

Application of Partitioning/Transmutation of Radioactive Materials in Radioactive Waste Management

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1 Introduction

The worldwide electro-nuclear capacity amounts to about 350 GWe and is subdivided into three blocks of each 100 to 120 GWe: the US, Europe and the rest of the world. For strategic evaluations a block of 100 GWe is therefore a representative portion of the electro-nuclear output on world scale. Two fuel cycle options have presently reached industrial maturity: the "Once Through Fuel Cycle" (OTC) and the "Reprocessing fuel cycle"(RFC) with recycling of plutonium and some uranium. Worldwide some 10,500 tHM spent fuel are discharged annually from the nuclear power plants (NPPs) and either stored till disposal or reprocessed. The industrial reprocessing capacity amounts to 3900 tHM which implies that only one third of the discharged spent fuel can be processed. Due to this situation the total cumulative inventory of stored spent fuel increased to 130,000 tHM while only 70,000 tHM has been reprocessed and transformed into High Level Waste (HLW) and spent LWR-MOX. Due to the relatively low uranium ore price this evolution is expected to continue in the next decades. Increasing amounts of repository space will become necessary to cope with these high inventories of spent fuel. On world-wide scale two repositories of the Yucca Mountain size or their equivalent are to be licensed for spent fuel, and one of the same size for HLW. The major difference between a spent fuel repository and a HLW repository is the high Plutonium inventory which has to be taken into account for its proliferation and criticality impact. When heat output and TRU nuclides are considered there is no significant difference between a spent fuel and a HLW repository since in both cases the medium-lived fission products (Cs137 and Sr90) are determining the design characteristics of an underground repository and the minor actinides (Neptunium, Americium and Curium) are determining the radiotoxicity potential.

Partitioning and Transmutation (P&T) is a complex technology which implies the availability of advanced reprocessing plants, facilities for fuel fabrication of TRUs and irradiation facilities beyond the present NPP facilities.

During the last decade Partitioning and Transmutation was thoroughly revisited by a large number of research institutes in many different countries in order to reassess its merits and to investigate new routes which could be followed to implement this alternative fuel cycle and waste management option. But the two concepts "Partitioning" and "Transmutation" are fundamentally different as well in technological background as in objective to be reached.

Partitioning is to a certain extent a broadening to other nuclides of the current reprocessing technique which has been operating at industrial level for several decades and for which the main facilities, at least in Europe and Japan, exist or can easily be extrapolated from present day nuclear plants. Partitioning is the technology which can be considered as a form of "super - reprocessing" by which,

in addition to U, Pu and Iodine (I-129) also the Minor Actinides (MA) and the Long lived Fission products (LLFP: Tc-99, Zr-93, Cs-135, Pd-107 and Se-79) would be extracted from the liquid high level waste.

Transmutation requires fully new fuel fabrication plants and irradiation technologies which are to be developed and implemented on industrial scale. The existing NPP's could in principle be used for transmutation but many practical obstacles may arise: e.g. interference with daily operation of the plants, core safety considerations and lack of transmutation or incineration yields. New irradiation facilities: dedicated Fast Reactors (FR), Accelerator driven transmutation devices and even Fusion Reactors have been proposed for transmutation and "incineration" purposes.

One of the main objectives of P&T has always been to reduce the **long term hazard** of spent fuel or High Level Waste (HLW), this hazard is associated with the radioactive source term itself. While the waste management community stressed the **longterm radiological risk**, which is a combination of potential hazard and confining properties of the geologic strata. The measures which have to be taken for hazard reduction are very different and much more fundamental than those for risk reduction.

The longterm hazard of the spent fuel and HLW is associated with the Actinides particularly the trans Uranium elements (TRUs) while the short and longterm risks are due to the mobility of Fission Products (FP) in the geosphere with possibility to enter the biosphere.

By reprocessing the spent LWR-UO₂ fuel the hazard associated with the major actinides is removed from the HLW and recycled to the fuel cycle facilities and NPPs. The resulting HLW contains essentially the FPs and the Minor Actinides (MA). Very much emphasis has been put on the extraction of MA from High Level Liquid Waste (HLLW) before vitrification, considering the vitrification as an adequate method for immobilizing FPs. However some FPs are not present in the HLLW: part of the insoluble residues (Tc, Ru and "platinum"-metals) and Iodine. High Pt elements concentration decreases the longterm glass stability.

Transmutation is, in principle, intended to reduce the longterm hazard by transforming long-lived radionuclides into short-lived or inactive elements. In some cases this approach is valid.

Transmutation is the most difficult issue to be investigated since the present LWRs are not suited for MA and LLFP transmutation. Only specially licensed LWR-reactors can cope with MOX-fuel and for increased Pu loadings (up to 100%) special reactor designs (e.g. ABB80+) are required. By using a combination of these reactor types the plutonium inventory of a reactor park could be stabilized at the expense of increased MA production.

In order to be really effective for the abatement of the longterm toxicity, the so called transmutation should be an "incineration" process by nuclear fission and therefore very hard neutron spectra are to be used. Specially designed fast critical reactors could serve as "incineration" facilities for Pu+MA concentrates but due to reactor safety considerations the MA loadings are limited.

A significant reduction of the total actinide inventories (Pu+MA) can only be achieved by combining the transmutation capacity of specially licensed LWR-MOX reactors with that of a dedicated fast reactor park including the latest generation of critical reactors of the CAPRA type for Pu consumption in the core, and MA transmutation in thermal blanket positions. The accelerator driven transmutation systems (ADS) with a fast neutron spectrum could be reserved for MA depletion. Some thermal ADS concepts with very high thermal flux may also provide a significant TRU destruction.

Transmutation of LLFP remains problematic because they occur in elemental mixtures which contain different isotopes of the same element. In order to be effective, isotopic separation of the long-lived fission nuclide would have to be realized. However their very low capture cross sections in thermal neutron fluxes reduces the transmutation yield and would need dedicated reactors with very high loadings to achieve a net transmutation yield. If specific nuclear interactions between light nucleons (neutrons, protons, mesons...) and target FP isotopes could be identified by fundamental research, this could perhaps open a new route to the reduction of the long term disposal risk.

Conclusions for waste management

In order to carry out a full P&T option throughout the global nuclear capacity of 350GWe the reprocessing capacity (aqueous and/or pyrometallurgical) will have to be upscaled from the scheduled 3900 tHM/year up to 10,500 tHM per year. The increase in HLW heat-output which determines the size of a repository will be proportional. The storage and eventual disposal of spent LWR-UO₂ will decrease while the inventory in surface facilities for spent LWR-MOX will remain unchanged. New types of waste forms: metallic, ceramic and vitro-ceramic will supplement the vitrified HLW form.

Whatever the penetration of an advanced fuel cycle in the nuclear energy production, a residual inventory of spent fuel resulting from the ultimate decommissioning of advanced FRs and ADS reactors will always have to be disposed of in geological repositories.

2 Conventional nuclear fuel cycle

2.1 The Once-through fuel cycle (OTC) with direct disposal of spent fuel (Fig 1)

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

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

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

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

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

A few specific **regulatory and safety aspects are associated with the waste management** in a OTC scenario:

- The potential for criticality has to be addressed in the licensing process of a spent fuel repository because substantial quantities of fissile material will be deposited, and Pu239 decays to U235.
- The entire Iodine-129 inventory which is the limiting nuclide in risk analysis of waste repositories will in a period of a few 10,000 years be fully

dispersed in the geosphere, unless very specific actions are taken to limit its solubility in the deep aquifers.

- Beyond 500 years heat emission of spent fuel due to the total actinide content is significantly higher than that of the fission products alone and has to be accounted for in the selection and design of a repository. However the absolute value of that long-term heat output is, significantly lower than that of the initially loaded fission products.
- In the USA, the maximum inventory of spent fuel is limited to 70 000 tHM per repository. Thus for a big country with a large number of nuclear reactors, like e.g. USA, a new repository may have to be installed about every 30 years.

2.2 Fuel cycle with aqueous reprocessing (RFC) and vitrification of HLLW (Fig 2)

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

By proceeding according to this RFC scenario the major fraction (~99.9%) of the U and Pu streams are extracted and only a very minor fraction of the so called “major actinides” are transferred to the HLLW (and consequently to the HLW) and eventually to the geologic repository.

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

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

The stock of Pu already accumulated at the reprocessing plants and which was intended to be used in LMFBRs became redundant in a cheap uranium market economy. Some years ago, the recycling in LWRs of Pu recovered in the reprocessing operations became an industrial practice. The use of LWR-MOX in a RFC option got industrial significance in Western-Europe where increasing quantities of PuO₂ were transformed into LWR-MOX fuel and irradiated in specially licensed reactors in France, Germany, Switzerland and Belgium. The reuse of Pu is to a certain extent a first step in a global P&T scenario which has to be brought into a broader perspective of reuse of resources and reduction of the long-lived waste produced during the nuclear age.

From radiotoxic point of view the overall gain is rather limited since only ~25% of the recycled Pu is consumed and about 10% is transformed into a long-term radiotoxic MA source term. Recycling of spent LWR fuel as MOX provides an overall mass reduction (a factor of about 5) of this very large radiotoxic source term, but this recycling does not significantly reduce the total radiotoxicity. Double, or perhaps at the limit, triple recycling of LWR-MOX is theoretically possible in LWRs if fresh plutonium is available but the resulting radiotoxicity drastically increases throughout the subsequent recycling campaigns.

If it were required to further reduce the global radiotoxic inventory it would be necessary to transfer the TRU content of spent LWR-MOX fuel into a FR-MOX fuel cycle scenario which becomes a part of the next-generation scenario: the advanced fuel cycle (AFC). (**Figs 3 and 4**) In the meantime, retrievable storage of spent LWR-MOX fuel is the most appropriate fuel cycle option until a fast neutron “incinerator” technology becomes industrially available. Reprocessing of spent LWR-MOX fuel with a view to recycling the TRUs in FRs producing energy is conceptually possible in present reprocessing plants.

The HLLW produced during reprocessing of LWR-MOX fuel would require an additional TRU separation module to reduce significantly its radiotoxicity in comparison with non-reprocessed spent LWR-MOX fuel. The same technology as that proposed for HLLW from LWR-UO₂ would be adequate.

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

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

a greater impact than e.g. neptunium. The production of large amounts of depleted uranium (8 to 9 tons per ton LWR fuel), which is stored as UF₆ constitutes an important long term radiotoxic and chemical hazard which will have to be managed in a safe way.

3 Advanced fuel cycle (AFC) with partitioning of actinides

Any AFC scenario must rely on the use of a generic form of spent fuel reprocessing as an indispensable first step for an LWR-FR fuel cycle (see **Fig 3**) and for a complete double strata fuel cycle (**Fig 4**).

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

- Improved reprocessing of LWR-UO₂ fuel with additional Np removal
- Separation of MAs from HLLW resulting from LWR-UO₂ reprocessing.
- Fabrication of MA targets for heterogeneous irradiation in LWRs.
- Quantitative recycling of U and Pu into LWR-MOX fuel (single or multiple recycling).
- Reprocessing of spent LWR-MOX fuel in adequate facilities (higher Pu inventory).
- Separation of MAs from HLLW and conditioning of individual elements (Np, Am, Cm).
- Longterm storage and eventual disposal of specially conditioned MA.
- Fabrication of FR (-MOX, -metal or -nitride) fuel with a limited MA content.
- Irradiation of FR-fuel in Fast Burner Reactors or dedicated hybrid facilities (very high burnup).
- Reprocessing of spent FR-fuel in specially designed (aqueous and/or pyrochemical) and licensed facilities.
- Quantitative separation of all TRUs from the spent FR fuel processing during multiple recycling.
- Multiple recycling of FR-MOX fuel with major TRU content until significant depletion.
- Separation of certain fission products with long half-lives if required for the disposal step.
- Revision of the fission product management: Tc-99 separation (head-end, HLLW)
- Optionally: Platinum metals separation and recovery.

P&T has to play an essential role in the future AFC which is intended to reduce as much as possible either the long-term radiotoxic inventory, or the dose-to-man or

just the mean half-life of the waste. However, the implementation of the partitioning steps in present reprocessing facilities or the design in future plants are of prime importance for the successful implementation of the P&T option in the AFC fuel cycle.

3.1 Advanced reprocessing with quantitative Np removal (Fig 5)

Recovery of Np-237 from the U-Pu product stream is technically possible in the PUREX process. During the current reprocessing operations, Np is partly discharged with the fission products into the HLLW and partly associated with the U, Pu, Np stream in TBP. The purification of Pu and its quantitative separation from Np is achieved in the second extraction cycle of the PUREX process. It would be advantageous to adapt the first extraction cycle in order to co-extract the three actinides (U, Pu, Np) quantitatively and to recover the purified Np stream directly during the reprocessing. This will require an adaptation of the first extraction column and a refurbishment of the current U-Pu separation (BX column) equipment. Such adaptations can only be performed either in specially designed facilities or in existing facilities after a very important refurbishment.

Up to now the reprocessing of LWR-MOX has mainly been done by diluting the LWR-MOX fuel with LWR-UO₂ fuel according to the ratio in which it occurs in the reactor-core (UO₂/MOX=2). Reprocessing of spent LWR-MOX without dilution in UO₂ fuel has been demonstrated at COGEMA La Hague UP2 plant in 1992 as a special campaign (~5 t) and can be performed industrially if the reprocessing plant has been designed for the treatment of increased Pu concentrations and licensed for a much higher total Pu inventory.

Quantitative recovery of Np from dissolved spent fuel streams could be realized in adequately designed and /or refurbished reprocessing plants. Separated Np has to be stored under strict safeguarded conditions since its critical mass amounts to 55 kg.

3.2 Separation of Am-Cm from HLLW resulting from spent LWR fuel

During the conventional reprocessing operations most of the MAs (Np, Am, Cm) are transferred to the HLLW. Am and Cm (together with shorter-lived TRUs, Bk, Cf...) are quantitatively (>99.5%) transferred to HLLW. The separation of Am (plus Cm) from HLLW is the first priority from the radiotoxic point of view, it is also a prerequisite for a significant reduction of the (very) long-term radiotoxicity due to Np-237. The separation of Am-241 implies obviously also the separation of the longlived Am-243 parent of Pu-239.

Partitioning of all MAs from HLLW is presently under investigation in many laboratories throughout the world (Japan, France, China and some minor nuclear

countries) and was studied formerly in the US national laboratories (ANL, ORNL, Hanford...).

The Am (Cm) fraction contains all the rare earth (RE) elements which are in terms of quantity, about 10 to 20 times more important than actinides depending on the burnup. At 45GWd/tHM the ratio is 16 (13.9 kg RE compared to 0.870 kg Am-Cm per tHM spent fuel). Several processes have been studied at the conceptual level and tested in hot facilities; among the most important are: the TRUEX (**Fig 6**), DIDPA, TRPO, DIAMEX for An-Ln group separation, coupled to recent CYANEX 301™ SANEX, ALINA processes which allow the An/Ln separation. (**Fig 7**).

The most important criterion to be used in ranking the different methods is the overall decontamination factor obtained during extraction of HLLW and its comparison with the required DFs in order to reach the 100nCi level of α active nuclides in the high level waste. As can be deduced from **Table 1** the highest DFs should be reached for Am-241 separation viz. $3.2 \cdot 10^4$ if immediate separation is scheduled.

Table 1

Required DF's to reach 100 nCi/g in vitrified/alternative HLW

Nuclide	Immediate	500 years	1 000 years	10 000 years
238Pu	$9.8 \cdot 10^4$	$1.88 \cdot 10^3$	36	-
238Pu (repro)	326	6.25	-	-
239Pu	10^4	$9.8 \cdot 10^3$	$9.7 \cdot 10^3$	$7.5 \cdot 10^3$
239Pu (repro)	33	32.6	32.3	25
240Pu	$1.63 \cdot 10^4$	$1.54 \cdot 10^4$	$1.47 \cdot 10^4$	$5.66 \cdot 10^3$
240Pu (repro)	54	51	49	18
241Pu	$3.14 \cdot 10^4$	(241 Am)	(241Am- 237Np)	237Np
241Pu (repro)	(beta- 1%alpha) (92 as alpha)	$1.04 \cdot 10^5$ 346	$2.25 \cdot 10^4$ 75	21 21
242Pu	69	69	69	69
242Pu (repro)	-	-	-	-
237 Np	11.2	11	11	11
241 Am	$3.2 \cdot 10^4$	$1.45 \cdot 10^4$	$6.4 \cdot 10^3$	(Np)17
243 Am	710	678	647	276
244 Cm	$8.27 \cdot 10^4$	(240Pu) 229	(240Pu) 205	(240Pu) 80
245 Cm	7.5	7.5	7	3.3

According to the hot cell tests carried out at the JRC-ITU at Karlsruhe the DIAMEX process represents the best compromise among the first series of methods. DFs of ~1000 have been obtained for the MAs from 3.5M acid concentrated HLLW. The separation of An/Ln has been demonstrated with the ALINA process based on the use of a new organo-sulfinic acid extractant and seems to work properly in laboratory conditions. DFs of about 30 were obtained in 0.5 M acid solution. One combined flowsheet DIAMEX-SANEX is shown as an example in **Fig 8**.

