the Am oxide and MgAl₂O₄ 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)O₂, (M, Am)₂O₃ type solid-solution in MgAl₂O₄ [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 PuO₂ and for AmO₂ powders. The γ -dose rate due to AmO₂ increases by a factor of 185 compared to PuO₂, 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 AmO₂ 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:

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

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

Both isotopes ²⁴²Cm and ²⁴⁴Cm are intense neutron sources. In case of a refabrication of fuel or targets five years after core discharge, ²⁴²Cm has mostly decayed while ²⁴⁴Cm 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 , $^{242\text{m}}\text{Am}$ 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		
	$\overline{\sigma}_f(b)^{a)}$	$\overline{\sigma}_c(b)^{a)}$	$\alpha^{b)}$	$\overline{\sigma}_f(b)^{a)}$	$\overline{\sigma}_c(b)^{a)}$	$\alpha^{b)}$
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) $\alpha = \overline{\sigma}_c / \overline{\sigma}_f$.

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 ²³⁵U) 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 $f = 10^{14} \text{ n/cm}^2 \cdot \text{s}$	Fast spectrum $f = 10^{15} \text{ n/cm}^2 \cdot \text{s}$
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 UO₂ 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 UO₂-fuelled PWRs

The Np and Am recycling was studied in a N4 type PWR, rated at 1 470 MWe, with UO₂ fuel enriched at 4% ²³⁵U, 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 (²³⁷Np or Am) are mixed with the UO₂ 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 ²³⁸U). 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 UO₂-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

* ²³⁵U 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 UO₂ fuel, and this requires a minimum decontamination factor of 640, which remains a quite high value.

Heterogeneous recycling of minor actinides in UO₂-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 and plutonium 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 UO₂ fuel irradiated at 33 GWd/tHM.

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

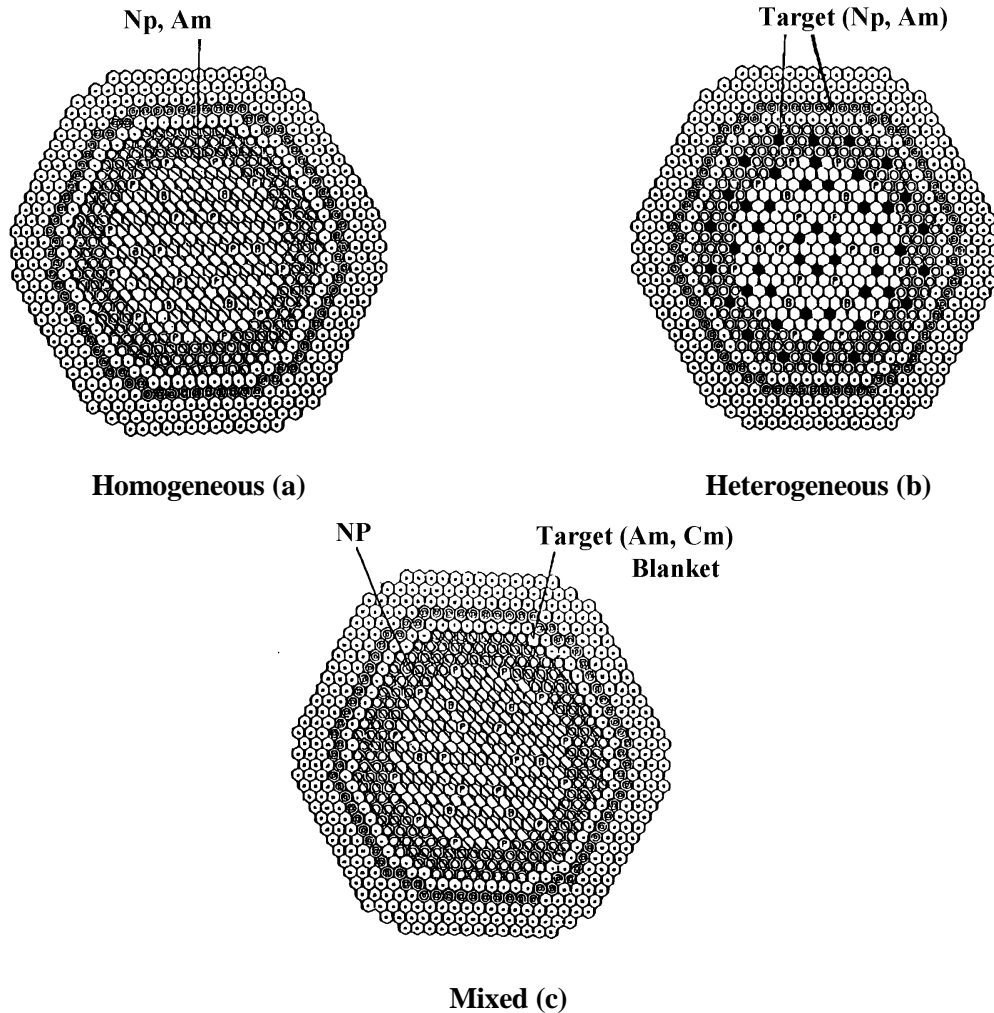
Multiple recycling of TRU components from reprocessed spent LWR-MOX fuel was investigated to assess the incineration capabilities of a FBuR [81].

Effects on core reactivity

These effects are roughly equivalent for americium and neptunium. The introduction of americium and neptunium instead of ²³⁸U 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 ²³⁷Np and ²⁴¹Am into more fissile isotopes. The effect is more marked for ²⁴¹Am because of its partial transformation into highly fissile ^{242m}Am.

Figure II.14 MA-loading methods in fast reactor



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 UO₂-fuelled PWR.

Recycling ²³⁷Np 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 ²³³Pa in equilibrium with ²³⁷Np (²³³Pa 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 ²³⁹Pu via capture by ²³⁸U. 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 $\text{DF} > 15$, the effect of lanthanides is compensated by the influence of the minor actinides.

The increase in the void effect ($\Delta\rho_{\text{void}}$) is +2.4% per vol.% of minor actinides and +0.89% per vol.% of RE in the core. The reduction of the Doppler effect ($\Delta\rho_{\text{dopp}}$) 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 ₂ O ₃	Al ₂ O 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%MAAs enriched	5% MAAs enriched
MAAs/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 (% $\Delta k/k$)	2.76	1.24	1.09	0.33
Internal conversion ratio	0.85	0.89	0.89	0.9
β_{eff} ($\times 10^{-3}$)	3.65	3.47	3.39	3.24
Void reactivity (% $\Delta k/k$)	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 ($\epsilon/\%$ -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.

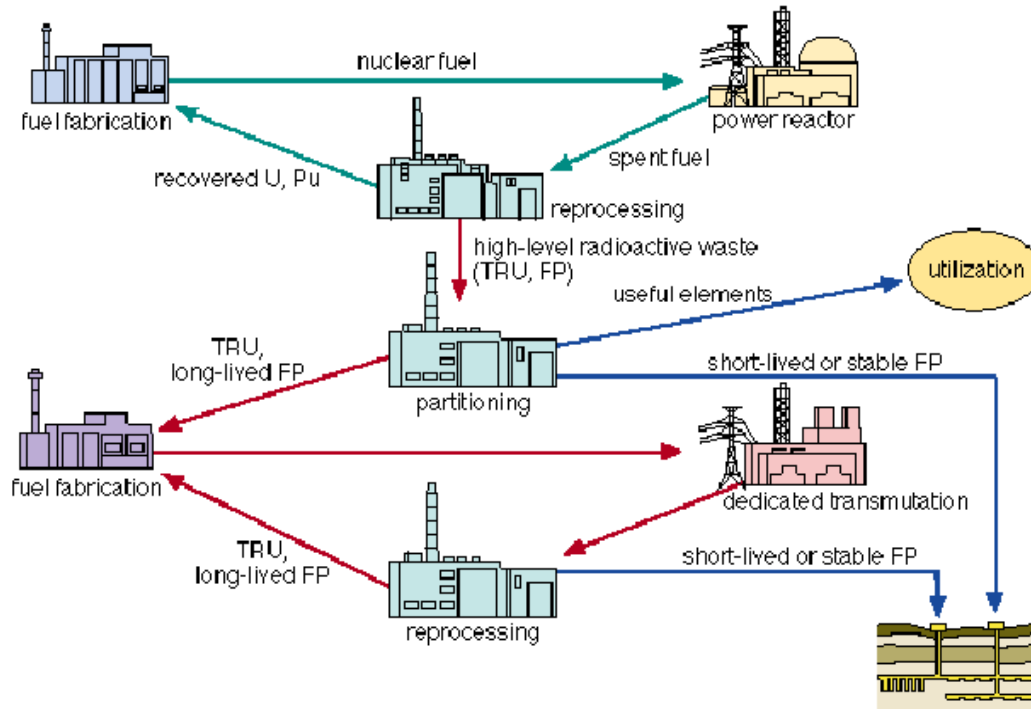
The first stratum is devoted to the electricity production and consists of standard power reactors (LWR-UO₂, LWR-MOX and FR), fuel fabrication plants and reprocessing plants. The U fuel irradiated in LWRs is reprocessed and the recovered Pu is recycled in LWR-MOX and in fast reactors. HLW from the reprocessing goes to the second stratum.