The TRUEX process is very effective for α decontamination of medium level and non-heating high active waste streams. In order to obtain a MA fraction with 90% purity a An/Ln decontamination factor of more than 100 is required for the RE fraction. A 99% purity involves an An/Ln separation factor of more than 1000.

Except for the US ex-military facilities where kilogram scale separations were performed the present research facilities in Europe have strong limitations with regard to the quantities of MA which can be handled in shielded facilities. For example the new MA laboratory of JRC-ITU has an authorization for maximum 150 g Am241 and 5 g Cm244.

A NPP park of 100 GWe produces annually about 1,600 kg of each Np and Am-Cm. A big chemical engineering effort will be needed to upscale the laboratory methods to pilot scale and subsequently to industrial prototype scale in order to include MA separation rigs from the design phase on, in the future advanced reprocessing plants.

Looking at the existing situation in Europe we can take the industrial RFC for granted. There is sufficient reprocessing capacity (La Hague, Sellafield) to cover the European and some overseas needs for the next 20-30 years. The first steps to implement the AFC are: the installation of the separation facilities for Minor Actinides (MA) from liquid HLLW and the conditioning of these nuclides for intermediate storage or as potential target material for transmutation.

"Actinide -free - HLW", could be produced by the vitrification plants and stored for cooling in surface facilities followed by geological disposal. There are no objective arguments to oppose geological disposal of such a waste stream which decays with more than four orders of magnitude during 500 years.

3.3 Impact of TRU separation yields on secondary waste generation

The secondary waste generation of the PUREX is mainly determined by the radiolysis of TBP and the elimination of DBP by washing with sodium carbonate. The standard DBP formation is about 200 mg/l at an irradiation level of 0.35 Wh/l. By extracting high burnup FR-MOX solutions an increase of at least a

factor 7 may be expected: ≥ 1.4 g/l. The secondary (organic) waste generation by FR-MOX at 2.25 to 2.5 Wh/1 amounts to approximately 1.4 kg/hour and 7 kg DBP per ton of FR-MOX. A similar quantity may be expected from the plutonium separation and purification flowsheets.

The use of salt-free reagents e.g. hydrazine, hydroxyl-amine-nitrate... are improvements which may decrease the sodium content in the HLLW stream and reduce its content in the glass.

The presence of DBP in the organic solvent will decrease the Pu recovery factors and increase the fission product contamination (particularly of Zr95) of the plutonium product.

The sources of secondary wastes from the MA separation methods are in general the following:

- Aqueous liquid waste from solvent cleanup
- α contaminated solvent
- Condensate from evaporation and denitration steps
- Scrub liquors from valency adjustment (in case of Np)
- Different liquid and solid wastes from Np,Am,(Cm) product preparation
- Solid contaminated wastes from Np,Am-Cm target preparation
-

Elimination of Np-237 during the advanced reprocessing operations will slightly increase the secondary waste as the stripping of Np from the Np-Pu stream takes place in the second Pu-purification cycle. However most of this waste streams can be recycled to the HLLW acid recovery step and mixed with the concentrated HLLW.

Very little is presently known about the secondary waste generation resulting from the introduction of Am-Cm separation methods.

Only rough estimates, based on the number of stripping operations, have been made of the secondary waste volumes generated by the TRUEX, TRPO and DIAMEX processes. The An/Ln separation factor is very difficult to estimate, but according to recent laboratory results obtained with the DIAMEX-DTPA method a ratio of about 30 was obtained between the Am-Cm-RE feed and the Am-Cm raffinate using a 8 stages liquid extraction process. This is obviously too small to be used in real conditions. The total effluent volume of the process is 1.5 times the initial Am-Cm-RE volume. Significant fractions of the Tc-99 and Zr-95 concentration from the High Active Raffinate (HAR) occurs in the Am-Cm resp. Np-(Pu) streams.

Per unit of extraction raffinate (HAR, before acid recovery i.e. ~ 5 m₃ / tHM) the following relative figures, summarized in **Table 2**, have been given:

Table 2

Estimated relative volumes of product stream per unit HLLW volume

Process step	TRUEX	TRPO-CYANEX	DIAMEX-SANEX
PUREX-HAR	100	100	100
Solvent cleanup	189	50	150
An/Ln separation	125	40	150
Np/Pu segregation	50	40	--

3.4 Conditioning of separated MA and fabrication of MA targets

The potentially separated MAs could temporarily be stored as pure oxides, but the critical mass of Am being 34 kg and that of Np 55 kg, severe safety and security precautions should be taken in order to safeguard the separated nuclides during their storage period.

For waste management purposes the separated Np, Am-Cm could preferably be mixed with a very insoluble matrix of the type Zirconolite, Hollandite and Perovskite known as "Synroc"; up to 30 wt% Pu and MAs can this way be immobilized. Once in the embedded form, retrieval of the nuclides from the matrix is very difficult. Their solubility in geologic fluids is several orders of magnitude lower (10^{-6} g/sq.m/d) than conventionally vitrified waste. Since the leach rate and the solubility in groundwaters determine the ultimate radiological risk such procedure would sharply decrease the long-time risk of a repository in comparison with conventional vitrification of HLLW. However the criticality issue involved when locally large amounts of MAs are transferred to a repository remains to be addressed and calls for additional studies. Mixing of conditioned MA fractions with very refractory neutron absorbing materials e.g. B₄C or RE mixtures is a possible approach.

Though the Cm²⁴⁴⁻²⁴⁵ content is only about 5% of the total MA fraction, the α radioactivity of the Cm²⁴⁴ isotope which is very high compared to that of the other MA, dominates the radiological and technological problems (critical mass, neutron- and heat- emission) during the conditioning operations. However separation of Cm from Am is very difficult and expensive. Separate Cm storage e.g. during 100 years would simplify the MA storage- and handling problems but amplify those of the pure Cm compound. After the decay period of 100 years the residual product would be composed of 97% Pu²⁴⁰ and could be recycled in the bulk Pu stream. **This option supposes the availability (after one century) of aqueous reprocessing plants to recycle the decayed targets.**

The second option is the transformation of the MA fraction into a ceramic irradiation target material ($\text{Al}_2\text{O}_3, \text{MgAl}_2\text{O}_4, \dots$) and its storage in critically safe fuel type storage racks till the transmutation technology becomes operational.

The management of the mixed MA fraction requires a very precisely determined strategy of what will be the fate of the separated nuclides in a time frame of 100 years prior to its transfer to a geologic repository. If transmutation of MAs is chosen as an option before disposal, the induced heat associated with the formation of Pu-238 and Cm-244 will determine the necessary cooling period before transfer to a repository. Within this time frame it would be necessary to keep a type of TRU recycling technology open in order to reclaim or recycle unwanted waste streams.

New facilities have to be designed and constructed for the conditioning of fuels or targets

- Hot pressing or High Isostatic Pressing at elevated temperatures have to be designed for use with highly α active powders emitting non negligible neutron irradiation. Since 20 to 30 w% α active material can be taken up in target compounds, facilities with an annual throughput of 5-8 tHM of both Np and Am-Cm, have to be made available for a 100 GWe-y spent fuel output. For a mixture of MAs a highly shielded and remote handled (and serviced) fuel/target processing facility with a throughput of 10 to 16 t MA has to be constructed and operated during several decades.
- For the fabrication of Am-Cm irradiation-(fuels or) targets new technologies e.g. sol-gel fabrication, or inert matrix pellet impregnation (INRAM), are to be envisaged. The first technique can be used for medium active α emitters, but is probably not suited for very hot material (Cm). The second technique is under laboratory investigation. An industrial pilot facility of 5-8 t per year would be required to handle the annual output. Since the Am-Cm content in the ceramic matrix does not exceed 20 to 25% the overall target plant-capacity would amount to 25 to 40 t per year.
- For the fabrication of Np containing fuel assemblies or target pins, conventional MOX fuel fabrication techniques can be employed. As the Np content of FR fuel may not exceed 2.5% a MOX-fabrication capacity of 64 t U-Pu-Np fuel would have to be reserved for that purpose. If thermal irradiation of Np would be proposed the resulting Pu-238 concentration in the irradiated target becomes the limiting factor in the Np enrichment of the initial U-Pu-Np fuel.

It may be assumed that within the period from 2015 till 2040 one or several of the above fuel or target fabrication technologies could become operational at the industrial level. Since the methods for waste conditioning and fuel fabrication are more or less similar, both options can be taken from the onset and preferably a dual option i.e. conditioning of MAs as potential irradiation targets.

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

The presence of large quantities of Am-241 accompanied by 1 to 10% RE will require fully gamma shielded and remotely operated fabrication facilities. The presence of 5% Cm-244 in an Am-241, 243 target will amplify the degree of technical complexity due to the additional neutron shielding resulting from the spontaneous fission rate and from the α -n reaction in oxide-type isotopic targets. Extensive experience has been gained in the production of isotopic heat sources, but the present radiologic context and the ALARA limitations to be expected from regulatory bodies on industrial activities render the recycling of MAs very different from what it was in the past for military and space applications.

Conditioning of pure MA fractions with formation of very insoluble and thermodynamically stable compounds is a positive contribution to the longterm risk reduction. Since the heat output has not been modified from its level in spent fuel, their occurrence as individual waste containers among the high level waste canisters in underground facilities does not influence the overall repository design. However it may have an effect on the local activation of structural- and backfill materials.

The presence of conditioned MA concentrates in the repository reduces its dispersion throughout the spent fuel or HLW canisters and decreases the contamination risk during accidental human intrusion. However it does not fully eliminates this hazard. Transmuted MA targets on the contrary have to be kept in surface storage for extended periods of time due to the increased heat output resulting from generation of Pu-238 and Cm-244 in the targets. The radiotoxicity of these targets is extremely high during a decay period which ranges from 200 years for "spent" Am targets due to the residual Cm-244 content and to 800 years for Np targets containing Pu-238.

3.5 Separation of long-lived fission and activation products

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

Tc-99 is a fission product with a half life of 213 000 years which occurs as Tc metal and TcO_2 in the insoluble residues and as soluble pertechnetate ion in the HLLW solution. Its generation rate is 26.6 kg/GWe-year and an overall specific concentration of 1.2kg/tHM. In order to effectively address the long-term radiotoxicity problem both, soluble (80%) and insoluble (20%), fractions ought to be combined before any nuclear action is taken towards depletion by transmutation.

The extraction of soluble TcO_4 is relatively easy. The similarity between Tc and the platinum metals in insoluble waste and the nature of the separation methods makes this partitioning operation very difficult. Separation from aqueous effluents is possible in an advanced PUREX scheme. However recovery from insoluble residues is very difficult. The present recovery yield could approach 80% at best (DF = 5).

A significant improvement of the Tc-99 recovery from HLW is only possible if it is converted into a single chemical species which is not easy to achieve. Pyrometallurgical processes are perhaps more adequate to carry out a group separation together with the platinum metals.

I-129 is in most of the land-based repository concepts for spent fuel the first nuclide to emerge in the biosphere due to its very high mobility in aquifers. In spent fuel it occurs as molecular iodine, as soluble CsI, as solid ZrI_{4-n} and as volatile ZrI_4 .

Iodine as fission element is generated in spent fuel at a level of 7.1kg/GWe-year. About 80% of this inventory is present as the very long-lived (16 million years) isotope I-129 and 20% as stable I127. During reprocessing (PUREX, UREX) it is removed from the dissolver solution with a yield approaching 95-98% (DF of 20-50) The radioactive concentration of this nuclide in spent fuel is, depending on the burnup $\sim 1.6 \cdot 10^9$ Bq/tHM and its ALI is $2 \cdot 10^5$ Bq. Since its radiotoxicity is the highest among the fission products ($1.1 \cdot 10E-7$ Sv/Bq), and being very soluble, it would be advisable to increase the separation yield from different waste streams to reduce the radiological impact. A target DF of ~ 1000 could be proposed as a significant improvement.

In order to improve this separation yield more complex chemical treatments are necessary. During high temperature pyrochemical processes higher separation yields could in principle be expected. Adapted "conditioning" methods for separated iodine (AgI, PbI_4 ,...) have been developed.

The separated fraction can either be stored on a specific (zeolite) adsorbent or discharged into the ocean. Since I-129 has a half life of 16 million years it cannot be prevented from entering into a world-wide dispersion in the geo- or biosphere. However in a salt-dome (evaporated sea water) type of repository the dilution of

eventually migrating I-129 by the mass of natural iodine (I-127) present in the body of the salt dome strongly decreases the radiological hazard.

In a world-wide dispersion scenario in the oceans, its radiotoxic importance is rather limited due to its dilution by natural (I-127) iodine, as long as the LWR-reprocessing capacity remains at its present levels.

But conditioning and confinement are the preferred management options to reduce its radiological impact and final storage in a salt dome is an alternative which undoubtedly has its merits.

Se-79 is a fission product (0.16 kg/GWe-y) with a half life of 65,000 years which occurs in the HLLW. Chemically this nuclide behaves as a sulfate and will be incorporated in vitrified waste. Its radioactive concentration in spent fuel is expected to be around $2 \cdot 10^{10}$ Bq/tHM and its ALI is 10^7 Bq/year. Separation from liquid HLLW is not obvious taking into account the very small chemical concentration in which it occurs, in comparison with natural sulfur compounds.

Zr-93 and **Cs-135** are two long-lived (1.5 and 2 million years half-life, respectively) nuclides which occur in spent fuel at relatively high concentrations of 23 kg resp. 12.5 kg/GWe-y. Separation of these radionuclides from the other fission products for eventual transmutation is almost excluded since they are accompanied by other radioisotopes which are either very radioactive (Cs-137) or present in much larger quantities (~ 23 kg Zr-93 among 118 kg Zr per GWe-y). In order to effectively reduce the radiotoxic potential by neutron irradiation, a series of isotopic separation processes ought to precede any target fabrication and this route is presently considered as an almost impossible endeavor as well from technical as from economical point of view.

Sn-126 has a half life 250 000 years, is partly soluble in the HLLW and occurs partly in the insoluble residues. Its chemical concentration amounts to 0.72 kg/GWe-y Sn-126 among 1.81 kg Sn /GWe-y in spent fuel. The radiochemical concentration of Sn-126 in HLLW ranges around $3.2 \cdot 10^{10}$ Bq/tHM and its ALI limit of $3 \cdot 10^6$ Bq/tHM. The radioactive species Sn-126 is accompanied by a series of stable isotopes (Sn-116, 118, 119, 120, 122, 123 and 124) which makes it difficult to consider its transmutation.

Activation products

C-14, with a half life of 5 730 years, is a difficult case because it can potentially enter into the biosphere through its solubility in groundwater and play an important radio-toxicological role because of its uptake into the biochemical life cycle. According to the nitrogen contamination of the initial UO_2 fuel its concentration in spent fuel is about $3 \cdot 10^{10}$ Bq and its ALI limit $4 \cdot 10^7$. Its role in the long-term radiotoxicity is dependent on the physico-chemical conditions

occurring in deep underground aquifers or in water unsaturated geospheres. The capture cross section in a thermal neutron spectrum is negligibly small.