In the second stratum, MAAs (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 MAAs 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 material	pin-bundle (⁶⁴ NpAmCm- ³⁶ U ³⁾ _{1.0} N ⁴⁾ _{1.0}	coated-particle (⁶⁵ NpAmCm- ³⁵ U 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 (% Δk/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) ¹⁵N enriched.

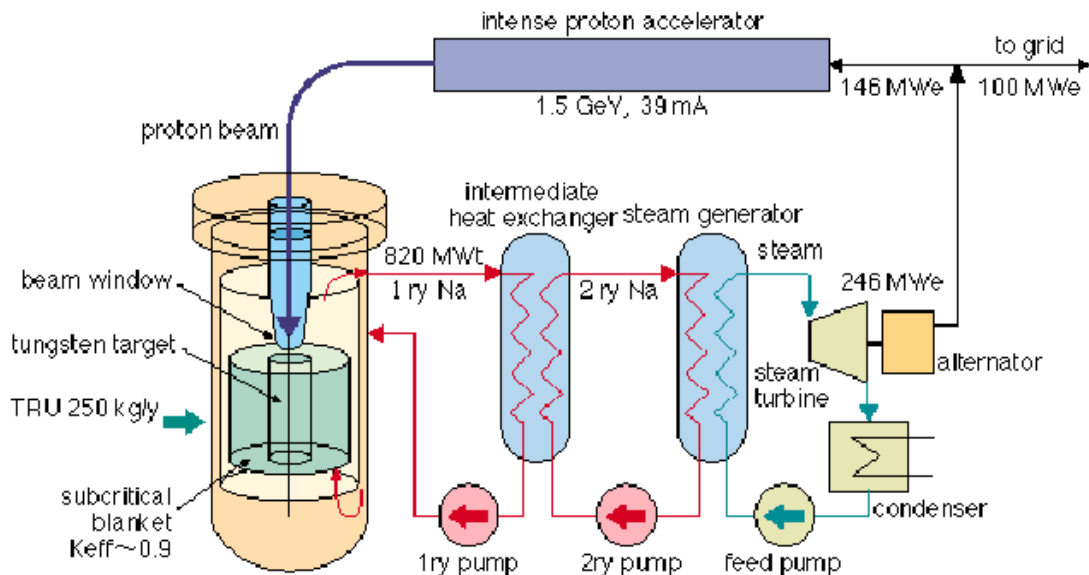
2.3.4.2 Accelerator-driven transmutation systems

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 $\pm 2\%$ accuracy on the system's energy balance is required, the spallation neutron yield should be calculated with an accuracy of $\pm 2\%$. 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 MAO₂ or PuO₂ 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 ⁹⁹Tc and ¹²⁹I and plutonium produced in 19 LWRs. ¹²⁹I 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 ($k_{\text{eff}}=0.95$) and EOC ($k_{\text{eff}}=0.80$) 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 MAO₂ or PuO₂ 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 (⁹⁹Tc and ¹²⁹I) 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 (% $\Delta k/k$)	+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/(cm²·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 (Istituto Nazionale di Fisica Nucleare) set up a basic R&D programme TASC0 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 , $^{242\text{m}}\text{Am}$ and ^{243}Am have been measured relative to that for $^{235}\text{U}(n,f)$ 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)	FBR 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 UO₂ fuel. One observes, for example, that ¹³⁵Cs, a long-lived isotope, accounts for only 10% of the mass of the chemical element caesium. Irradiation of the caesium would thus produce ¹³⁵Cs from the isotopes 133 and 134, which would severely compromise the desired objective.

Table II.14 **Production of long-lived fission products (PWR UO₂ 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 ⁹⁹Tc and ¹²⁹I

In practice, ⁹⁹Tc and ¹²⁹I are the main FPs to be considered as candidates for transmutation in present reactors: only ⁹⁹Tc 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 transmuted 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/(cm²·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 UO₂ 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 R1 scenario covers the period from 2000 to 2100. The reactor population consists of PWRs supplied with UO₂ fuel at 4% ²³⁵U 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 UO₂ 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 RP1-2 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.

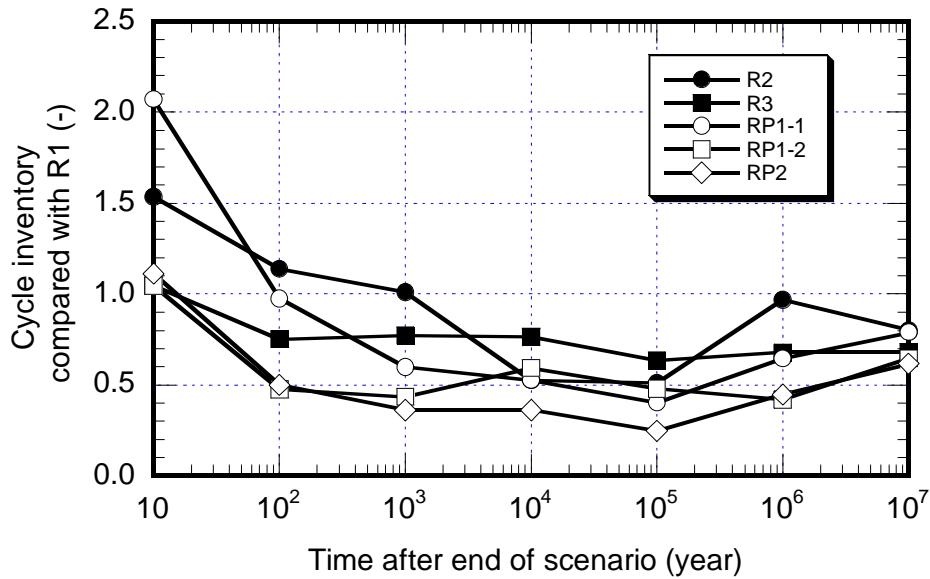
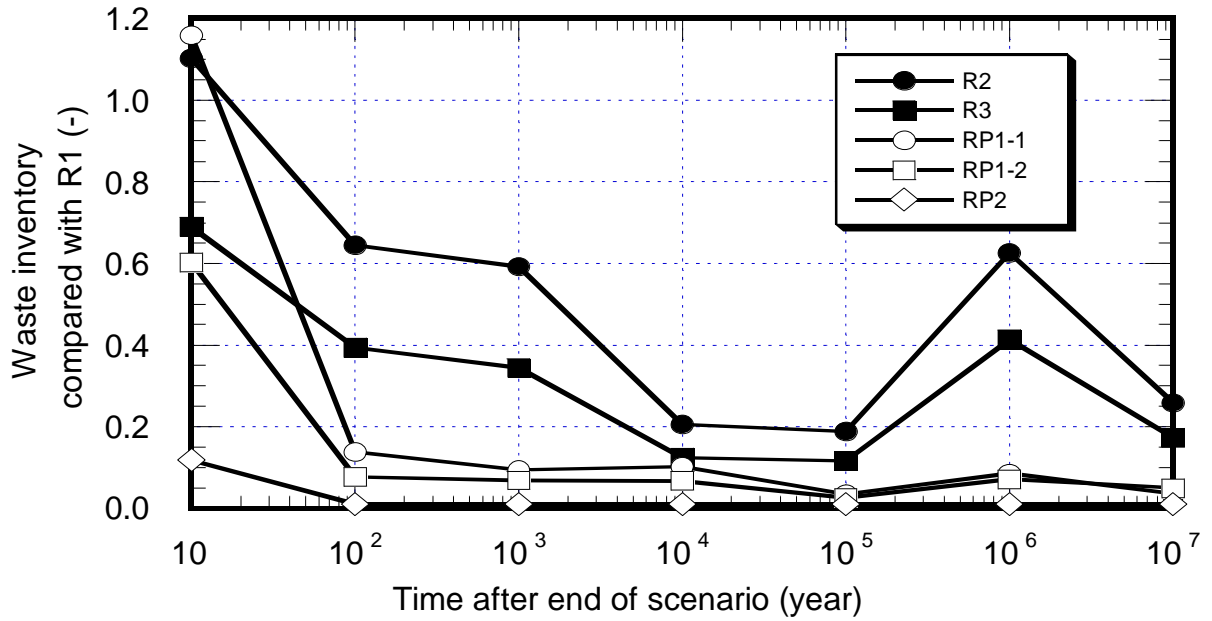
The reductions in radiotoxicity for the RP1-1 and RP1-2 scenarios are nearly the same as in the R2 and R3 scenarios respectively. There is an improvement by a factor of 6 between 10^2 and 10^3 years because of the decrease in Am inventory and similarly between 5×10^5 and 5×10^6 years due to the smaller 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 ²⁴¹Am is generated during spent fuel cooling by the β -decay of ²⁴¹Pu. 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 UO₂-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.