Cl-36

Zircaloy cladding contains some natural chlorine impurity at the level of 5 to 20 ppm. During irradiation this Cl-35 is transmuted into Cl-36 with a half life of 300,000 years. This activation product arises partly in the dissolver liquid and partly remains within the washed Zircaloy hulls. At 45 GWd/tHM about $2 \cdot 10^6$ Bq are calculated to be globally present in the HLW and MLW. The ALI by ingestion is $2 \cdot 10^7$ Bq. Due to its chemical characteristics this nuclide is gradually dissolved in groundwater and could contaminate water bodies around a repository. This radionuclide cannot be considered in a recovery or transmutation scenario. The presence of natural Cl-35 in all natural waters precludes further transmutation

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

Conclusions on the advanced aqueous reprocessing options with P&T

- Separation of Np-237 is technically possible in refurbished reprocessing plants or in newly designed facilities
- Separation of MAs has been demonstrated at the "hot-cell" laboratory level. The flowsheets are very complex and need to be simplified in order to allow upscaling to technological levels.
- The secondary wastes expected from MAs separation are of the same order of magnitude as from the second cycle of the PUREX process.
- Separation of MAs followed by conditioning as a ceramic waste form and retrievable storage is a valid option for the MAs management.
- Among the long-lived fission products only I-129 has a radiotoxicity comparable to that of the actinides. The radiotoxicity of the other fission products is ~1000 smaller. Isolation and specific conditioning of I-129 is advisable.
- Transmutation of Tc-99 is technically feasible but economically very difficult to realize.
- All the other long-lived fission products can not be separated effectively from HLLW unless isotopic separation techniques are envisaged.
- The activation products C-14 and Cl-36 are troublesome contaminants of medium level wastes.

4 Advanced management of TRU nuclides with recycling

4.1 Plutonium recycling in LWR-s

Reprocessing of LWR-UO₂ fuel has for several decades been realized in Europe on industrial scale due to the existence of the two large reprocessing plants of Cap de la Hague in France and Sellafield in the UK with a total nameplate capacity of 2800 tHM LWR-UO₂ and 1100 tHM GCR-U. This process capacity is sufficient to cover the internal needs of Europe and some from overseas customers. The use of separated plutonium was initially started to feed a fast reactor program, but has been delayed or even put to an halt due to technical and economic constraints. The effective reprocessing capacity produces a throughput of about 25 t of Pu per year.

Presently the use of separated plutonium is almost exclusively connected with the production of LWR-MOX fuel which is used as substitution for LWR-UO₂ fuel. But the delay in the construction of the LWR-MOX facilities has led after several decades to an increased fraction of stored separated plutonium. Presently the overall LWR-MOX fuel fabricating capacity in Europe has reached about 300 tHM, (~ 25 t Pu/year) which is in equilibrium with the reprocessing output capacity.

The LWR-MOX fuel output with Pu enrichments varying from 5.25 to 8.2% depending on the fissile isotopic content and on the delay between reprocessing and fuel fabrication is used in thermal reactors in a proportion of 20 to 33% of a reactor core. The balance between the consumption and the production of Pu in the reactor core depends on the burnup of the fuel.

In a generic case of a reactor core loaded with 1/3 LWR-MOX fuel and 2/3 LWR-UO₂ and irradiated till 50 GWd/tHM the balance between the Pu consumption in the MOX fuel and the production in the UO₂ fuel is slightly negative, but if the production of MAs in both parts of the core are being considered there is an increase in the overall TRU content.

The inventory reduction of Pu in irradiated LWR-MOX amounts to 33% but almost 10% is transformed into MA. The net TRU inventory reduction is in this case ~24%. When totaling the generation of TRU in such recycling process the overall radiotoxicity increase is small (about 8.6%) but not negligible.

Advanced LWRs capable of having a 100% LWR-MOX fuel load could decrease the Pu inventory at a more rapid pace since at each cycle the Pu inventory would be decreased by 33% without compensating production of Pu in the core. The mass reduction of the TRU nuclides does not mean a reduction in actual radiotoxicity since this depends on the radioisotopic composition of the irradiated fuel as a function of storage time compared to the initial composition.

Multiple recycling of plutonium in LWR-MOX is not efficient in terms of radiotoxicity reduction because of the low thermal fluence. A significant reduction of the radiotoxic inventory of nuclear materials can only be

accomplished by performing transmutation at very high thermal neutron fluence ($>10^{22}$) followed by irradiation in fast spectrum devices (FRs or ADS). The radiotoxic inventory of spent LWR-MOX fuel is about 8 times higher than that of spent LWR-UO₂. Conventional reprocessing will remove U+Pu which accounts for about 30% of the total alpha activity and the residual 70% made up of Np, Am and Cm, enters into the HLLW. However the Cm and Am isotopes constitute the overwhelming majority of this α activity.

In a perspective of P&T, it would be indispensable to remove the TRUs from the HLLW before vitrification. The techniques to be used are in principle the same as for the LWR-UO₂ fuel (see section 3.2), but the higher alpha activity level will interfere with the extraction because of increased radiation damage.

Another option is to store the spent LWR-MOX fuel for example during 50 or more years and to let Cm-244 decay (18 years half-life) to Pu-240 before carrying out the reprocessing. The chemical extraction processes are much easier to perform after the extended "cooling" period as the alpha decay heat is reduced by a factor of 7 or more, depending on the isotopic composition.

4.2 MAs recycling in LWRs

The Minor Actinides constitute the main source of radiotoxicity in the vitrified High Level Waste. Their confinement from High Level Liquid Waste (HLLW) produced during advanced reprocessing and conditioning in a insoluble matrix, is a first, but not sufficient step. In the risk reduction of long-lived waste, transmutation of long-lived MAs with formation of shorter lived nuclides and "incineration" with the formation of the much less-toxic fission products could be a second step which would be crucial if successful to reduce the longterm radiotoxicity. This very complex issue has been investigated during the preceding decade. Since the present reactor park is for 99% constituted by thermal neutron reactors and particularly by LWRs, the question was raised whether these worldwide operating reactors could be used to eliminate by thermal neutron irradiation the MA inventory produced during power production.

Homogeneous recycling of MAs in LWR-MOX fuel is a practice which does not keep the surplus MA source term confined from the driver fuel and would further dilute the MAs, especially Np-237, in a larger volume of spent LWR-MOX fuel which would be very difficult to reprocess later on.

Heterogeneous recycling of MAs in the form of irradiation targets or individual fuel pins in a LWR-UO₂ or LWR-MOX core is an alternative which is much more attractive from fuel cycle point of view, since target fabrication and chemical processing are independent from the industrial large scale operations on spent fuel.

The difficulty to handle highly irradiated MA targets in a multirecycling operation led the P&T community to consider the "once through irradiation" as a possible

option. Per year Europe produces 1.6 t of Np and a similar amount of Am-Cm. For reactivity reasons only 1% Np or Am-Cm may be loaded in a large LWR core. The addition of MAs in a LWR-core would call for an equivalent increase of the U-235 enrichment.

4.2.1 Irradiation of Np

The capture cross-section of Np-237 prevails in LWRs and leads to the production of Pu-238 with some minor Pu-239 and fission product impurities. The transformation of Np-237 into Pu-238+239 is almost quantitative (~95%). Only if recycling is performed in particular conditions of low burnup, pure Pu-238 is obtained but it needs frequent reprocessing of the irradiation targets.

Homogeneous irradiation of 1% Np in standard PWR-UO₂ fuel to a burnup of 47 GWd/tHM leads to a depletion of about 50% i.e. only a reduction factor of 2 is obtained. Further reduction of the Np inventory would require repeated and selective reprocessing of this "Np doped" fuel, which is a complex and very expensive process. The reprocessing of this fuel would transfer all the generated Pu-238 content to the recycled plutonium.

Quantitative conversion of Np-237 into Pu-238 reduces the (very) longterm radiological impact of a repository but increases the medium term radiotoxicity of the target by a factor of $2.4 \cdot 10^4$ for a period of several hundred years. From technological point of view the Np-237/Pu-238 conversion has important consequences for the heat dissipation in a repository. The decay heat of Pu-238 amounts to 0.55 W/g or 0.55 MW/t Pu238.

Quantitative transmutation by heterogeneous recycling of Np targets totaling 1.6 t Np, produced annually by a 100 GWe LWR-UO₂ fuelled reactor park would increase the heat load of the equivalent repository by 880 kW (1.6 Mg x 0.55 W/g) or 40% of the heat load produced by an equivalent mass (2200tHM) of 50 years cooled spent fuel elements. (~1 kW/tHM).

4.2.2 Irradiation of Am

Irradiation of Am is a much more complex nuclear reaction scheme as two different isotopes (Am241 and Am243) are transformed into Cm242 and Cm244 (see **Fig 9**). But due to the short half life of Cm242 (162 d) this nuclide decays during its stay in the reactor to Pu-238 and is transmuted on his turn into Pu-239. The discharged target after 3 years of irradiation contains still 27% Am, 60% (Cm+Pu) and 13% fission products. Homogeneous irradiation results in 83% Am depletion but the produced mix of nuclides is the same. Fabrication of special 1% Am enriched LWR-UO₂ fuel assemblies, followed by their irradiation in a conventional PWR could be a preliminary step in the gradual decrease of this very radiotoxic element. However from radiotoxicity and heat generation points of

view this practice can only be an intermediary step in the Am inventory reduction which has to be completed by high flux thermal and fast neutron irradiations. Moreover from economic point of view the inclusion of 1% Am into LWR-UO₂ fuel needs an surplus enrichment of 1% U-235.

4.2.3 Conclusion on thermal neutron irradiation of MAs

Conventional LWRs cannot be used to decrease significantly the Np and Am radiotoxic inventories resulting from LWR-UO₂ reprocessing as neutron capture reactions dominate the nuclear transmutation processes. The presence of highly active α emitters (especially Cm244) in irradiated targets make multi-recycling nearly impossible.

In order to get a net MA destruction yield the elimination of the nuclide must exceed the internal production. A conventional 1 GWe LWR-UO₂ reactor core generates 1.8 kg/TWh of Np and 1.6 kg /TWh of Am-Cm. A dedicated 1 GWe LWR-UO₂ reactor could accumulate gradually a MA inventory of 1% i.e. about 800 kg Np or Am-Cm. After a long residence time of say 7 to 8 years, the fissioned fraction amounts to only 7% for Np (~56 kg) and 10% for Am (~80 kg) the balance being a mixture of residual Np (60%) or Am-Cm (40%), and their transmutation products Pu-238, Pu-239 and Cm-244 leading to a specific depletion of 4.2 kg Np/TWh and 5.8 kg Am-Cm/TWh

Dedicated LWR transmutation reactors could serve as "intermediary storage" plants for separated actinides if an additional enrichment of 1% U-235 has been provided. Four powerplants dedicated to the "uptake" of separated MA would have to be licensed annually for these purposes to absorb the yearly MA production from a 100 GWe power grid. This scenario will gradually transform the introduced Np and Am-Cm targets into a mixture of fission products, Pu238 and Cm-244. This approach would reduce the mean lifetime of the nuclides involved but increase very drastically their radiotoxicity.

Calculations performed with several nuclear transmutation codes (SCALE, MCNP and ORIGEN-S) have shown that in order to achieve a inventory reduction of 100 in a Np-237 target a total thermal fluence of $1.15 \cdot 10^{22}$ n/cm² is necessary which corresponds to an irradiation period of 30 years in a conventional power plant. The transformation into 95% Pu-238 multiplies the radiotoxicity during the first 870 years after discharge from the reactor with an initial factor of up to $2.4 \cdot 10^4$. Only after 10^5 years, the radiotoxicity decreases significantly below the original value of unirradiated Np237.

Somewhat better results were obtained with Am-241-243 targets, but the order of magnitude of the irradiation time is the same. The final radiotoxicity decreases faster because a larger fraction is fissioned but about 20% is transformed into Pu238-239 and Cm244.

The low MA transmutation capability of LWRs has been demonstrated by different R&D groups in Europe and have shown that by following the once-through irradiation, only with very long irradiations (20 to 30 years) and with higher U-235 enrichment, significant results could be obtained.

The stabilization of the TRU inventory with LWRs is only possible by generalizing the use of LWR-MOX fuel with an increased U-235 enrichment in the entire reactor park. It would profoundly influence the fuel fabrication needs for LWR-MOX and require additional U-235 enrichment capacity. It is outside the scope of this report to discuss the issues involved.

4.3 Multirecycling of TRU's in FRs :possibilities, issues, consequences

In the recent past consideration was given to using the separated plutonium and MA as a feedstock for FR-fuel for "accelerated incineration" of TRUs. This option asks for special reactor core designs (IFR, CAPRA...), advanced fuel types (oxides, metals, nitrides) and last but not least new or advanced reprocessing techniques (e.g. Pyrochemical reprocessing).

4.3.1 MOX-Fuel fabrication and refabrication problems

The largest industrial experience has been gained in the FR-MOX fuel fabrication since for several decades FR programs were undertaken in many nuclear countries. The fabrication of FR-MOX fuel with 15 to 25% Pu has been realized routinely and on a commercial basis. But the Pu quality used for these purposes was derived from low burnup UO₂ fuel with low Pu-238 and Pu-242 contents.

In the mean time the burnup of spent LWR-UO₂ and LWR-MOX has reached 50 GWd/tHM. The isotopic composition of Pu resulting from the reprocessing of such fuels is seriously degraded, with high Pu-238 and Pu-242 levels, and low Pu-239 and Pu-241 concentrations.

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

For homogeneous recycling of MAs in FR-MOX, admixtures of 2.5% Np-237 and/or Am-241 are currently studied. Np-237 is a pure alpha emitter (except for the small in-growth of Pa-233) and there is no major handling problem involved; but the admixture of Am-241+243 at the 2.5% level will induce a gamma field around the glove-boxes or hot cells. However, the major interfering nuclide in FR-MOX is Pu-238 at the 3% level which is a heat and neutron source (7.4 kWth/tHM; 7.210^8 n/s·tHM).

The FR-MOX fuel fabrication with 2.5% Am admixture will also be influenced by the degree of separation of the rare earths (strong γ emitters) and last but not least by Cm-244 which will accompany the Am fraction when separated from HLLW. The presence of ~17% Cm-244 in the Am-Cm fraction of recycled FR-MOX fuel will further increase the neutron emission up to $\sim 4.4 \cdot 10^{10}$ n/s per tHM

The separation coefficients from rare earths and Cm-244 required in order to permit industrial fuel fabrication operations will greatly depend on the permissible RE concentration acceptable in fresh FR-MOX fuel and on the permissible Cm-244 concentration during in the fuel fabrication plant.

It has to be remembered that specially equipped MA fuel fabrication laboratories e.g. those of the Institute for Transuranium elements in Karlsruhe are licensed to handle 100g of Am-241 and 5 g of Cm-244.

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

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

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

Very recently, attention was drawn on the potential of nitride and carbide fuels for fast burner reactors (FBuR). Nitride TRU fuel containing macroscopic quantities of MAs can be produced by a combination of an internal gelation method and a carbothermic synthesis. These nitride fuels can be reprocessed by electrorefining methods similar to the technology developed for metal fuel.

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

Reprocessing of metal and nitride fuel relies on the use of pyro-chemical processes, and is followed by pyro-metallurgical fuel fabrication for recycling in FRs or ADS systems.

4.4 Irradiation of MAs in critical FRs

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

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

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

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

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

Complete elimination of the actinide inventory by neutron irradiation can only be accomplished in a fast neutron spectrum, of either "critical" or "subcritical", reactor. The real net reduction results from fission and is consequently proportional to the burnup of the fuel or target. High burnups are the prerequisite condition for a reasonably fast reduction pace but involve on the other hand the industrial implementation of the most difficult reprocessing operations on very "hot" nuclear materials.

The maximum burnup obtainable in a fast neutron spectrum device (FR or ADS) is in principle not limited by the fissile material content, since all TRU's are to a certain extent fissionable, but by the resistance to fast neutron radiation damage of the cladding material expressed in "displacements per atom". If the fast neutron dose exceeds 200 dpa the stability of the cladding material is at stake and the highly irradiated fuel or target (150 to 250 GWd/tHM) will have to be withdrawn from the reactor and recycled or disposed of.

Irradiation as dissolved fuel or target in molten salts is not considered as realistic in the present state of technological development. However it might become in the far future an option to be investigated if adequate construction materials could be employed.