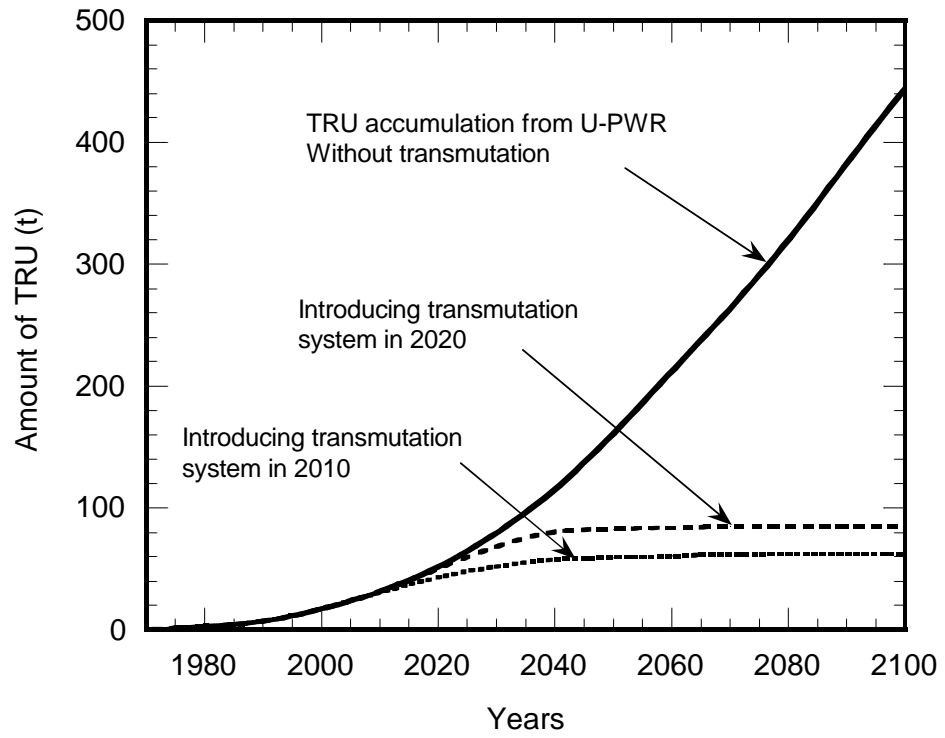
The calculated results show that the conditions can be satisfied by introducing realistic transmutation systems at the rate shown in Figure II.18.

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 ($1 \text{ Gy} = 1 \text{ J/kg}$).

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:

$$\begin{aligned} RR (time^{-1}) = & [probability\ of\ a\ detriment\ per\ unit\ dose(dose^{-1})] \\ & \times \mathbf{S} [probability\ of\ occurrence\ of\ an\ event\ i\ (time^{-1})] \\ & \times [dose\ delivered\ by\ event\ i\ (dose)] \end{aligned}$$

Hence:

$$RR (time^{-1}) = p (dose^{-1}) \times \mathbf{S} P_i \times (dose)_i$$

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:

$$RR = \sum (dose)_i P_i$$

For stochastic effects, in case of chronic low exposure, the doses are in Sv and $p = 0.063 \text{ Sv}^{-1}$ 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:

$$RR (\text{year}^{-1}) = p (\text{Sv}^{-1}) \sum P_i (\text{year}^{-1}) E_i (\text{Sv})$$

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-UO₂, 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.

$$^{238,239,240,241,242}\text{Pu} > ^{241,243}\text{Am} > ^{242,244,245,246}\text{Cm} \gg ^{237}\text{Np}$$

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 (⁹⁹Tc, ⁹³Zr and ¹³⁵Cs) and constitute a radiotoxic inventory which is roughly 1 000 times smaller than that of the actinides. ¹²⁹I 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 ¹²⁹I 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 ¹³⁵Cs, ¹²⁹I and in some cases ⁹⁹Tc, 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)

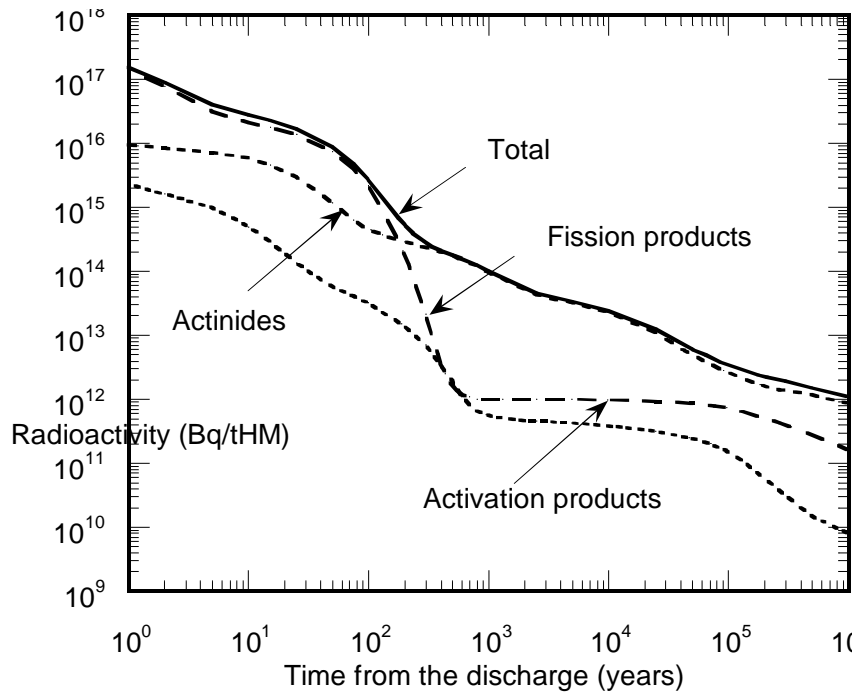
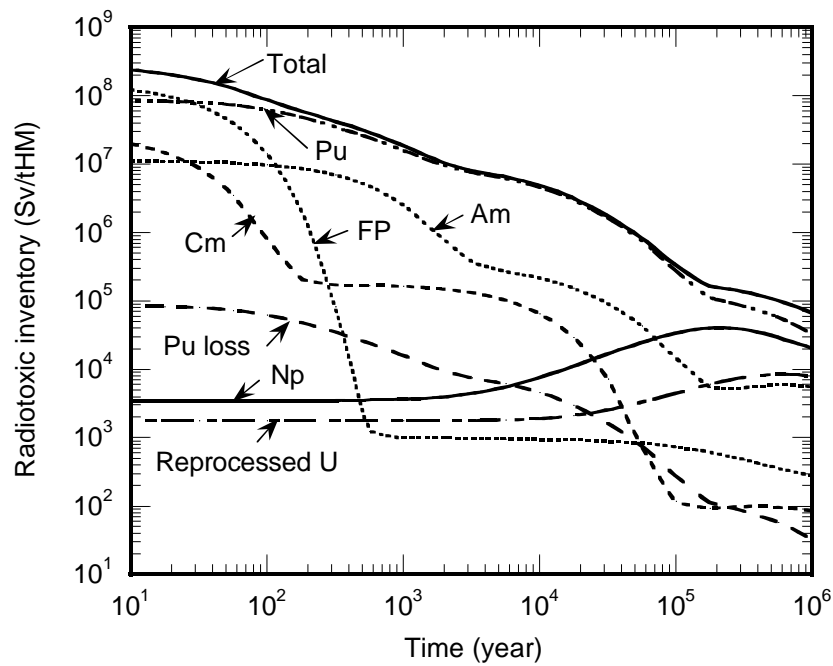


Figure II.20 **Radiotoxic inventory of UOX fuel as a function of time (3.7% ^{235}U , 45 GWd/tHM)**



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 of 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)

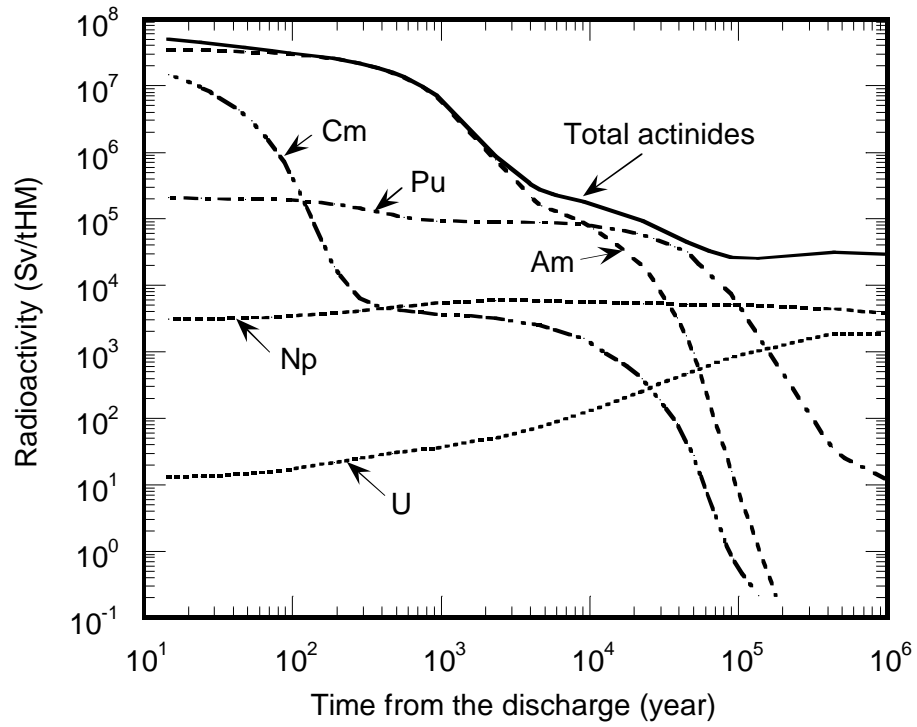


Figure II.22 Potential radioactivity of main fission products in the glasses from the standard reprocessing of PWR type spent fuel (4.1% ^{235}U , 40 GWd/tHM)

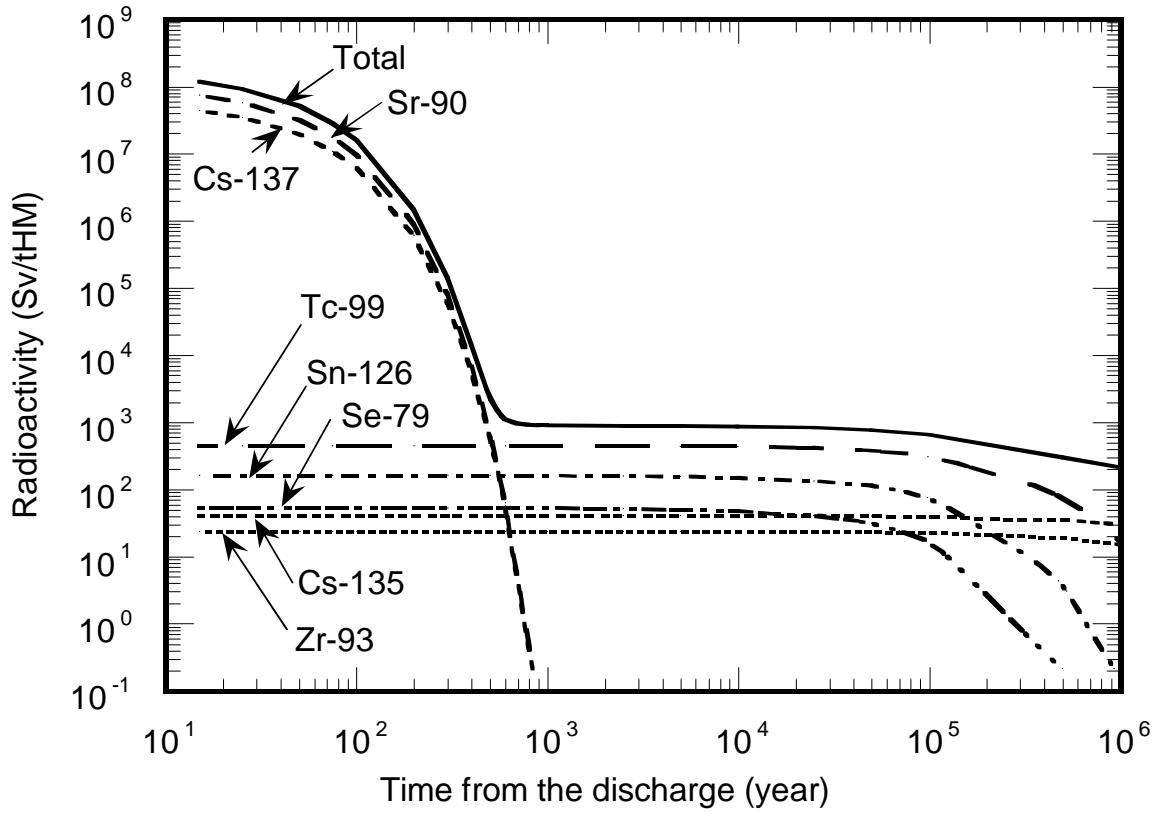
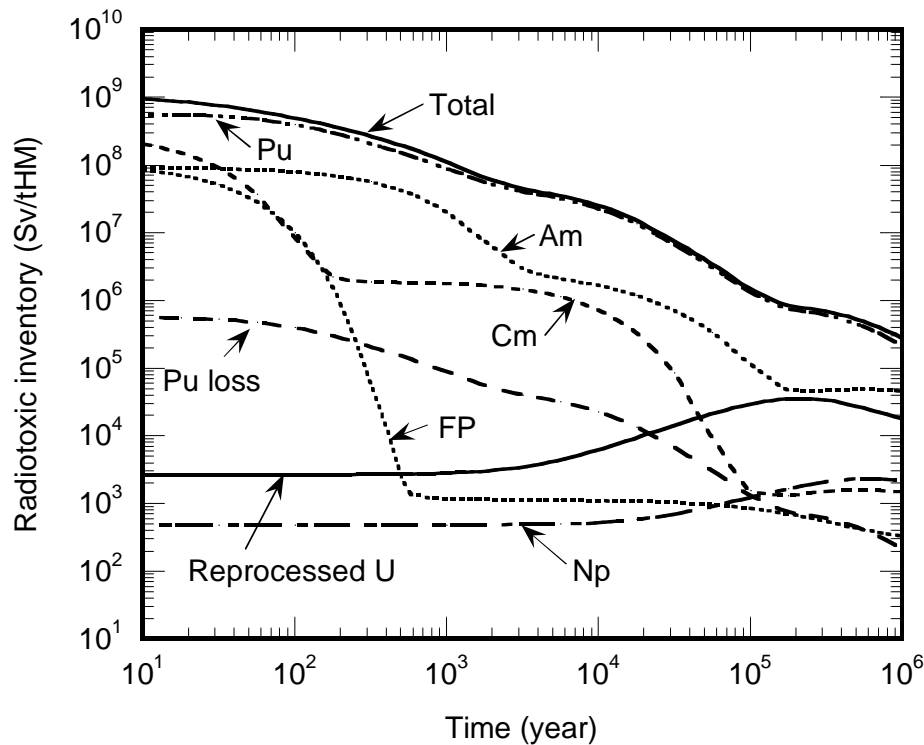


Figure II.23 Radiotoxic inventory of MOX fuel as a function of time (8.28% Pu, 45 GWd/tHM)



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]