Experimental and theoretical work which has been performed on the irradiation of MAs or on a mixture of Pu + MAs, point at the fact that multirecycling is necessary to achieve a significant inventory reduction factor. Burnups of 150 GWd/tHM could be taken for granted but yield a TRU depletion of only 17%, which corresponds to a situation in which 83% TRU fraction is still present in the target or fuel. The "discharged" TRUs have equal or higher masses than those of the initial target. Burnups of 250 GWd/tHM are at the higher bracket of present achievements

4.4.1 Transmutation/incineration of Np

Homogeneous recycling of Np in a typical fast reactor (EFR) has not much impact on the neutronics of the core as long as the Np content remains below 2.5%. Such type of reactor could be used as a "storage" reactor since approximately 1 ton of Np could be accumulated in the core. As a result 1.6 GWe EFR capacity is capable to absorb all the Np resulting from the LWR-UO₂ spent fuel reprocessing produced annually by a 100 GWe grid. However this option will not significantly deplete the Np content since the fuel has to follow the reactor cycles and has to be periodically discharged from the reactor. Reprocessing of such hot fuel dissipating > 30 kW/tHM and containing high levels of Pu-238, is not industrially feasible in the present aqueous reprocessing plants. Aqueous reprocessing could perhaps be achieved by diluting the spent FR-MOX-MA fuel assemblies with spent LWR-MOX fuel assemblies according to a minimum ratio of 1FR/2LWR.

The non-reprocessed FR-MOX spent fuel has in any case to be stored for an indefinite period of time in engineered facilities or transferred in a retrievable way to a repository where it would possibly be "diluted" among a large number of LWR-UO₂ spent fuel assemblies, in order to decrease the local heat load. The long term radiotoxicity of such fuel assemblies is very high and decreases mainly with the Pu-238 half life (87 years).

Transmutation of Np targets in FRs at a burnup of 120 GWd/tHM yield a depletion of 60% but a fission rate of merely 27%. In the burnup bracket of 150 to 250 GWd/tHM the depletion yield will further increase but the final composition will be determined by the fission to capture ratio of Np-237. Unless multirecycling of the targets is performed no significant (factor of 10) depletion can be achieved. Reprocessing of the irradiated targets is possible in small dedicated facilities where either fast contactors are used for aqueous extraction or possibly in the future pyrochemical separation methods are put to work. By multirecycling, the Np inventory of the targets could be decreased by a factor of 10 after 5 recycles. This is the price to be paid to decrease a very longterm hazard without any benefit in the short and medium term.

Transmutation of Np in FRs is theoretically possible but the radiotoxic advantage is not very obvious since the transmutation product is to a significant extent Pu-238 and Pu-239.

4.4.2 Transmutation of Americium

Since Am 241 is the precursor of Np-237, and constitutes the highest radiotoxic potential after the Pu-isotopes, its transmutation is much more defendable from radiotoxic point of view. Great progress has been achieved in the separation of Am-Cm from HLLW and it may be anticipated that concentrates of Am-241-243 + Cm242-244 (with 10 to 50% Rare Earths) might be produced in an extension of the reprocessing plant. For obvious reasons it is preferable to keep the separated Am-Cm fraction in targets (mixed e.g. with Al_2O_3) to be irradiated at a later stage. However the fabrication of Am-Cm targets is the main technological problem to be overcome before any significant irradiation campaign can be envisaged.

Transmutation /incineration of the Am fraction in a ZrH_2 or CaH_2 moderated peripheral core position, is by all means the best way to reduce the Am inventory in a once through irradiation up to the limit of cladding resistance. In longterm irradiations (15 to 20 years) 90 to 98% of the initial Am inventory can be "incinerated" i.e. nearly quantitatively transformed into fission products and some percentages of Pu238, Cm244-245.

In the specific case study carried out by CEA-EDF in the frame work of the EUR program on P&T, the Am targets were considered to be irradiated in a CAPRA type reactor. At 98% Am depletion the residual target contains the following nuclides: 80% FPs, 12.1% Pu (mainly 4.4% Pu242; 3.8% Pu 238 and 2% Pu 239) 2.16% Am and 5.2% Cm (2.38% Cm244; 2.8% Cm245) The radiotoxicity (expressed in $\text{Ci } \alpha$) of the initial target is $2.6 \cdot 10^6 \text{ Ci } \alpha$ and increases after irradiation to $3.8 \cdot 10^6 \text{ Ci } \alpha$, mainly due to the formation of Am-243, Cm-244 and Pu 238. After an intermediate storage of 100 years the radiotoxicity drops with 60% to about $1.1 \cdot 10^6 \text{ Ci } \alpha$.

The heat output of such discharged targets amounts to ~ 100 kW/tHM Am, among which the Fission products contribute for 20%, and the Actinides for 80%. The main heat contributing TRUs are: Cm-244 (71%), and Pu-238 (26%) decaying each with their half-lives of 18.4 and 87.8 years respectively. The main source of longterm radiotoxicity is due to the Pu isotopes (44 kg Pu242; 20kg Pu 239; 7.5 kg Pu240.per initial tHM Am) The Am irradiation targets are from waste-management point of view, very different from the classical spent fuel or vitrified HLW

Disposal of such intense heat and neutron sources is a challenge for the repository design in order to avoid hot spots in a mining environment. The annual Am output from a 100 GWe reactor park amounts to 1.77 tHM Am+Cm which will have to be "fractionated" in small targets and will generate an additional heat load by 465 kW/ year which is still small (21%) but not negligible in comparison with the total LWR-UO₂ spent fuel heat load after 50 years cooling.

The conclusion to be reached from these computations is that the (once through) irradiation of Am targets provides a solution to decrease the Am-(Np) inventory by a factor of 50 but that the radiotoxicity merely decreases only with a factor of 2.5 after 100 years cooling. Moreover this result shows that a preliminary Am/Cm separation which was studied previously in the framework of the P&T program does not seem to be necessary. The main issues to be resolved are the target fuel fabrication and the FR technology.

4.4.3 Multirecycling of Pu+MA in critical fast reactors

A combined irradiation of Pu and MAs is an approach which has been investigated in several reactor technology projects: Metal fuelled "Integral Fast Reactor" at Argonne National Laboratories(USA), the Minor Actinide Burner reactors at JAERI (Japan) and the CAPRA reactor project at CEA - Cadarache (France).

Each of these approaches displays a number of advantages and drawbacks which can be summarized as follows:

- the fuel types and configurations aim at the generation of an as hard as possible neutron spectrum
- metal or nitride fuels are slightly better than oxide fuel
- the Pu concentration increases from 33 to 45% to compensate the anti reactivity of the MAs
- the void reactivity coefficient is positive with Pu as fuel and Na as coolant
- substitution of Pu by U235 and Na by Pb greatly improves the reactor safety parameters
- burnups of 150 to 250 GWd/tHM have to be achieved

This set of improved irradiation conditions has resulted in an expected increase of the "incineration" yield of TRUs but has shown that, to reach a significant (factor >10) TRU depletion, multirecycling cannot be avoided.

Multi recycling of Pu+MAs from high burnup FR-fuel is still at the conceptual level and important progress has to be made before this option can be implemented. Computations have shown that an isotopic equilibrium state is reached from the fifth recycle. Aqueous reprocessing is probably inefficient for this purpose because long cooling times (10 to 12 years) between each recycle step are necessary to reduce the α radiation damage due to Cm-244 on the aqueous extractants. Pyro(electro)chemical reprocessing is in the long term the only viable option for multi-recycling with short cooling times, but this technology is still in its infancy, and encounters severe material problems.

The major reactor problem encountered in the design of dedicated FRs for MA burning is the positive void reactivity coefficient when the reactor is Na cooled. Therefore the MA content must be kept below 2.5%. The Am consumption rate lies around 14 kg/TWhe but the generation rate in a Pu fuelled FR amounts to 5 to 10 kg/TWhe. The net destruction rate ranges thus from 4 to 9 kg /TWhe and corresponds to the production of minimum 2 and maximum 4 LWR-UO₂ reactors. As a consequence a large electro-nuclear FR capacity (25 to 50% of the park) would be necessary to first stabilize the Pu+ MA inventory and then to decrease the overall actinide level (see **fig 10**).

4.5 Irradiation of TRU in Accelerator Driven 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. 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 neutron cascades and 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. (see **Fig 11**). 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 utilized for transmutation, conversion, and breeding purposes. These features may be exploitable to prepare a safe and efficient means 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 programs 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.

4.5.1 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 for 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.

In the high-intensity accelerator, beam loss should be minimized 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 development while operational experience will further increase the dependability.

The interaction of the beam with the surrounding materials in a direct or indirect way will strongly influence the activation of the structural and target materials. Any directional instability of the proton beam will result in an interaction of the high energetic protons with the wall structure of the beam tube. Apart from accidental penetration, the most important consequence will be the gradual activation of the tube particularly in the regions where the beam changes its direction. The nature of the radioactive contamination will depend to a large extent on the type of material used. Since stainless steel will generally be used,

the activation of the wall material will be similar to that encountered in LMFRs and in conventional proton accelerators used in research and isotope production.

4.5.2 Spallation target

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 materials, particularly the beam window, suffer radiation 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 of the impact of the high-energy proton and neutron beams on the window and the structural materials is very important for the future of ADS development.

In order to impinge on the (Pb, Pb-Bi, W) target, the proton beam must leave the high vacuum area of the accelerator to enter the subcritical core. A so called metallic "window" has to insure a relatively easy proton passage without too high energy loss in the metallic membrane. The window will according to recent calculations be submitted to very intense stress possibly inducing cracking of the window. Remote replacement of such membrane without interruption of the production process is the most challenging engineering task.

The highly active target material underneath the window can escape and contaminate the accelerator ducts and the surroundings of the ADS system.

In case Pb-Bi-eutectic (LBE) is selected as target material a wide range of spallation nuclides will be formed. The most important from safety and waste management point of view are the nuclides with a high vapour pressure and a relatively long half-life. **Table 3** gives an overview of these nuclides.

4.5.3 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 utilize 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.

Table 3

Main spallation nuclides with long half-lives in Pb-Bi.

Isotope	Half-life	Liq.-Gas T°	Predominant radiation	
Po-210	138.3 days	254-962°C	Alpha	5.3 MeV
Po-209	103 years	254-962°C	Alpha	4.88 MeV
Po-208	2.93 years	254-962°C	Alpha	5.11 MeV
Bi-208	3.7 10 ⁵ years	135-270°C	EC- γ	2.6 MeV
Bi-207	30 years	135-270°C	β^+	0.56 MeV
Tl-204	3.77 years	303-1457°C	β^-	0.76 MeV
Hg-203	46 days	-38 -356°C	β^-	0.2 MeV
Hg-194	1.9 years	-38- 356°C	EC	0.4 MeV

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.

The problems which have an impact on waste management are very similar for ADSs and LMFRs if the same cooling liquid is used. Very much experience was gathered in the past for sodium as LMFR coolant, but it is not obvious that sodium will be selected as ADS coolant since its void reactivity coefficient is positive.

Lead-bismuth eutectic (LBE) is preferred for its thermal, neutronic and safety features but is less attractive from corrosion point of view. The most important impact on waste management is the neutron activation of the LBE with an important generation of Po-210, a semi-volatile alpha emitting isotope with 138 days half-life.

The accelerator-driven subcritical system 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 supercriticality 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.

JAERI has designed two 820 MWth types of subcritical cores with nitride fuels. One type is Na-cooled core with a solid tungsten (W) target at the core center and the other is lead-bismuth target and coolant system. The spallation targets are bombarded by high energy and power protons of 1.5 GeV and 45 mA. The parametric studies were conducted to obtain the optimal neutronic characteristics of the subcritical core to maximize the MA transmutation rate and to minimize the burnup reactivity swing during irradiation by adjusting the MA and Pu fraction of nitride fuels with ZrN inert matrix as thermal diluent. The core design parameters are given in **Table 4**. The coolant void reactivity is negative in the lead-bismuth cooled core, though it is positive in the Na-cooled one. The MA transmutions in both cores are 250 kg/y and this amount corresponds to the annual production of MA in about 10 LWRs. FP (Tc-99 and I-129) transmutation is calculated for loading the FP target assemblies with ZrH₂ moderator pins in the core-reflector region. The core performance for FP loading is also given in the table.

Table 4

Characteristics of the Na and Pb-Bi cooled 820MW-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-effective (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/y) (MA/FP)	250 / -		250 / 40

The potential advantage of an ADS system lies in the flexibility of core loading and in its ability to transmute nuclear charges with a very high TRU or MA content. However from pure neutronic point of view there is no difference between the "transmutation" output of a LMFR compared to an ADS. In other words for the same transmutation output you need as many ADS systems as LMFRs. The technology of ADS systems is much more complicated than that of LMFRs and the investment costs per unit "neutron" output is roughly twice as

much. In a global waste management scheme the ADS systems will have to be reserved for a small percentage of very specific duties where conventional LMFRs cannot provide a safe solution e.g. residual MA transmutation. (see **Figs 4 and 11**).

4.6 Transmutation issues of long-lived fission products

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

The main fission products determining the size of the repository and the overall radioactivity of HLW are ^{137}Cs and ^{90}Sr . These two isotopes with half-lives of 28-30 years are not considered in P&T operations since their radioactive "life" is limited to about 300 years.

The fission products which play an important role in the long term dose-to-man originating from the back-end of the fuel cycle are in order of radiologic importance: ^{112}I , ^{99}Tc , ^{135}Cs , ^{93}Zr , ^{95}Se and ^{126}Sn . Their significance varies according to the type of repository host formation.

Some activation products are also of importance in the determination of the dose-to-man: ^{14}C and ^{36}Cl

The determination of the separation yield and the DF's from HLW depends to a great extent from the policy decisions with regard to HLW.

Iodine 129

Transmutation of ^{129}I is a difficult task because it has a small thermal cross section (27 b), is transmuted into a gaseous decay product (^{130}Xe and ^{132}Xe) and does not withstand large temperature excursions during irradiation. Up to now no thermally stable iodine matrix has been found and most of the calculations have been done for NaI , CaI_2 and CeI_3 . Each of these candidates have their limitations, but CeI_3 seems the most promising. Two opposite safety requirements have to be fulfilled: on the one hand the confinement of the iodine compound in the target capsule during irradiation and on the other hand the discharge of the produced Xenon. A vented capsule with iodine filter is to be investigated. The upscaling of such complex irradiation procedures to industrial quantities is not obvious.

The transmutation yield depends on the thermal(ized) flux level, the loading of the reactor and residence time. One dedicated FR-incinerator could theoretically

transmute 50 kg Iodine /year while the annual production in a 100 GWe park amounts to 700kg/year.

ADS technologies with extremely high thermal neutron fluxes (10^{16} n/cm²/s) should in principle be capable of reducing the transmutation half-life. This technology is presently in the conceptual phase. However, any type of thermal neutron transmutation will be energetically very expensive.

Technetium 99

Technetium 99 is one of the most important long-lived fission products which occur in spent fuel and in several waste streams from fuel reprocessing. Due to its long half-life (213 000 years) and its diverse chemical forms in which it can occur, its radiologic significance is important if the repository surroundings are slightly oxidic. In reducing conditions it is remarkably stable and insoluble as Tc metal or TcO₂ suboxide in deep aquifers.

If Tc-99 is a real radiological hazard in some repository conditions, fully new separation technologies have to be developed. Transmutation of Tc-99 is technically possible if it is present as a metallic target, since the transmutation product is inactive Ru-100 with an isomorphous metallic phase. In a standard PWR, the transmutation "half life", i.e., the time necessary to deplete the target with 50% varies from one thermal reactor to another but amounts to about 30 years in the best conditions. The simultaneous production of Tc-99 by fission in the LWR-UO₂ driver fuel decreases the net transmutation rate and necessitates large Tc loadings. The reactor ought to be dedicated for that purpose. Moreover a dedicated reactor must be overenriched to compensate the reactivity loss due to the negative reactivity of the Tc targets in the fuel assemblies.

Taking into account the very small thermal cross section of 20 b it is important to have a high thermalized flux, a high loading in the reactor and an optimized moderator to target radius. A fast reactor with a ZrH₂ moderator, surrounding a metallic Tc-pin is the best configuration.

In a "thermalized" blanket of a dedicated FR fuelled with excess plutonium and burning MAs, an irradiation time of ~15 years is needed to deplete the Tc-target by about 50%. This transmutation half life is valid for evaluating the number of dedicated transmuters which would be necessary to absorb the annual production in the nuclear electricity generating park. The annual production in a LWR-UO₂ is ~ 20 kg/year and a moderated blanket of a 1200 MWe EFR type reactor could transmute annually 120kg. Roughly 16 units would be necessary to compensate the Tc-99 production in a 100 GWe-LWR park.