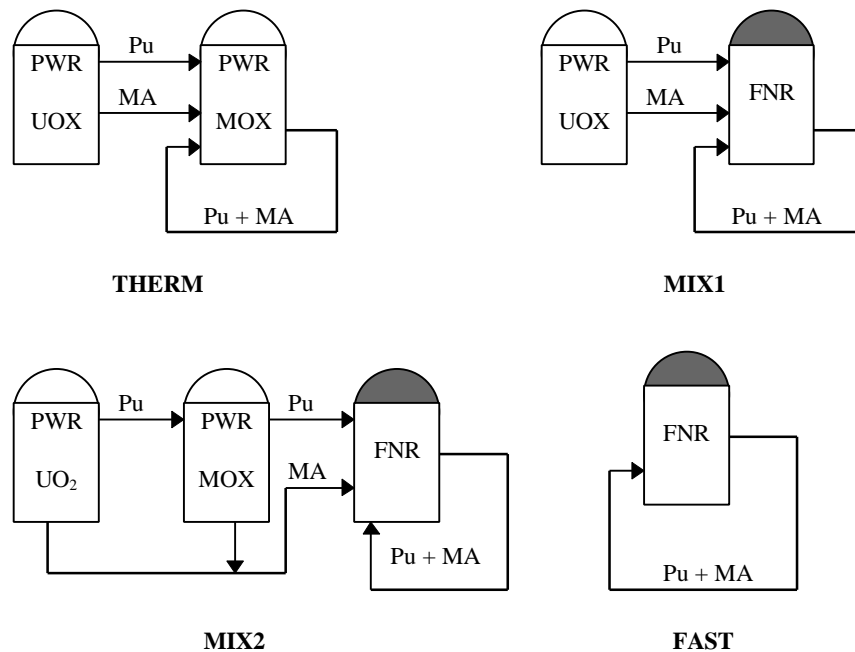
Research conducted in recent years has helped to clarify the feasibility of recycling actinides (mainly Am and Np) in the different types of reactor (FBR, PWR) in different recycle modes (homogeneous or heterogeneous).

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_a/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** 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 UO₂ 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 TWh/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 UO₂ 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 UO₂	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 UO₂ 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)**

	UO₂	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

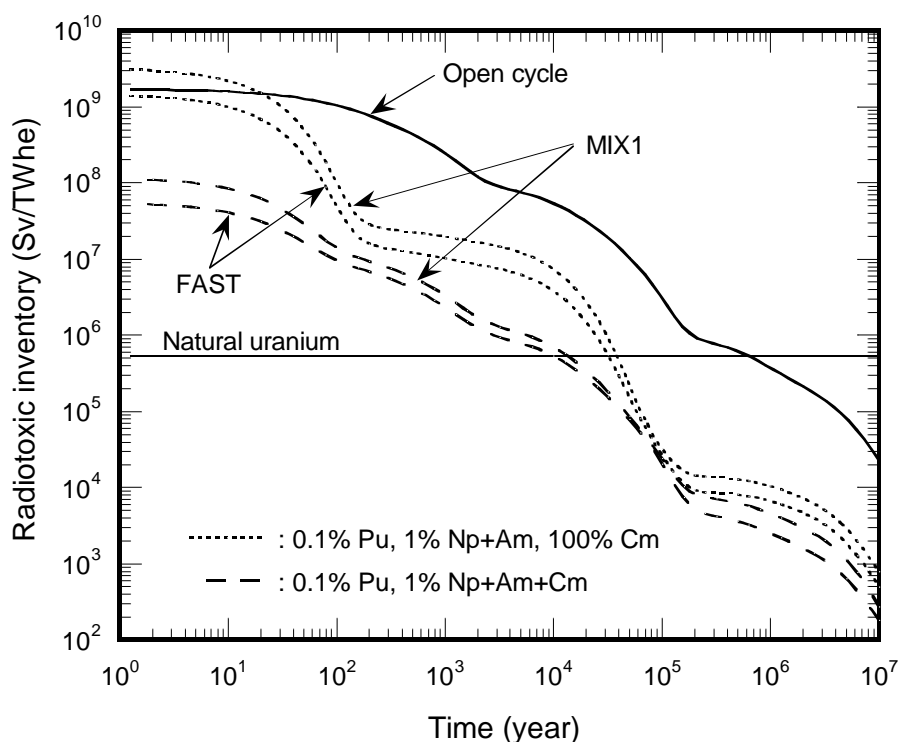
The comparison of the data of Table II.19, with the curium recycling option, to those of Table II.21 with curium discharge in the waste streams, shows that the radiotoxic inventory reduction factor is decreased from the range of 70-150 down to 7-115 depending on the time interval. This obviously has 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	1.12	0.00684	0.00656	0.00299
²³⁹ Pu	14.1	0.0329	0.0159	0.0857
²⁴⁰ Pu	6.44	0.0462	0.0372	0.0573
²⁴¹ Pu	3.44	0.00903	0.00706	0.00713
²⁴² Pu	2.54	0.0139	0.0140	0.00536
Total Pu	27.7	0.109	0.0807	0.159
²⁴¹ Am	1.09	0.0508	0.0520	0.0330
^{242m} Am	0.00271	6.0×10 ⁻⁴	0.00285	0.00188
²⁴³ Am	0.550	0.0553	0.0588	0.0158
Total Am	1.65	0.107	0.114	0.0507
²⁴² Cm	2.63×10 ⁻⁵	2.89×10 ⁻⁵	2.39×10 ⁻⁵	1.85×10 ⁻⁵
²⁴³ Cm	0.00325	0.00220	0.00174	0.00159
²⁴⁴ Cm	0.220	0.0557	0.0535	0.0236
²⁴⁵ Cm	0.0198	0.00282	0.00271	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-UO₂, 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 ($^{240}\text{Pu}/^{245}\text{Cm}$).

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 $1\,300 \pm 300$ 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

Year	La Hague			Sellafield		
	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

Year	La Hague			Sellafield			Tokai
	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 ¹³⁷Cs and ⁹⁰Sr 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 ²⁴¹Am and ²⁴⁴Cm 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 ²³⁷Np 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 ²⁴¹Am which decays to ²³⁷Np. 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 UO₂ 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-UO₂ 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-UO₂ 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 ²³⁸Pu, ²⁴⁰Pu and ²⁴²Pu) 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 (²³⁸Pu, ²⁴¹Am and ²⁴⁴Cm) 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-UO₂ 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-UO₂ 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-UO₂ NPP park of 100 GWe producing 700 TWh/year discharges annually 1 760 tHM LWR-UO₂ 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-UO₂. The TRU content of spent LWR-MOX is 42.3 kg/tHM compared with

11.4 kg/tHM for UO₂ 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 ²⁴¹Pu.

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 UO₂ 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 UO₂ fuel) owing mainly to ²⁴⁴Cm, ²⁴¹Am and ²³⁸Pu. The radiotoxic inventory will decrease with the half-life of ²⁴⁴Cm (18 years) and then with the half life of ²³⁸Pu (87.7 years). The total α -activity per tHM would decrease to 5 217 TBq α 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 ¹³⁷Cs ($t_{1/2} = 30$ years) and ⁹⁰Sr ($t_{1/2} = 28$ years), their disappearance by natural decay takes about 300 years. Partitioning of ¹³⁷Cs and ⁹⁰Sr 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 m² is required (including 200 000 m² 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

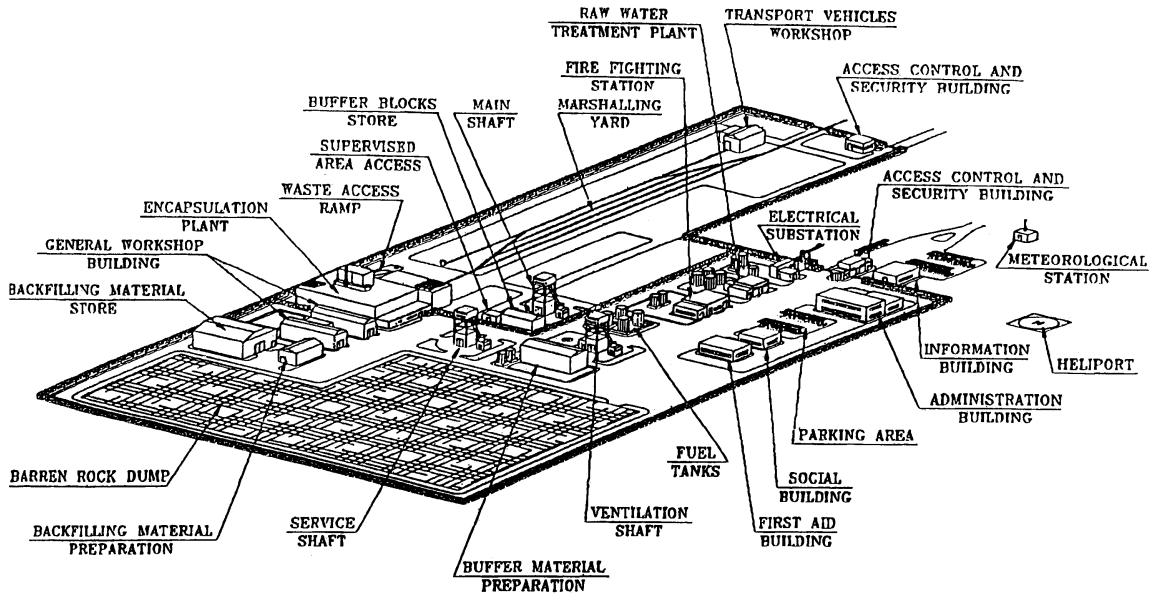
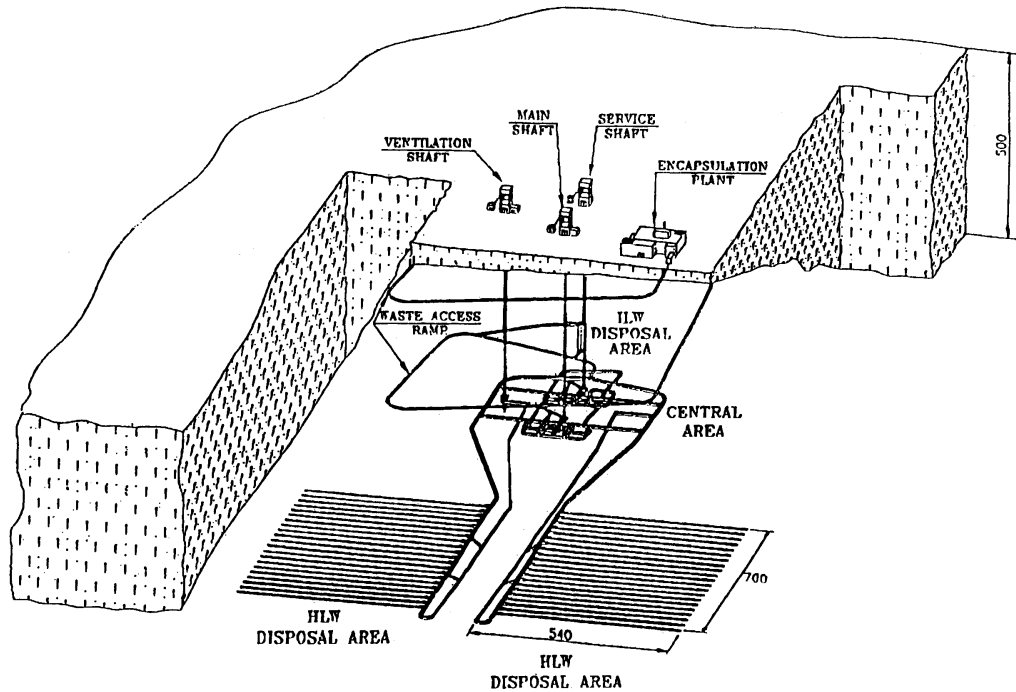


Figure II.27 Underground facilities. General layout



The repository area needed is 2.58 km² for clay, 4.1 km² for granite and 1.2 km² for salt, and the total excavated rock volume amounts to about 765 000 m³ for clay, 569 000 m³ for granite and 579 000 m³ 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 m² 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.

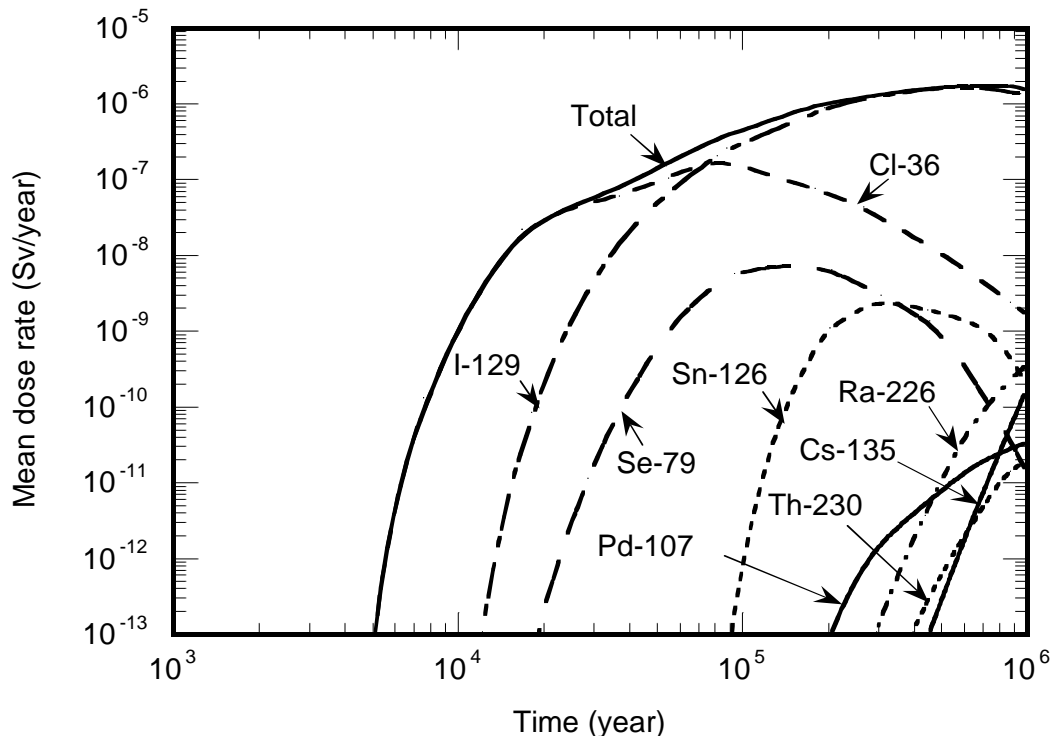
Once the emplacement activities are finished, all open space inside the repository mine must be backfilled and sealed. Different backfilling materials will be used (in-situ compacted reconstituted clay for the clay option, in-situ compacted mixture of bentonite and sand for the granite option, and in-situ 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 μ Sv/year, well below the regulatory limit of 100 μ Sv/year, is reached 600 000 years after waste disposal, with ¹²⁹I 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.

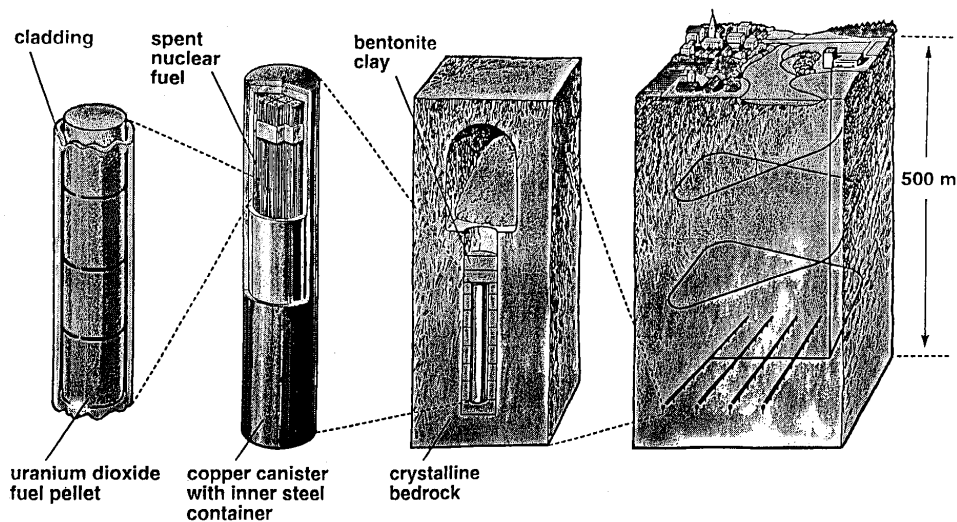
Deep repository

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 km².