Cs-135

Caesium occurs in several isotopic forms Cs137, Cs134, Cs133 and Cs135. In terms of radiologic significance Cs137 is the major constituent of HLW. Cs135 has a very long half life (2 million years) but its radioactivity is a million times lower. Cs137: $1.1 \cdot 10^{17}$ Bq/GWe-year; Cs135: $5 \cdot 10^{11}$ Bq/GWe-year). Cs is very mobile in the geosphere if not conditioned in a suitable matrix e.g. glass.

Presently no Cs-135 separation is envisaged since isotopic separation from the highly active Cs-137 would be necessary in order to isolate this nuclide.

Transmutation to stable Cs136 in order to deplete Cs-135 is very difficult since stable Cs133 and Cs134 are present in the fission product mix and would generate during long range irradiations new Cs135.

Zr-93

Zirconium 93 is to a certain extent similar to Cs-135, since it has a very long half life (1.5 million years) but is present as a relatively small fraction (14%) of the total Zr load present in the fission product mix. Separation of Zr-93 involves the development and operation of isotopic separation procedures.

Sn-126

Sn126 has a half life of 100 000 years, is partly soluble in the HLLW from aqueous reprocessing but occurs also as an insoluble residue (similar to Technetium). Its isolation involves a special treatment of the HLLW and the use of isotopic separation techniques.

Carbon-14

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

5 Fast Reactor-spent fuel reprocessing

The reduction of the radiotoxic inventory resulting from **multiple recycling** of spent FR-MOX or FR-Metal fuel is only possible if repeated reprocessing of FR-spent fuel is part of the AFC scenario. Since irradiation of FR-MOX or FR-Metal fuel is carried out to very high burnup levels the reprocessing technology should be adapted to these new fuel conditions.

Compared to LWR-MOX fuel the target burnups of FR-fuel are 2 to 3 times higher, i.e., burnups ranging from 120 to 180 GWd/tHM are currently to be envisaged, and even higher burnup levels may be attained in a more distant future. However in order to reach an overall TRU depletion yield of e.g. 90% it is necessary to recycle the FR-spent fuel many times.

Any reprocessing campaign of spent FR-fuel based on the use of the PUREX process see **Figure 12**, or on an equivalent aqueous extraction process cannot be conveniently carried out within a short time interval after the discharge from the fast reactor because of the high decay heat.

Cooling times between discharge and reprocessing have, as a result, to be increased from 7 to 10 or more years. This way the duration of the total AFC cycle would amount to 15 or 17 years per cycle. The time interval to reach a global depletion of 90% is consequently ranging from 100 up to 250 years.

Pyrochemical reprocessing of spent FR-fuel was developed in the frame of the IFR project. In this case FR-Metal fuel irradiated at very high burnup is transferred to a “on-site” pyrochemical hot-cell for reprocessing. The molten salt bath (CdCl_2 , LiCl , KCl ...) is not subject to alpha radiation damage and much shorter cooling times can be envisaged. However this type of reprocessing technology is still in its infancy and will require extensive R&D before reaching industrial maturity in the civil nuclear sector.

5.1 Aqueous reprocessing of high burnup spent fuel

High burnup fuel means: high heat output, high initial plutonium concentrations, high alpha contaminated streams.

The first problem encountered in the reprocessing plant is the chopping of high heat emitting fuel. The heat dissipation during the FR-MOX chopping operation is roughly 10 times higher than for LWR- UO_2 fuel, for which the bundle choppers have been designed. Dismantling of the spent fuel sub-assembly followed by pin- or small-pinbundle chopping is an alternative which has been successfully tested during pilot plant test-operations at Dounreay and La Hague plants. Industrial operation would ask for adapted bundle-shears with increased cooling capacity.

The second issue which has to be faced is the high initial plutonium concentration which implies solubility problems in HNO_3 and criticality concerns. Suitable solutions to these problems have been found by installing a "residue dissolver" equipped with electrolytic generation of Ag(II)NO_3 solution and the addition of neutron absorber liquids. Complete dissolution of plutonium can be obtained at the expense of the installation and operation of an additional dissolver, a silver recovery circuit and an increased capacity of the α waste treatment plant preceding the vitrification.

The very high α concentration in these "residue"-dissolver liquids are to be diluted in the main LWR- UO_2 process stream in order to keep the overall radiolysis as low as acceptable for the extraction process with TBP and to keep the heavy metal concentration constant throughout the reprocessing campaign. Specially equipped extraction columns have been designed based on the use of e.g. hafnium as structural material and on the use of neutron poison in the fuel solutions. By choosing the proper blending of LWR- UO_2 , LWR-MOX and FR-MOX solutions, a suitable "cocktail" with a well defined composition can be processed in an adapted aqueous reprocessing plant.

The dilution of (10%) LWR-MOX and (20%)FR-MOX into the (70%)LWR- UO_2 reprocessing stream from a 100 GWe-year park increases the plutonium throughput with a factor of ~ 3.5 to 4. The mean heat output and the radiolysis of the solvent in the extraction plant increases with a factor of ~ 1.8 to 2 compared to a homogeneous LWR- UO_2 reactor park. It has consequences for the plutonium purification cycle and for the vitrification process.

The total plutonium inventory of the reprocessing plant is generally regulated by the licensing authorization ; an increase with a factor of about 4 will have consequences for the licensing of existing plants and the design of new dedicated processing plants. The main impact of coprocessing thermal and fast spent fuel relates to the radiation damage of TBP during the first and second extraction cycles. During the first cycle the presence of high concentrations of Pu-238 and Cm-244 in FR-MOX determine the degree of radiation damage to TBP. The impact of this α radiation source can be mitigated by reducing the contact time between aqueous and organic phase. Fast contactors could in principle solve this problem but their technological sophistication is a source for continuous surveillance and maintenance.

The presence of Pu-238 extends beyond the first extraction cycle and may become the limiting factor in the loading of fast reactor fuel in a mixed reprocessing strategy. In a complex reprocessing plant it may be difficult to control all the residence times of the different steps in the PUREX process without any effect on the quality of TBP as extractant.

If the MAs are kept with the fission products the coprocessing of thermal and fast fuel types will increase the heat production of the HLLW in the storage tanks, and eventually increase the heat output of the conditioned / vitrified HLW canisters. In LWR-UO₂ the contribution of MAs to the waste decay heat is only 10% while it amounts to 62% in LWR-MOX and ~75% in FR-MOX. **Table 5** gives an overview of the specific decay heats generated in different fuel types.

Table 5

Decay heats in kW/tHM of the different types of fuel 5 years after discharge

Fuel type	Burnup GWd/tHM	Total heat	Fission products	Actinides	Pu-238	Cm-244
LWR-UOx	50	2.76	2.28	0.477	0.2	0.25
LWR-MOX	50	6.56	1.97	4.59	0.46	3.35
FR-MOX	150	30.3	6.60	23.7	5.22	15.8
FR-MOX	210	33.7	8.14	25.6	5.68	17.2

In conclusion, the total decay heat influences the transport and storage of spent fuel, and the head-end operations in the reprocessing plant. The actinides in general and the Pu-238 concentration in particular interfere with the liquid extraction operations, the plutonium purification, the PuO₂ storage and the MOX fuel fabrication. The MAs and more specifically Cm-244 is the main contributor to the decay heat of HLLW during the first decades. The liquid storage of HLLW will have to take this into account.

The separation of MAs as proposed by the P&T community is a means to reduce the heat output and the toxicity of the HLLW but this proposal will face the same problems as the aqueous reprocessing technology. Several techniques which have been developed (TRUEX, DIAMEX, SANEX,...) are based on the specific extractability of some nuclides by complex organic molecules which are very vulnerable to ionizing radiation.

Multiple recycling of FR-MOX by aqueous processing of spent fuel cannot be accomplished without "dilution" of FR-MOX into a mainstream of LWR fuel. Industrial recycling of FR-MOX fuel in a dedicated FR-aqueous processing plant will be jeopardized by safety issues connected with radiolysis and criticality. Increasing the cooling time of the spent fuel does not solve the problem of radiolysis since the decay curve is determined by the half life of Pu-238 (87.7 years) and Cm-244 (18.2 years).

Multi recycling of FR-MOX fuel with aqueous processing can be carried out on an experimental base and on a limited scale. However operation at the industrial

level is not technically and economically possible. The increase of the TRU levels throughout the nuclear fuel cycle facilities, the build up of an extensive out of pile inventory of nuclear materials and the generation of secondary wastes are fundamental difficulties in the recycling of MAs or TRUs by aqueous processing methods.

5.2 Pyrochemical reprocessing

For several decades the pyrochemical and pyrometallurgical technologies have been investigated to multi-recycle the very hot nuclear materials resulting from FR-irradiation. The most important progress was achieved in the US during the IFR project and in Russia at the Dimitrovgrad facility.

Recently the Transmutation of Waste project (TWA) renewed the interest for this technology and initiated the Roadmap project which reviewed and updated the current state of the art.

The main particularity of the Roadmap proposal is the "dogmatic" requirement that pure plutonium may not be separated or handled. The process treats all the "Trans- Uranium" nuclides (TRU) as an homogeneous group of elements which have to be treated simultaneously during the chemical processing and the transmutation phase.

5.2.1 The Roadmap baseline flowsheet

The Roadmap project uses the US stockpile of LWR-UO₂ spent fuel as source term and submit it to the conventional chop-and-leach process in order to eliminate by aqueous extraction (UREX process) the bulk of uranium, technetium and iodine from the dissolved fuel mixture. (see **Fig 13**)

The raffinate of that extraction containing the fission products and the TRUs (~6% of the initial mass) is calcined and transferred to the pyrochemical section of the plant. The calcined oxide mixture of fission products and TRUs is submitted to a reduction-chlorination step with Li-LiCl and Cl₂ in a furnace at 650°C. The alkaline fission products Cs-Sr remain in the salt mixture and are treated as waste. The TRUs, Rare Earth and residual metals (Zr, Mo,...) are treated in an electrorefining furnace at 500°C. The electric potential of the Cd electrode is selected at a potential where the Rare Earth do not yet deposit. The metal concentrate of the TRU is mixed with Zr metal to produce by casting a TRU-Zr metal alloy fuel. **Figure 14** shows a general flowsheet of the pyrochemical process.

In a future scenario of multirecycling the spent TRU-Zr metal fuel discharged from an ADS transmuter is directly submitted to the electrorefining step and recycled by pure pyrochemical techniques.

The technology of pyrochemistry relies on the use of inert-atmosphere-hot-cells with a very low oxygen and or water content (1 to 10 ppm). The high temperature processes require the use of very sophisticated materials (Monel, Hastelloy, Pyrolytic graphite, Tungsten,...) because the molten salts and molten metals are very corrosive at high temperature. The proposed pyrochemical infrastructure is therefore directly connected to the fuel fabrication and transmutation facility in order to limit the number of transfers and the size of the treatment plant, and to restrict as much as possible the transfers outside the controlled atmosphere.

Online argon or nitrogen purification equipment has to keep the atmospheres of the work spaces oxygen- and moisture free. Vacuum tight levitation and transfer locks have to be installed in order to avoid diffusion of air through the plastic sleeves and joints. The chemical- and electrochemical equipment inside the hot-cells ought to be as vacuum tight as possible in order to limit ingress of very corrosive gasses in the hot cell atmosphere.

Large amounts of Cl_2 gas and chemically equivalent quantities of liquid metals (Li, Cd or Bi) have to be introduced in the chemical reduction reactor, while its exhaust contains oxygen, unconsumed Cl_2 , I_2 (I-129), Kr-99 etc. and other gaseous impurities. A very complex but efficient process-gas purification system will have to be installed and operated in order to limit as much as possible the discharge from toxic chemicals to the surrounding atmosphere.

At the cathode of the electrochemical reactor the TRUs and some Rare Earth metals are separated from the fission products. Complete separation from TRUs is theoretically impossible because some free energies of formation (Pr, Ce, Nd, and Y) are overlapping with those of Pu, Np, Am and Cm. This is not a fundamental drawback since a certain amount of Rare Earths (presumably between 1 and 10%) may be present in the TRU mixture which is recycled in a fast neutron spectrum device. (FR or ADS). Recent studies showed that the liquid Bi cathode is better suited to separate the TRU's from the bulk of Rare Earths. But very much R&D&D will be necessary to transform the present IFR "batch" process into a countercurrent extraction system operating at high temperature. Technological support in upscaling may be expected from the metal refining industry which uses similar techniques for the purification of a wide range of metals.

The waste streams discharged from the pyrochemical processes are chemically very different from the wastes produced by the aqueous reprocessing.

The LiCl salt waste is absorbed by a zeolite and submitted to hot isostatic compression (HIP technique) which operates at 850 °C and 15 bar pressure. The vaporization of Cs, one of the main fission products, has to be reduced as much as possible. The glass-ceramic matrix resulting from this process is very similar in confinement characteristics of conventional boro-silicate glass.

The hulls discharged from the UREX process and the insoluble "platinum" metals can be conditioned by melting as an homogeneous metal waste in the form of stainless steel 15% Zr. The homogeneity of this waste form has not yet been established. The same technique applies to a certain extent to hulls' waste resulting from direct pyrochemical reprocessing though the actinide contamination of these hulls' fragments is not as well documented as for hulls from aqueous reprocessing.

5.2.2 Alternative pyrochemical processes

Alternative reprocessing technologies have been investigated in the past (1957-1965) within the framework of the fluoride volatility projects aiming at the separation of U and Pu as hexafluorides from spent LWR-UO₂ fuel. The major drawback of this approach was the partial volatilization of gaseous PuF₆ decomposing to solid PuF₄ and co-volatilization of some fission products e.g. ZrF₄, TcF_x and RuF_x.

The most important recent (1996) technological demonstration of pyrochemical reprocessing was performed in Russia where a sequential electrolysis process in Li-KCl was demonstrated. In this process the spent FR-MOX fuel is chlorinated by gaseous Cl₂ gas in a liquid bath of LiCl-KCl at 600-650°C. After extraction of UO₂ by electrolysis at the cathode, the salt bath is oxidized leading to the precipitation of PuO₂. It is planned to incorporate the separation process of Np into this flowsheet in order to recycle TRU –Pu-Np fuel into the BOR-60 reactor. This flowsheet is not applicable to Am-Cm.

5.3 Nature and management of irradiated TRU and MA targets

a) Spent FR fuel types

In a complex reactor park including dedicated FR and ADS reactors to burn Plutonium resp. MAs or a mixture of TRUs, the equilibrium composition of the spent fuel is very different from conventional LWR-UO₂ discharged from the present nuclear power plants. **Table 6** shows the comparative composition of Plutonium, MAs, and the bulk of fission products as well as the decay heat after 5 years cooling expressed per GWe-year.

Table 6

Bulk composition and decay heat of spent reactor cores per GWe-year

	LWR-UO ₂ 50 GWd/tHM	LWR-MOX 50 GWd/tHM	FR-MOX-(CAPRA) 210 GWd/tHM
tHM/GWe-y	22.12	22.12	5.26
Pu total kg	240	1320	2067
Pu-238 kW	4.75	34.4	30
MA total, kg	34.1	155.5	155
Np	16	4.21	21.6
Am	16.4	126.6	129
Cm	1.74	24.73	25.7
MA, kW	4.80	70.6	74
Cm	4.76	61.25	65.88
Fiss. Prod. kg	1133	1107	1127
Gaseous FP	200	186	
FP, kW	50.4	43.5	42.8
Total heat, kW	61	145	146

For intermediate storage and waste disposal it is important to note the equivalence of LWR-MOX and FR-MOX in terms of MA inventory and decay heat dissipation per unit produced energy. However the fuel mass generating the heat is 3 times smaller for FR-MOX and consequently the specific heat per weight of fuel is three times higher having direct consequences for the cooling times in the reactor building, the design of transport flasks and the frequency of transportation. However the plutonium inventory of the fuel drastically increases (a factor of 5.5) when it is recycled in a LWR-MOX and increases still further with a factor of 8.6 when it is used in a FR-burner. From fuel cycle management it is a transfer of the long-term radiotoxic TRUs from the waste, in case of direct disposal, to the reactor cores. However this "concentration" of the plutonium inventory has strong consequences for the fuel cycle facilities if multi-recycling is carried out indefinitely. The reprocessing and fuel fabrication plants are directly involved in this strategy. The waste treatment plants will have to handle much larger quantities of intermediate- and alpha waste. But in an ideal P&T strategy the waste to be disposed of will become an almost pure Fission Product deposit with a as low as possible actinide concentration.

b) Minor actinide targets

In most of the present P&T scenarios, aqueous reprocessing of spent fuel precedes the partitioning of MAs, their purification and their transformation into irradiation targets. According to the type of reactor used, and the duration of the irradiation period, different kind of targets will be obtained. These targets will ultimately be discharged from the reactor and either recycled to a dedicated processing plant or disposed of for interim storage preceding geological disposal.