Figure II.29 Swedish repository concept

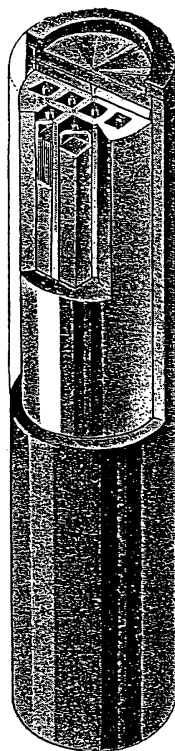


Encapsulation of spent nuclear fuel

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



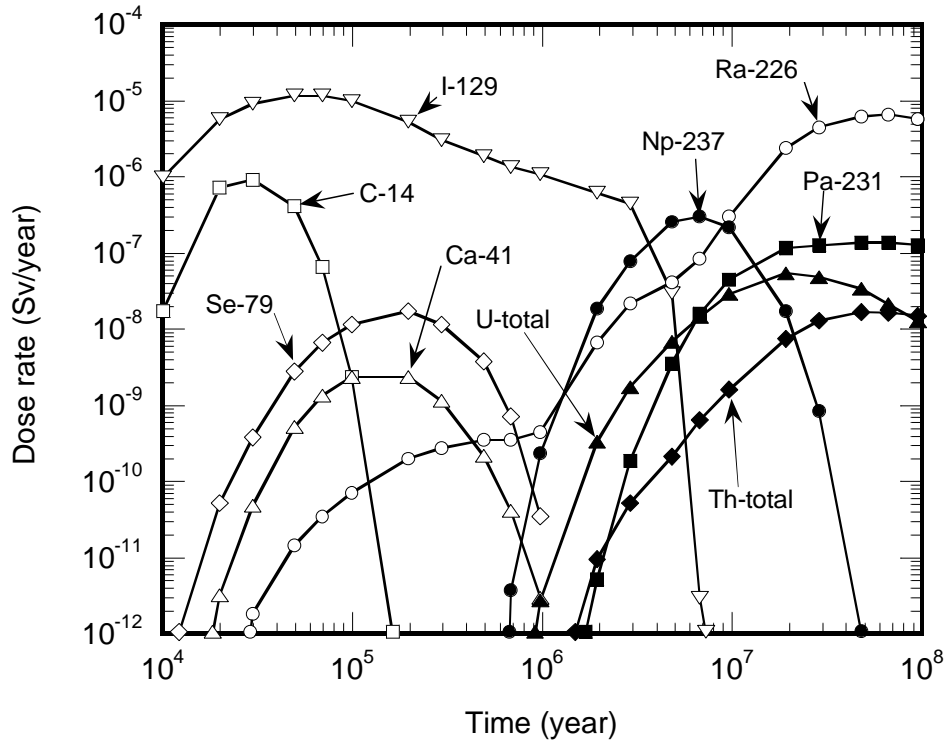
The steel insert is cast with thick steel walls between fuel assemblies. This gives a good mechanical stability besides providing adequate protection against criticality in the unlikely event that the canister at some unspecified future time should be filled with water.

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 $\mu\text{Sv}/\text{year}$ 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 $\mu\text{Sv}/\text{year}$. 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

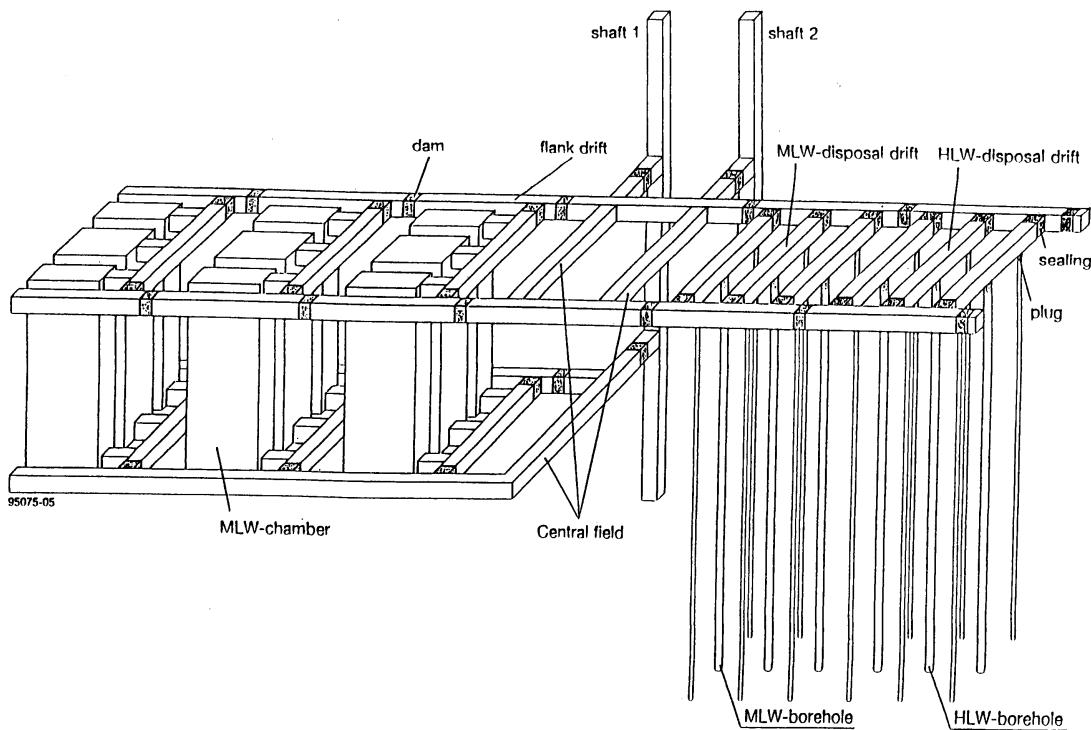
In the case of the RFC, attention in waste management and disposal is focused on the conditioning of the High Level Waste (HLW) and Medium Level Waste (MLW) forms produced during the reprocessing operations and their disposal in geological formations. Two case studies are briefly described in this report: 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 $14 \times 4 \text{ km}^2$. 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 $7 \times 10^{-16} \text{ m}^2$. 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 ($10\,454 \text{ m}^3$) corresponding to a nominal initial quantity of 73 000 tHM spent fuel will be disposed of. Feed sludges and cladding wastes totalling $80\,000 \text{ m}^3$ 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.

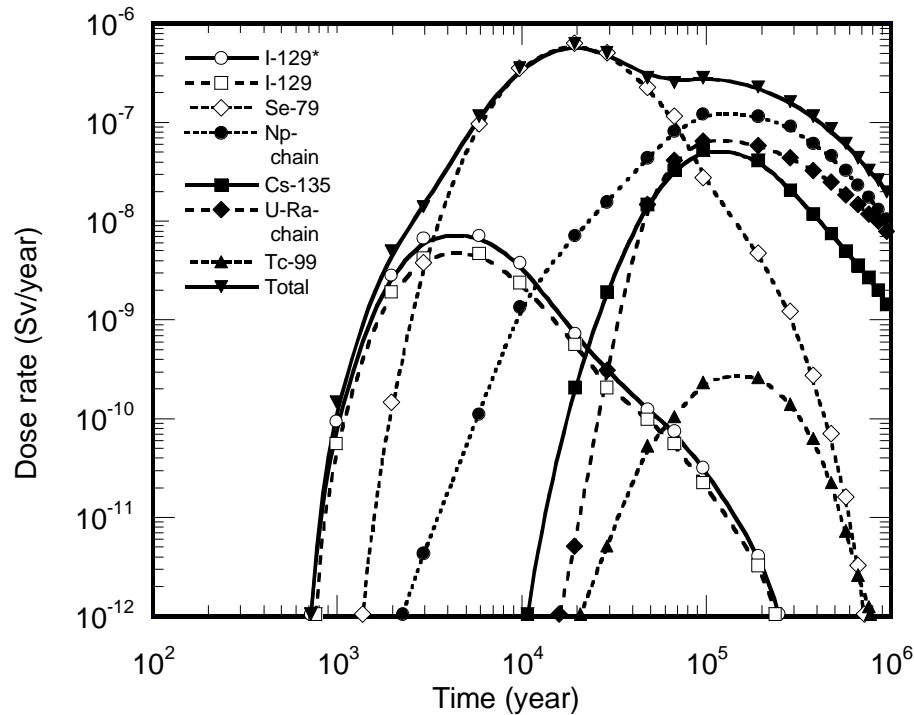
Figure II.32 Reference repository in salt formations in the Gorleben salt dome



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 mSv/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 mSv/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 500~1000 years after closure (cavern-convection scenario), the peak annual dose is one order of magnitude higher (1 nSv/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.