Table 7

Composition of irradiated MA targets from LWR, FR and ADS reactors

	LWR	HFR	FR		ADS
	THERMAL		EFR FAST	CAPRA (therm)	MYRRHA FAST
EFPD	2460	800	4500	1500	800
Cycles	X		X		
once-thr.		X		X	X
Np residue	58%	6.4%	40%		76.5%
FP	7%	75.9%	24%		10%
Pu-238	35%	6.9%	~30%		12.8%
Pu total	~40%	12.6%	36%		12.9%
Am residue	42%	4.3%	37%	2%	76.5%
FP	10%	55.4%	22%	80%	7.7%
Pu238	~20%	17.2%		3.8%	10.2%
Pu total	--	27.3%		12.1%	11.1%
Cm244	20%	8.4%		2.38%	2.3%

The data in **Table 7** show that very high thermal or thermalized fluxes are necessary to deplete MA targets. **The HFR calculations and experiments have shown that the thermal flux is most efficient in "burning" the MAs in the targets.** The thermal cross sections of Np-237 and Am-242 (transmutation product of Am-241) are 50 to 150 times higher than the fast cross section and explain this result. The results for the once-through irradiation in the thermalized blanket of an FR (CAPRA) fully confirms this approach. **The neutron economy in a fast neutron spectrum is much better than in a thermal one and FRs or fast ADS have to be used in order to produce as efficiently as possible the neutrons necessary for the transmutation.** The pure fast neutron spectrum of an ADS requires much longer irradiation times but can eliminate in the end the residual longterm TRU content from a thermal irradiation campaign.

5.4 Effects of longterm policy changes on radioactive waste inventory

The most important aspect associated with P&T is the long-term implication of the decisions taken. Once a given path has been traced and the first steps have been made it is difficult not to say impossible to eliminate the consequences.

The **once through cycle** is reversible as long as an irreversible disposal in a deep geologic repository has not been realized i.e. until shaft closing. As long as the repository is filled but not sealed-of, the process of changing the initial decisions is yet very expensive but not impossible within a period of say 100 years. However the fuel cycle facilities which are necessary to reopen the cycle are in principle not available and have to be recreated. The case of the US is to a certain extent typical for this situation. The civil reprocessing was abandoned in the early seventies and a certain form of reprocessing (UREX+Pyrochemistry) has to be re-introduced in order to eliminate the TRUs from the HLW.

The **reprocessing fuel cycle** as developed industrially in Europe reduces the Pu content of HLW, but transfers it to the MOX fuel. The vitrified HLW contains the long-lived FPs and the MAs which constitute the longterm radiotoxicity but do not involve a criticality hazard. By transferring the plutonium to the MOX fuel the nuclear community has bought some additional decades to take the decisions which have longterm impact. A stabilization of the Pu inventory at the current level can only be obtained if an important (~20%) part of the reactor park is equipped with FRs or equivalent fast neutron spectrum facilities. They are not existing and it will take several decades before a viable fast reactor industry has been built up. The disposal of LWR-MOX could therefore favourably be delayed till the next generations decide how to equip their nuclear park.

Since reprocessing of LWR-UO₂ leads to the production of HLLW which has to be stored in tanks for a number of years, decisions have to be taken how to proceed further. Separation of MAs has to be decided within the interval between the reprocessing and the vitrification. If the separation is performed the question arises how to carry on with the separated nuclides.

In most repository conditions only Np-237 is among the MAs a longterm radiological source term, but Am-241, being the parent of Np-237 has to follow the same sequence of decisions. The radiologic impact on the biosphere of Np-237 in vitrified HLW is non-existent during the physical lifetime of the glass i.e. ~10,000 to 100,000 years. But once conditioned as glass the source term remains untouchable forever. Partitioning of MAs and conditioning into a stable ceramic form and storage in retrievable conditions is therefore an opportunity to keep the decisions on this source term within the hands of the future generations without increasing the risk. If possible and efficient, transmutation, aiming at "incineration", could be undertaken in the future when dedicated burner reactors are available.

Reprocessing provides "access" to some fission products which are important radiological source terms: I-129, Tc-99. The existing technologies allow to partition these nuclides and to condition them in appropriate matrices to be disposed of in specially suited repository conditions e.g. I-129 in salt dome and Tc-99 in a stable reducing host rock. Transmutation of these nuclides is nearly impossible to perform economically. However once separated and retrievably stored any decision on their fate could be reversed.

The **advanced fuel cycle with P&T** incorporated is without any doubt the most comprehensive approach. It requires very important and dedicated fuel cycle and reactor facilities which go far beyond the presently existing nuclear technology. Especially the transmutation approach calls for the development of FR-burners and ADS facilities which may take 20 to 30 years before these are industrially available. But this option is the only one which offers a final solution for the plutonium problem.

A serious problem occurs when the recycling is interrupted after e.g. 50 or 100 years. In this option the enrichment of plutonium in the nuclear energy generating plants will have reached an equilibrium level and the whole inventory is bound to be disposed of at that time. **Recycling of TRUs in a composite park of nuclear reactors, comprising LWRs and FRs, depends on a long term energy policy with a continuous political and economical backing of nuclear energy in the global energy mix.**

The impact of this "interruption" scenario on the repository design is far reaching: the radiotoxicity of the nuclear fuel streams is multiplied by several orders of magnitude, the heat dissipation requirement is much higher and the effect of the longterm radiotoxicity reduction is not attained unless after several hundred years, determined by the decay time of Pu-238.

The actinides which have a direct impact in long-term waste management are the plutonium isotopes (238,239,240,242), ^{237}Np , ^{243}Am and ^{245}Cm , because of their long half lives. However due to the interrelation between the actinide decay chains $4n$, $4n+1$, $4n+2$ and $4n+3$ short lived mother isotopes e.g. ^{241}Pu , ^{241}Am , ^{243}Cm and ^{244}Cm influence the composition of the long-lived actinide mixtures, long time after the decay period of the mother isotopes.

The most important decay chain (see **Fig 15**) from radiologic point of view is the $4n+1$ decay chain comprising ^{245}Cm , ^{241}Pu - ^{241}Am - ^{237}Np . The $4n+3$ decay chain includes a.o. ^{243}Cm , ^{239}Pu and the $4n$ chain ^{244}Cm - ^{240}Pu (see **Fig 16**).

The determination of a target decontamination factor depends on the time period in which the radionuclide mixture is considered (1000-10 000 years), the composition of the high-level waste (depending on the burn-up i.e. 33-60 MWd/tHM) and the moment when the reprocessing operation is scheduled: 3 to 10 years for direct treatment and 50 to 100 years for delayed reprocessing.

There are two time intervals which are generally considered 1 000 years, being the time limit for the canister materials and 10 000 years the limit of the "technical lifetime" of a repository structure.

If the reprocessing period is scheduled between 3 and 10 years the contribution of ^{241}Pu as source of ^{241}Am - ^{237}Np is limited to the quantity initially present after discharge. However when reprocessing occurs after 50 to 100 years 90 to 100% of ^{241}Pu has been transformed into ^{241}Am and the ^{243}Cm and ^{244}Cm isotopes have during this time interval practically decayed to their daughter products ^{239}Pu and ^{240}Pu .

6 Impact of advanced fuel cycles with transmutation on waste management

6.1 Radiotoxicity evolution, hazard and risk

The radiotoxicity evolution of spent fuel expressed in Sv/THM is very well known and depends on the type of fuel: LWR- UO_2 , LWR-MOX, FR-MOX, FR-metal, and the attained burnup. During the first 150 years the fission products Cs-137-Sr-90 determine the radiotoxicity and the heat emission. From 150 years till 250,000 years the Plutonium isotopes are the main contributors to the radiotoxicity. Beyond 250,000 years Np-237 emerges together with the daughter products of uranium.

The total radiotoxicity of HLW from reprocessed LWR- UO_2 is during the first 150 years almost equal to that of spent fuel. The α radiotoxicity of vitrified HLW depends on the precise moment when the reprocessing takes place and on the achieved decontamination factors for the Pu isotopes. With a half life of 14.2 years the decay of Pu-241 determines to a great extent the concentration of Am-241 in the HLW. Once the waste has been vitrified the radiotoxicity will decrease with the half life of Am-241 (432.7 years). Beyond 4000 years the radiotoxicity of residual Pu and gradually that of Np-237 prevails.

The recycling of Pu into LWR-MOX fuel generates a very radiotoxic spent fuel type which up to now remains in the interim storage till disposal or could be recycled if a second era of FR-MOX would become an economic reality within a few decades. Since the decay heat of this fuel is much higher than that of LWR- UO_2 the cooling period must be prolonged anyway. The radiotoxicity of this fuel follows the cumulated decay curves of Cm-244, Pu-238, Am-241, Pu-239, Pu-240 and Pu-242. The other α nuclides e.g. Am-243, Cm-245 and Np-237 are in this context negligible.

The α radiotoxicity of FR-MOX fuel is ~3 times higher taking into account the increased Pu concentration (from 8.2% to 22%) in the fuel of a number of FRs

which operated during several decades (DFR, PFR, PHENIX, SUPERPHENIX). But the radiotoxicity expressed per unit of energy produced is nearly the same.

The radiotoxicity of conceptual fuel of fast burner reactors (CAPRA) and accelerator driven transmutation systems is undoubtedly an order of magnitude higher. (up to 42% Pu and burnups of 210 GWd/tHM).

The notion and significance of the term "radiotoxicity" has been challenged in the waste management community as not being related to the essential risk criterion for the biosphere which is: **dose to man**. In this context the water soluble fission products I-129, Cs-137, Cs-135, Se-79 and Sn-126 are the most important nuclides from radiological point of view due to a combination of toxicity, half-life and concentration.

The most toxic of the fission products is I-129 which has a toxicity ($1.1 \cdot 10^{-7}$ Sv/Bq) similar to that of the actinides. Cs-137 one of the main fission product in HLW has a toxicity which is 10 times lower ($1.3 \cdot 10^{-8}$ Sv/Bq). The other soluble fission products are still less toxic by two or three orders of magnitude compared to the actinides. Some fission products e.g. Tc-99 and Pd-107 have long half-lives but are only slightly soluble in chemical reducing media representative of deep geologic repository conditions. The behaviour of Tc-99 in oxidic conditions (Yucca Mountains) is an exception.

The toxicity of the spent fuel and HLW, coupled to a very long half-life is the main concern of the public and a showstopper for the deep geologic disposal. The involved hazard is very substantial but the risk, except for I-129, is small.

An advanced fuel cycle with **partitioning** followed by "improved" **conditioning** of some nuclides substantially decreases the risk, but not the hazard. Separation of MAs before vitrification and special conditioning of these nuclides in e.g. ceramic matrices with very low solubility in water is an opportunity which would offer substantial advantages in the reduction of longterm risk.

On the other hand such practice does not reduce the radiotoxic inventory and its associated hazard. **The reduction of the actinide inventory, or its mean half life is the only possibility to reduce significantly the longterm hazard.** In order to achieve this goal only nuclear processes are capable of modifying the nature of the isotope involved either its half life or its decay scheme.

The most drastic hazard reduction is the **fissioning process** of higher actinides decreasing the intrinsic radiotoxicity by a factor of 100 to 1000. This nuclear process is preferably carried out with fast neutrons.

But simultaneously any neutron interaction with higher actinides leads to **neutron capture** and formation of other higher actinides which may fission again. In case the resulting half life is shorter the specific radiotoxicity of the target increases proportionally. An easy example is the transmutation of Np-237 leading to the formation of Pu-238 in an inverse relation with their respective half lives; the resulting half life is reduced from 2,000,000 years to 87.7 years and the

radiotoxicity of the irradiation target increases with a factor of $2,140,000/87.7=24100$. In a waste management option the risk of this irradiation product, which is less soluble and shorter lived, is lower than the initial product but the radiotoxicity is 4 orders of magnitude higher.

The reverse can also occur e.g. with Cm-244 which occurs in FR-MOX fuel at high burnup. A separated Cm target is very difficult to handle and decays after 18.1 years for 99.9% into Pu-240 with a half life of 6560 year. By neutron irradiation a Cm-244 target would partially be transformed into Cm-245 with a half life of 8500 years having a slightly reduced radiotoxicity.

In the perspective of disposal of radioactive waste with a guaranteed technical lifetime of 1000 years for the canisters and 10,000 years for the repository structure and its confinement, partitioning of MAs followed by advanced conditioning is in the present state of the technology the preferred solution. It has to be thoroughly investigated for which nuclides transmutation is a beneficial process and which transmuter would provide the "cleanest" end product. In the **partitioning-conditioning scenario** the partitioned fractions remain available for possible transmutation in the future if this technology breaks through.

Transmutation with emphasis on fissioning (incineration) can without reservation be defended as technology to reduce the actinide inventory and consequently its radiotoxicity and risk.

This is undoubtedly the case for plutonium isotopes with a high fission cross section as well in FRs, as MOX or metal fuel, as in ADS, as metal or nitride fuel.

But transmutation of a well defined actinide element (pure Np, Am or Cm) separated from HLLW by partitioning would transform it again into a mixture of actinides and fission products, with very different chemical and physical properties, which would become similar to spent fuel (see **Tables 6 and 7**).

If a **transmutation-incineration** process would produce an end product with a significant TRU reduction (factor 10 to 100), this endeavour will reduce the disposal risk for workers only in the case of **inadvertent human intrusion** into a repository.

6.2 Impact of MA transmutation on waste management operations

During the active phase of the advanced nuclear fuel cycle with P&T an important increase of the waste treatment, storage and disposal operations is expected. The high α level of the process and waste streams will strongly affect the radiological and economical impact of these additional operations.

6.2.1 Neptunium processing and transmutation

The following facilities will be necessary to produce and process the irradiation targets

- Neptunium chemical purification plant with a capacity of 3.2 t per year
- Neptunium target fabrication facility of equivalent capacity.
- Interim safeguarded storage of Np targets.
- Dedicated processing rig for irradiated targets.
- Hot refabrication plant for recycling Np-237- Pu-238 targets.
- High level waste connection with LWR-UO₂ vitrification plant.
- Medium level α waste treatment plant for effluent processing.

This complex ought to be associated to one of the big industrial reprocessing plants to avoid unnecessary transfers and transport on the public roads.

A dedicated FR or ADS facility should be erected in the immediate vicinity.

It is not possible to estimate the size of such irradiation facilities which will depend on the "incineration" rate in the dedicated FR or ADS reactor system. Very long irradiation periods are necessary to achieve a sufficient Np-237 depletion.

Alternatively one might favour a high flux thermal ADS or a thermal "island" in a fast flux ADS facility, which is equivalent to the irradiation in an HFR. It would convert ~95% of the Np-237 into fission products, Pu-238 but it would leave a residue containing some (~16%) higher actinides.

6.2.2 Americium processing and transmutation

The separation of Am from HLLW would realistically produce a waste stream containing between 90 to 95% Am-Cm and 5 to 10% Rare Earth. The facilities to separate, condition and transmute the separated Am-Cm stream are still much more sophisticated than those for Np-237. The following partitioning plants are necessary to implement this option.

- α - γ -n shielded Americium-Curium chemical purification plant
 - Additional Rare Earth separation
 - Chemical purification
- Americium – Curium target preparation
- Interim storage of conditioned targets
- α - γ -n Dedicated hot recycle rigs for irradiated targets
- Disposal of once through irradiated targets
- Am/Cm separation facilities
- Dedicated Cm storage facility for Pu-240 recovery.
- Connection to HLLW vitrification

- High and medium α waste treatment plant

As described in sections 3.2 and 4.4.2 the industrial implementation of Am-Cm transmutation needs a highly sophisticated infrastructure. In a first stage, group-separation of the Am-Cm-RE fraction contains up to 50% RE and has to be treated in order to reduce the RE content to e.g. 10 or 5% which is tolerable in a FR-MOX reactor. The Am-Cm stream still contains chemical impurities which have to be eliminated in order to prepare a suitable target matrix for irradiation.