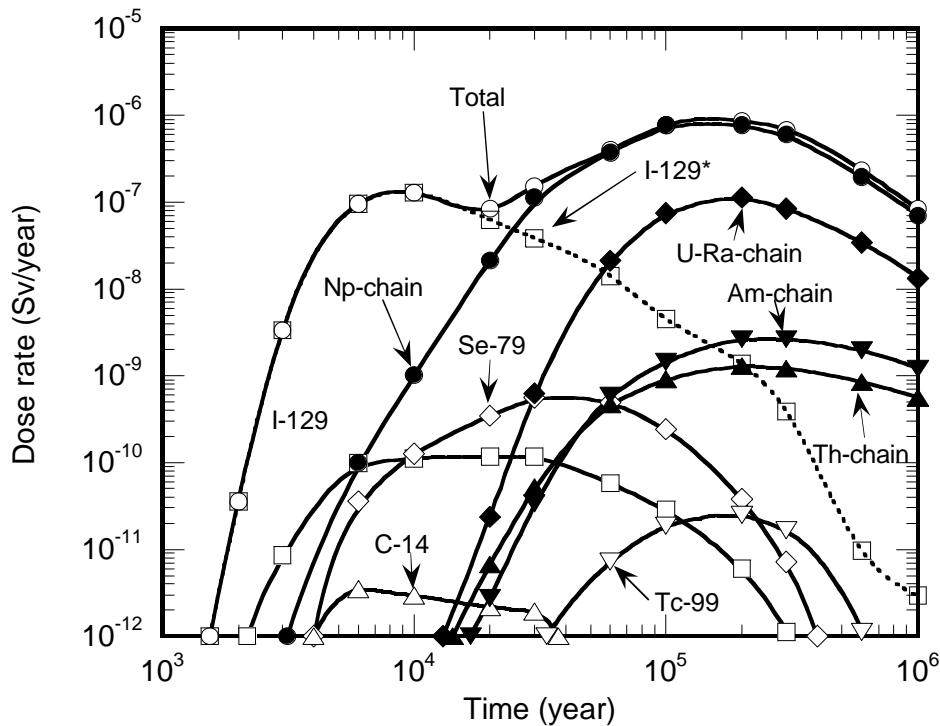
In the uplift scenario the doses depend essentially upon the uplift rate. The maximum dose rate ranges from 0.6 nSv/year for an uplift rate of $3.3 \times 10^{-5} \text{ m/year}$ to 100 nSv/year at an uplift rate of $2 \times 10^{-4} \text{ m/year}$. However, the time of arrival of the peak concentrations 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 μm . 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 m³ 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.

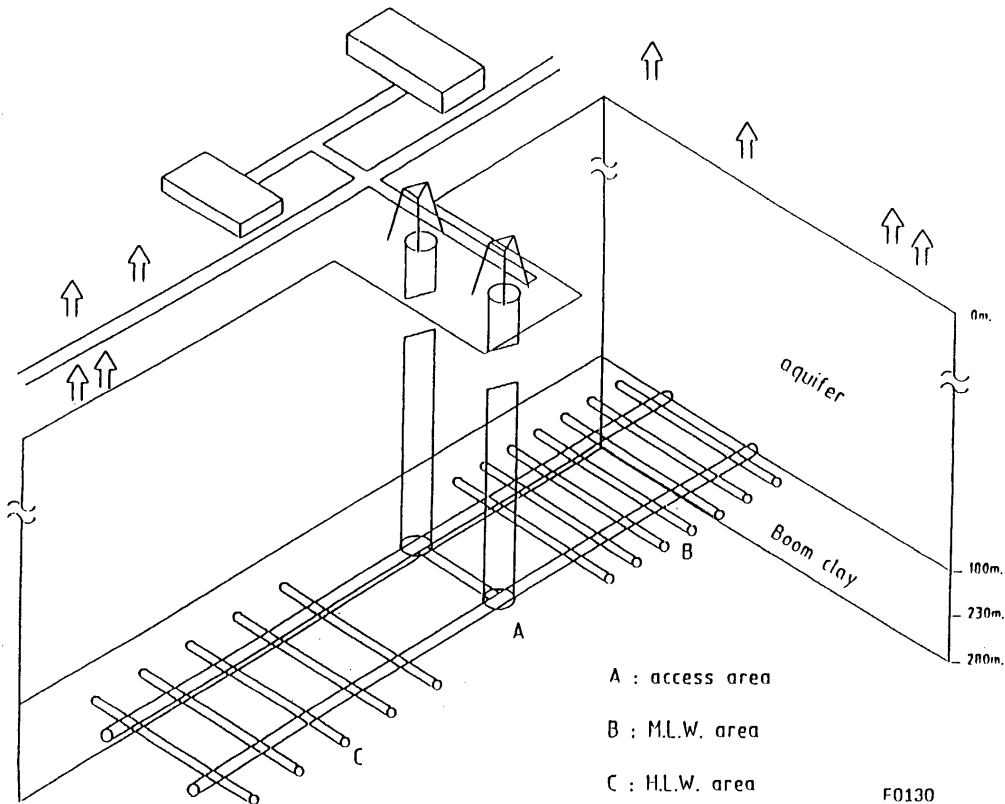
The actinides Am and Cm and the daughter product ^{240}Pu decay completely within the first 10 m of the clay layer. This is not the case with ^{237}Np or with uranium if it were disposed of. The maximum annual dose due to the presence of ^{237}Np is about 0.02 $\mu\text{Sv}/\text{year}$ when the use 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 $\mu\text{Sv}/\text{year}$ and 0.15 $\mu\text{Sv}/\text{year}$, 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 $\mu\text{Sv}/\text{year}$. 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

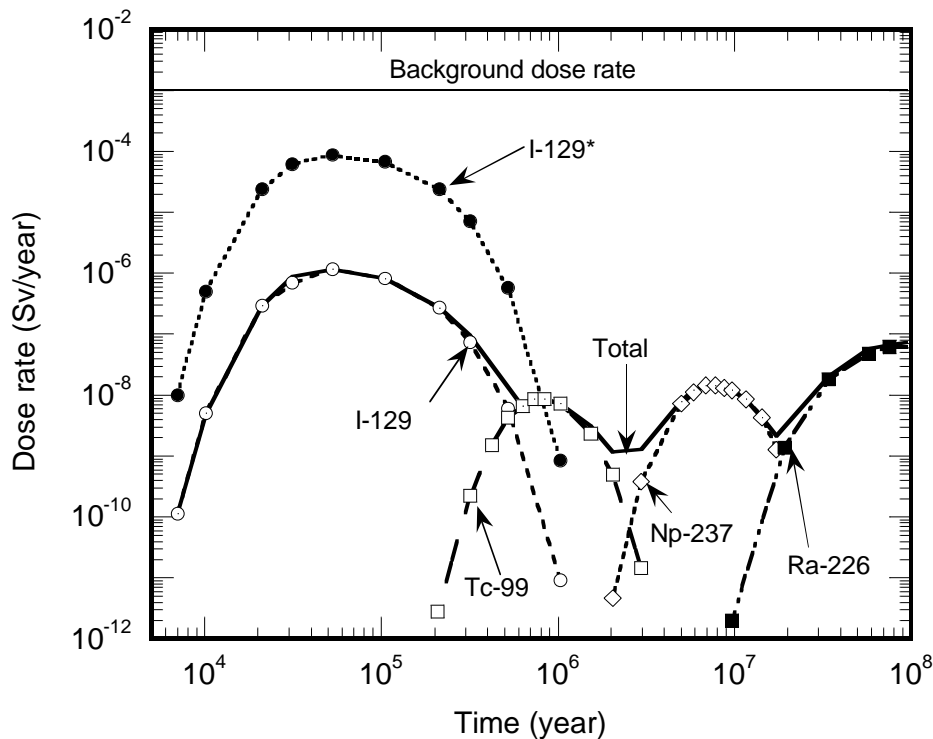
4.5.3 AFC-specific waste management issues (targets, reactor cores)

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 ($^{129}\text{I}^*$: 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 ($<5 \times 10^{-4}$);
- 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 , ^3H 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 $^{137}\text{Cs} - ^{90}\text{Sr}$ 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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