Fully gamma shielded facilities are required to handle the effluent from the preliminary separation since the REs are strong gamma emitters. Once the bulk of REs has been removed the concentrated Am-Cm stream needs hot-cells with heavy neutron shielding.

Am-Cm targets contain significant amounts of Pu-238, Cm-244 and Fission products. Due to this combination the irradiated capsules are very difficult to handle and to process. Therefore one alternative consists of carrying out a once-through irradiation in a thermal island of a Fr-MOX reactor till 200 dpa (limit of capsule/target pin cladding) in order to eliminate further processing.

According to this scenario the Am inventory could be eliminated for 98% by irradiation during ~20 years. The capsules from such a test if they resist to the intense radiation and the high He pressure have to be overclad to avoid leakage inside the storage facilities. The transformation of the Am target into a mixture of 80% fission is possible in a target type capsule but cannot be extrapolated to industrial quantities since in this case the global impact of the depletion in the target and the generation in the driver fuel has to be taken into account.

The transmutation of Am-Cm is a technical challenge in its operational phase but the radiological impact and its influence on waste management is rather limited. Once an inventory has been transformed (almost) completely in a fission product mixture the management of it is identical to "actinide-free" vitrified HLW. The main longterm benefit is the elimination of Am-241 as parent of the very long-lived and slightly mobile Np-237 and its partial (~12%) transformation into some longlived plutonium isotopes.

In order to provide the necessary neutrons to transmute a fertile Am-241-Am-243 mixture it is obvious that ADS systems could be more efficient than critical FRs in which the driver fuel will be a generator of Am-Cm. From safety point of view the loading of the ADS reactor is more flexible since the void reactivity coefficient remains negative even with very high TRU loadings.

6.2.3 Thermal impact of MA transmutation on an advanced fuel cycle strategy

In a complex reactor park with 70% LWR-UO₂ 10% LWR-MOX and 20% FR-MOX reprocessed 5 years after discharge and decaying in 50 years which may be estimated at

$$1520 \text{ t LWR-UO}_2 \times 1 \text{ kW/t} = 1.52 \text{ MW}$$

The non-reprocessed LWR-MOX fuel contributes to the heat as follows:

$$224 \text{ t LWR-MOX} \times 6.56 \text{ kW/t} = 1.47 \text{ MW}$$

If this spent fuel type is recycled to the FR-MOX and submitted to advanced reprocessing with partitioning the heat contribution would be reduced to

$$224 \text{ t LWR-MOX} \times 2 \text{ kW/t} = 0.45 \text{ MW}$$

The discharged FR-MOX fuel reprocessed after 5 years contributes in the following order of magnitude:

$$153 \text{ t FR-MOX} \times 8.14 \text{ kW/t} = 1.24 \text{ MW}$$

Irradiation of Np and Am targets will increase this output:

$$1.6 \text{ t of Np-237 transforms into a heat source of 190 kW}$$

$$1.7 \text{ t of Am-Cm transforms into a heat source of 620 kW}$$

In a "all reprocessing" option the contribution of the MA transmutation would increase the total annual heat output by ~ 20%.

7 General conclusions

- **The present waste management for already vitrified HLW has to be considered as an irreversible process for which disposal in geologic strata is the most advisable and unavoidable solution**
- **Spent fuel discharged from nuclear power plants should be stored in engineered facilities as long as there is no definite choice of longterm management of long-lived actinides. Retrievable storage in underground facilities is an alternative which could have its merits as a medium term policy.**
- **Conventional reprocessing of spent fuel is a necessary step in the reduction of the actinide content of HLW. The recycling of Pu and U into the MOX-fuel cycle is a transient solution to reduce the volume of actinide loaded waste.**
- **Spent LWR-MOX fuel with its high actinide content should be stored in engineered facilities till a safe fast reactor technology becomes available.**
- **Advanced reprocessing of (high-burnup) spent fuel with removal of U, and TRUs and production of "actinide-free-vitrified-HLW" is in the medium term the most defensible waste management option.**

- **The aqueous partitioning technology could be upscaled to industrial levels within the next 15 years. Conditioning and /or target fabrication of Minor Actinides may be developed in parallel.**
- **Final disposal of actinide-free (or -poor) HLW in geologic strata (salt, clay) is fully acceptable if the actinide content has been reduced with two (or more) orders of magnitude.**
- **Transformation of separated minor actinide concentrates into a ceramic type of waste form is beneficial from radiologic point of view even if the deep disposal is the last resort. The sharply reduced solubility (compared to glass) reduces the long term environmental risk. Transformation into a matrix which could also be used as a future nuclear fuel- or target form is the most versatile option.**
- **Transmutation of MAs in power reactors does not sufficiently decrease the MA inventory and leads to an accumulation of higher actinides with a drastic increase of the radiotoxicity of the actinide inventory.**
- **Transmutation in a fast neutron reactor facility is the only possibility to incinerate the overall actinide (Pu+MA) inventory and to make use of the large amounts of plutonium present in spent LWR-MOX fuel. However the current FR technology with sodium cooling can, for safety reasons, not be considered for that purpose. The use of less dangerous metallic coolants (e.g. Pb-Bi) is to be investigated. Multiple recycling of plutonium in critical FRs is the most economic way to reduce the accumulated plutonium inventory of LWR-MOX. This option asks for the development of new reprocessing technologies.**
- **Pyrochemical reprocessing techniques appear as almost indispensable to carry out multiple recycling of high burnup FR fuel within short cooling times. However their technical implementation at industrial level may become very difficult to accomplish.**
- **Critical fast reactors with an adapted coolant system could also be used as "storage reactors" for Minor Actinides and reduce significantly their inventory during a long irradiation period in a thermalized blanket.**
- **Separated Minor Actinides can theoretically be "incinerated" in accelerator driven transmutation systems (ADS). But a long range R&D effort will be necessary to develop the system from its present conceptual design stage to an industrial reality.**
- **High proton accelerators exist as high-energy-physics tools, but not as industrial type facilities for continuous operation. The number of beam trips must be decreased (~1000 times) and the current density increased with two orders of magnitude.**

- **The connection between the proton accelerator operating under vacuum and the subcritical reactor core operating under cover-gas pressure requires the development of a replaceable beam window.**
- **The subcritical core design implies a lot of investigations on the homogeneity of the neutron flux, the burnup reactivity swing, the criticality control and the in-core actinide fuel management.**
- **Dedicated transmutation facilities (FR or ADS) will have to be erected near the large reprocessing plants (La Hague, Sellafield) in order to reduce as much as possible fuel recycling and transportation of highly radioactive materials.**
- **Development of dedicated transmutation facilities (refurbished FR and/or ADS) may take at least 25 years to reach the mature industrial level. The operation of a combined P&T prototype facility on a single site may possibly start in the second quarter of the 21st century.**
- **In a nuclear phase out scenario, the ADS technology may have a very important role to play in order to reduce the residual Pu+MA inventory resulting from discharged spent FR-fuel.**

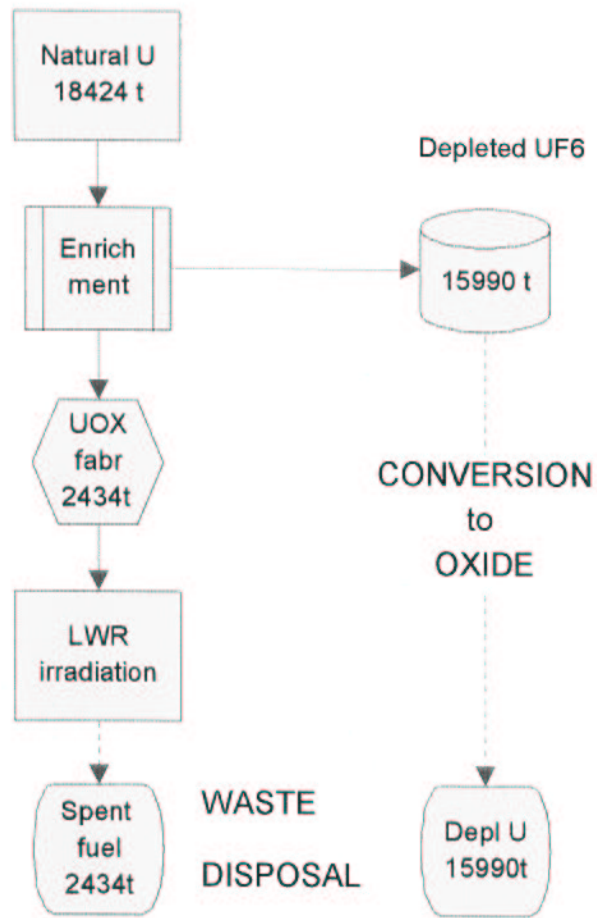


Fig 1 Once-through fuel cycle

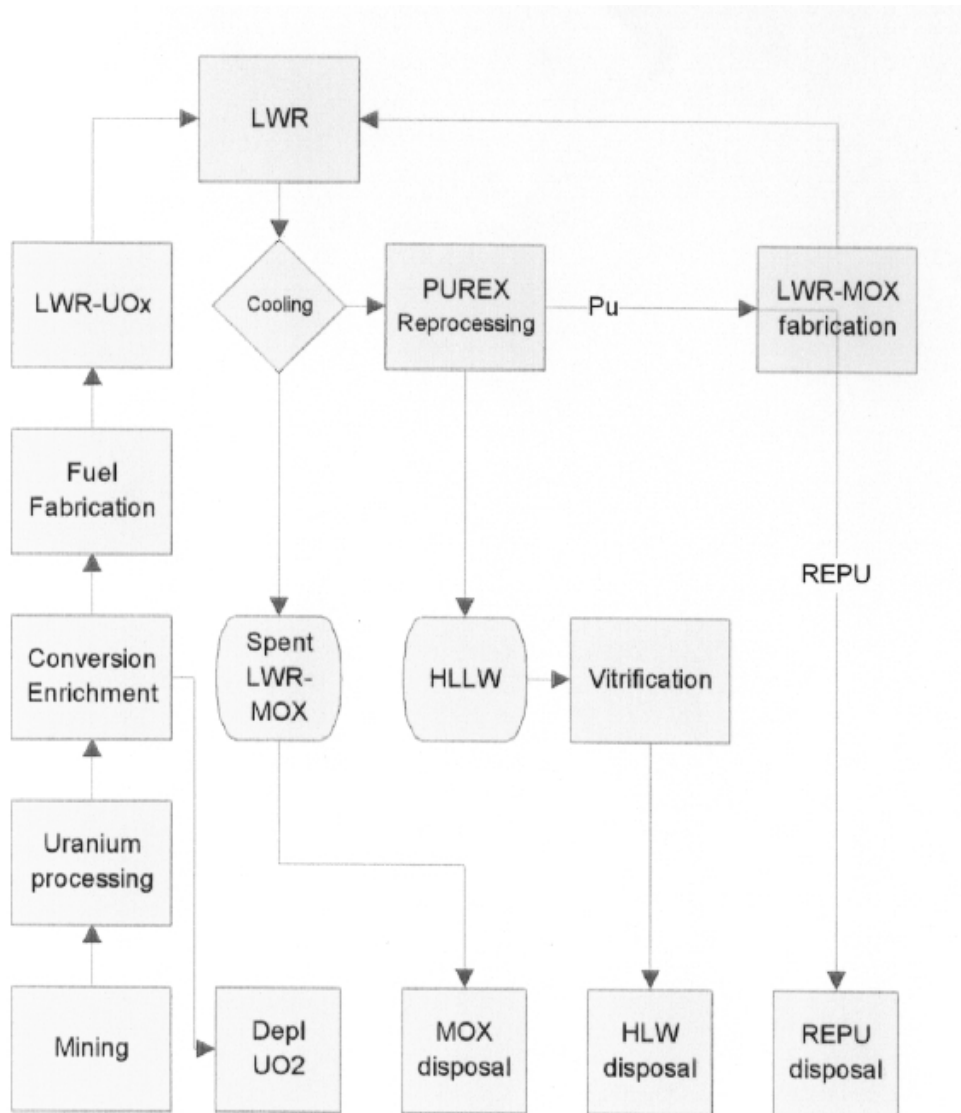


Fig 2 LWR-reprocessing fuel cycle

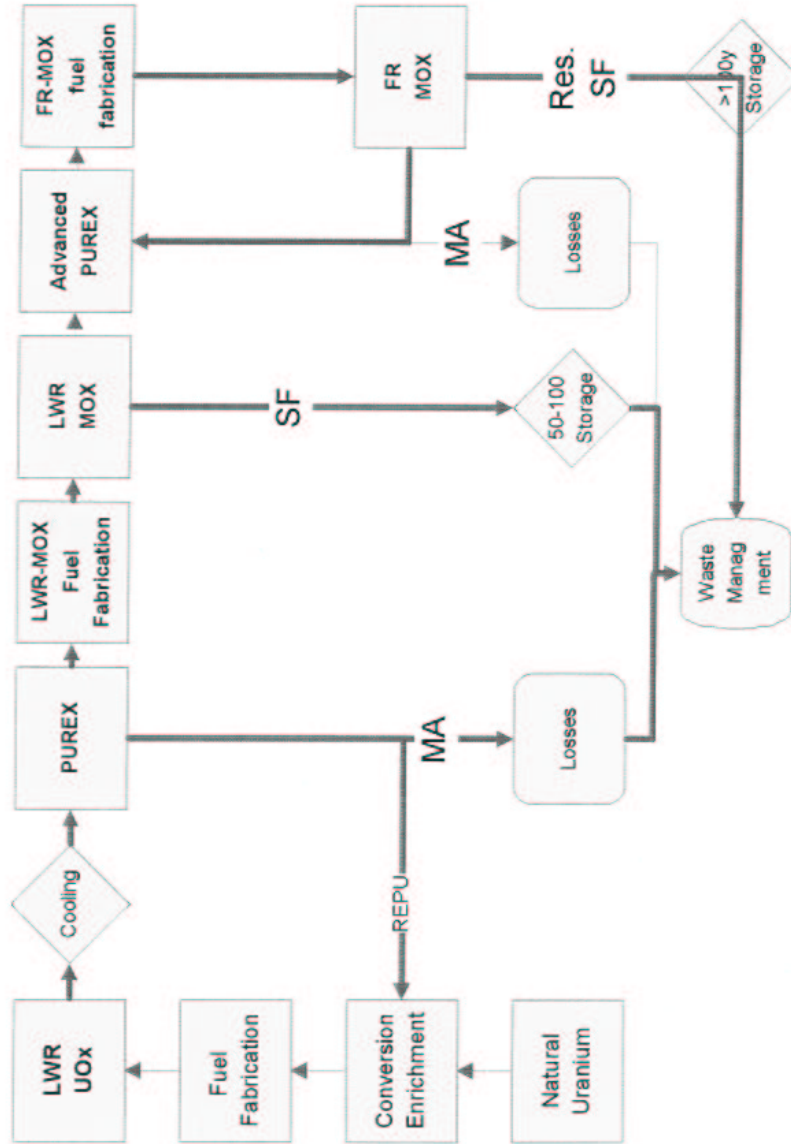


Fig 3 Conventional LWR-FR-MOX fuel cycle

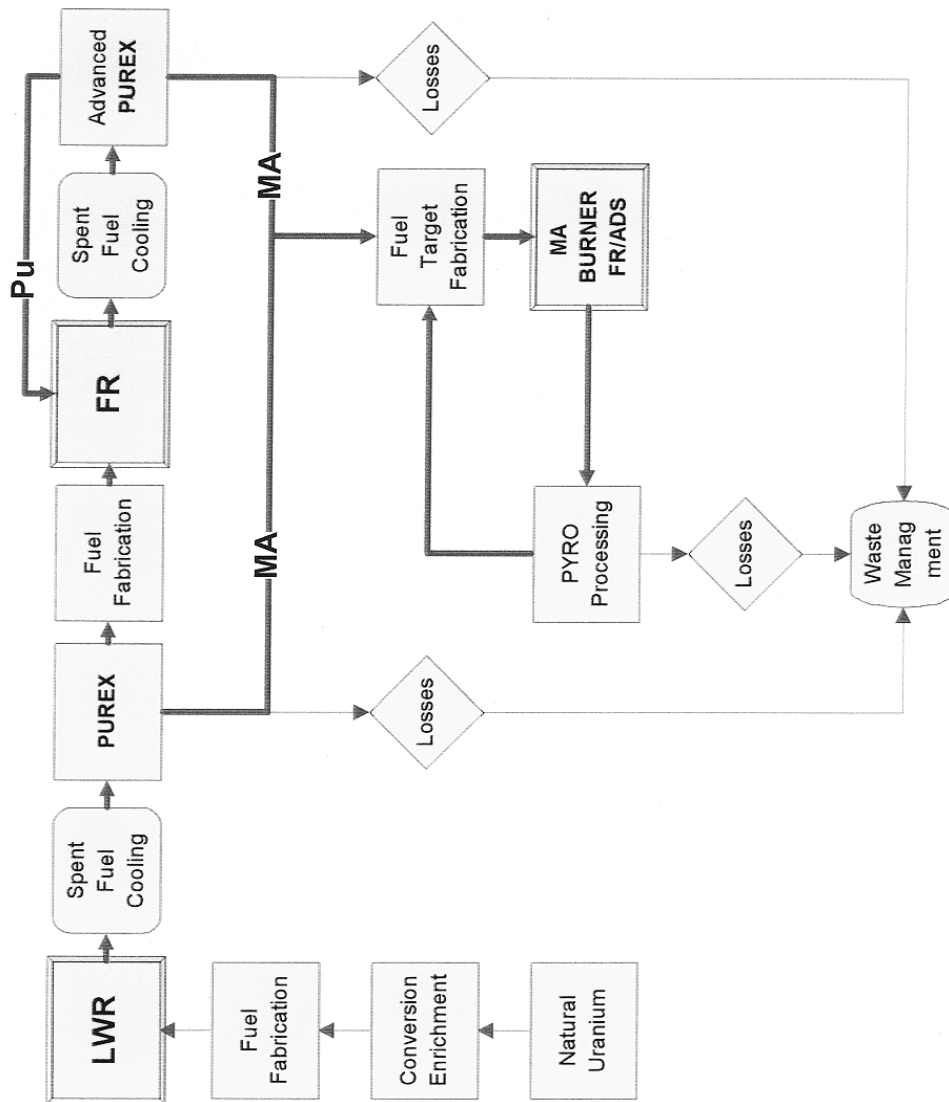


Fig 4 Double strata fuel cycle

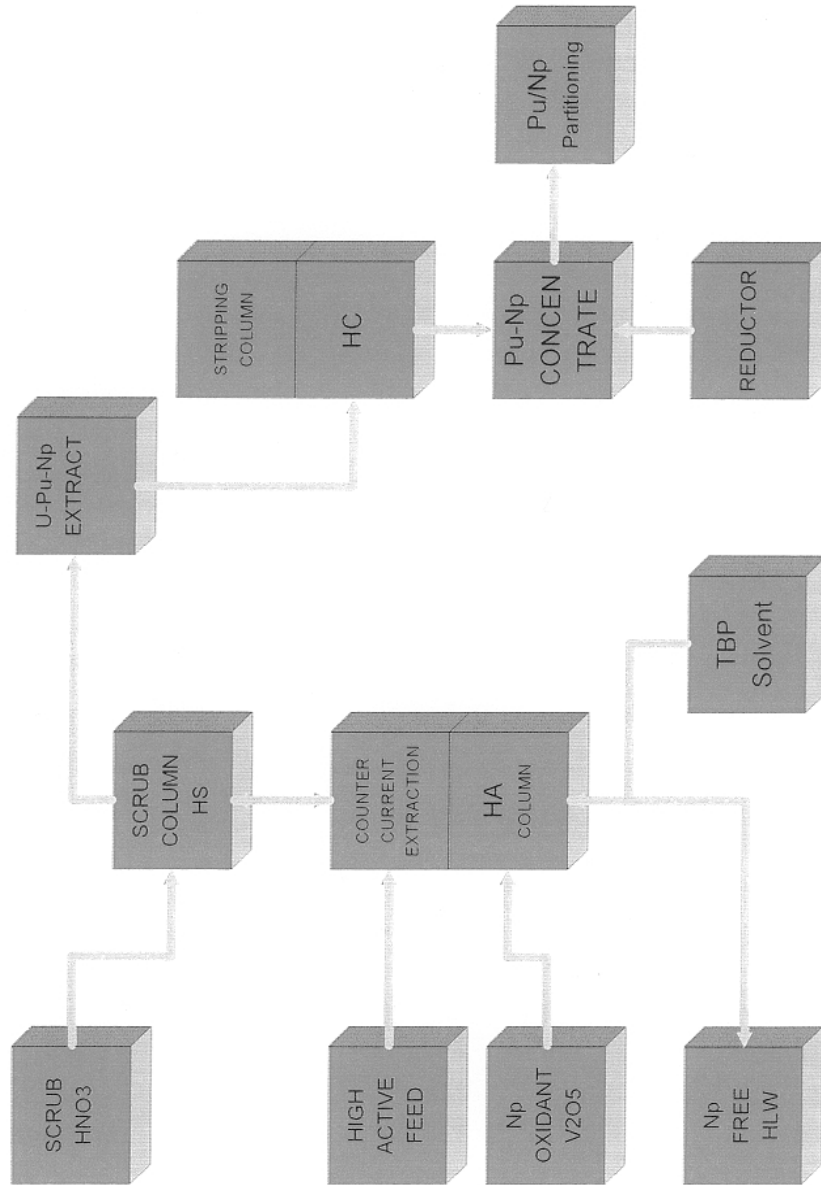


Fig 5 PUREX conceptual flowsheet with Np recovery

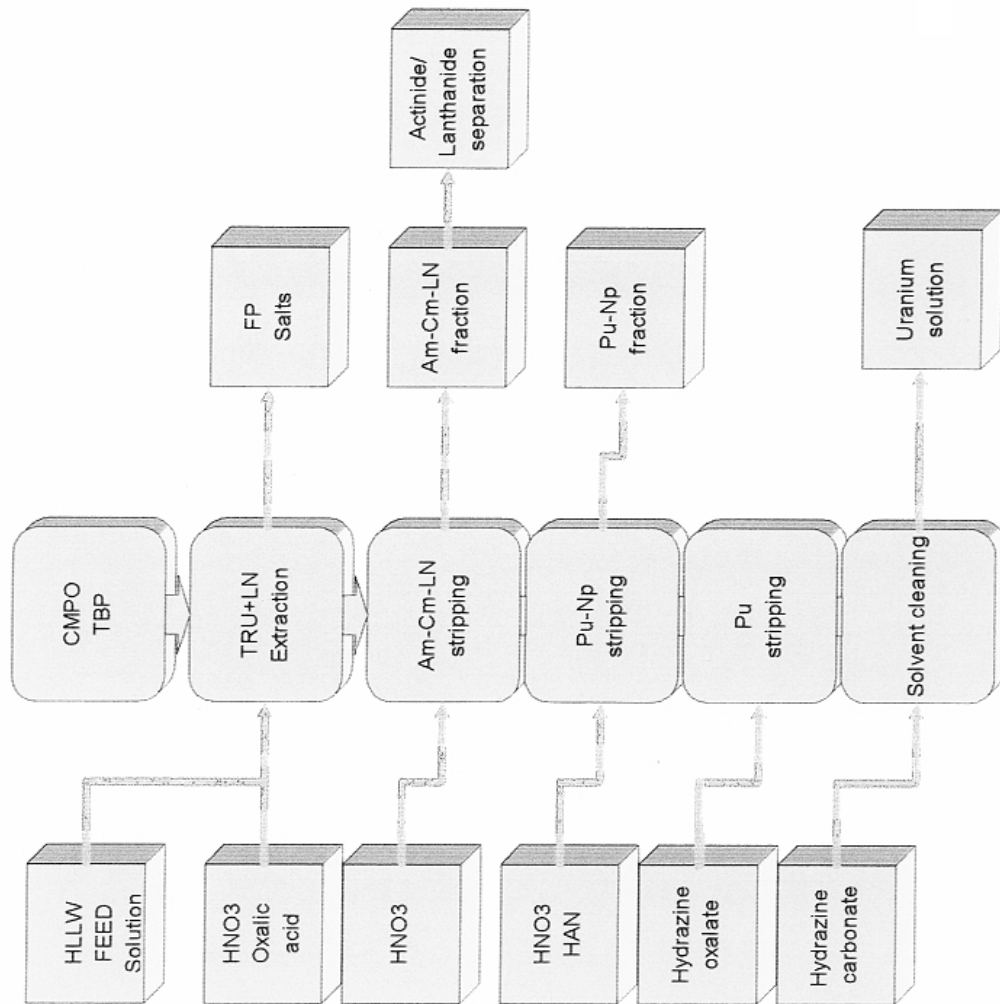


Fig 6 TRUEX flowsheet for minor actinides extraction

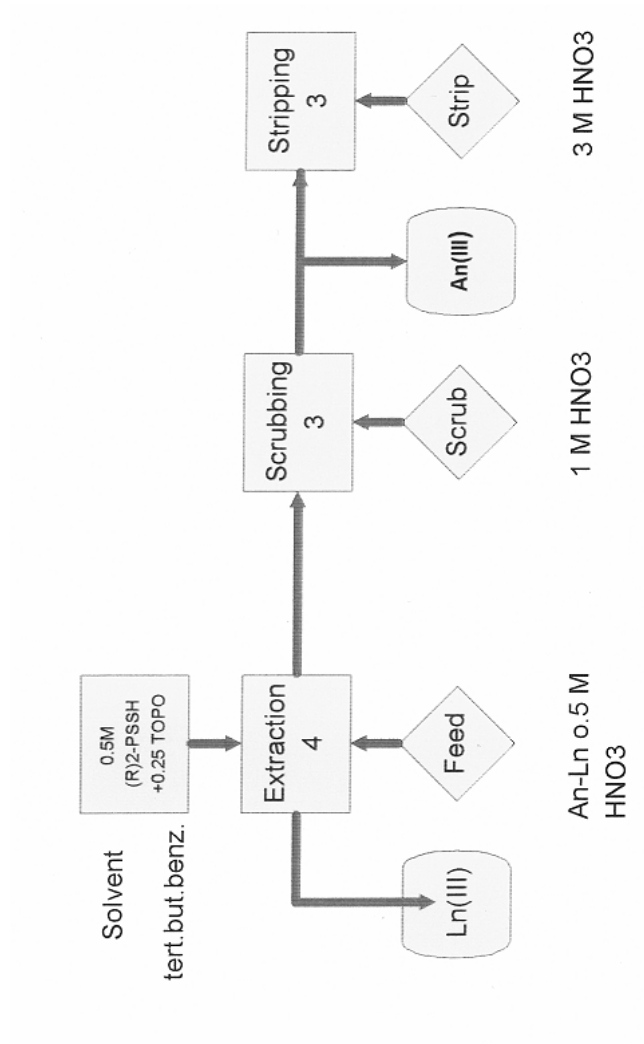


Fig 7 "ALINA" Lanthanide-actinide separation process

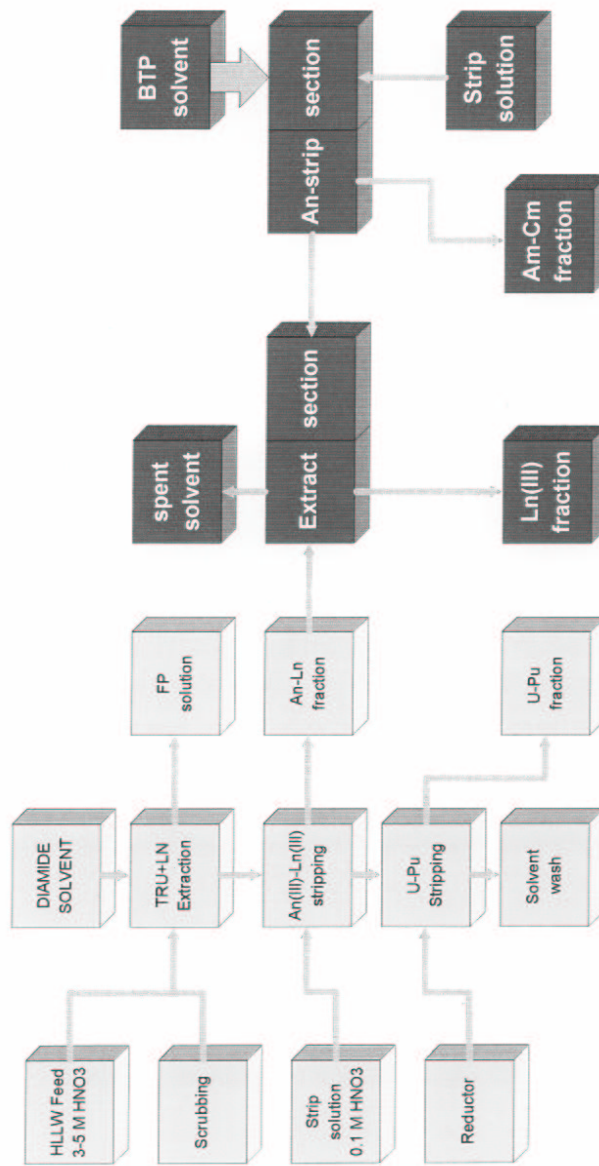


Fig 8 Conceptual DIAMEX-SANEX flowsheet for MA recovery from HLLW

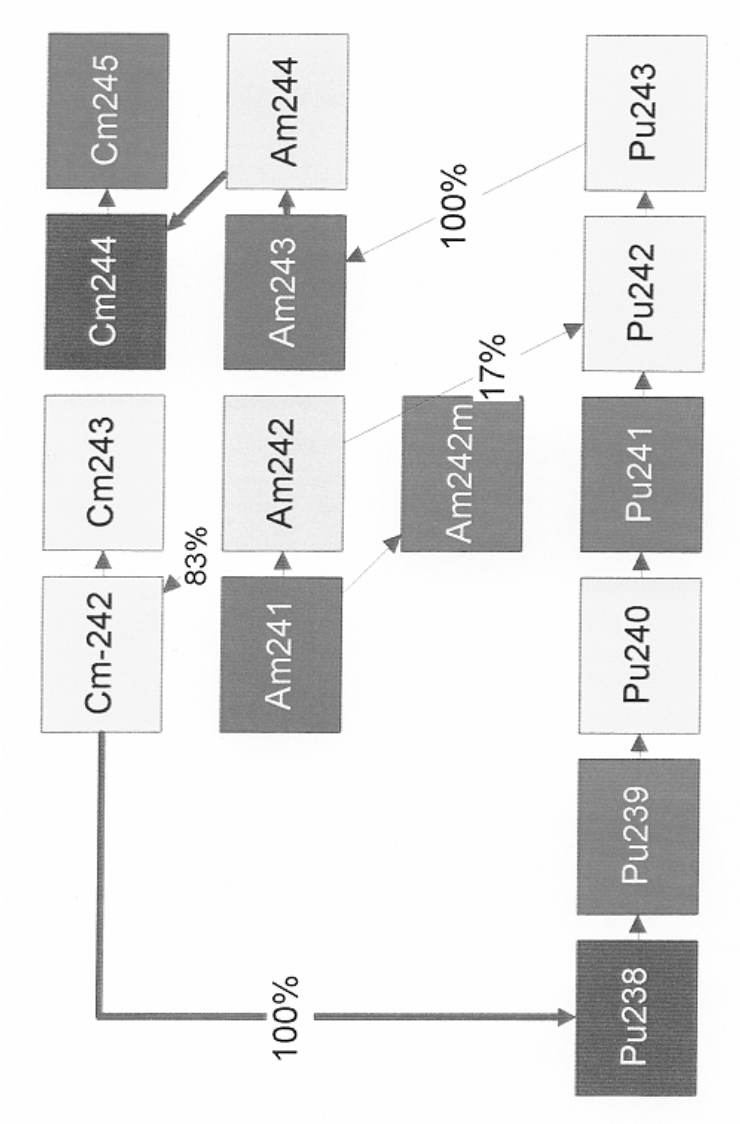


Fig 9 Americium irradiation scheme in a thermal neutron flux

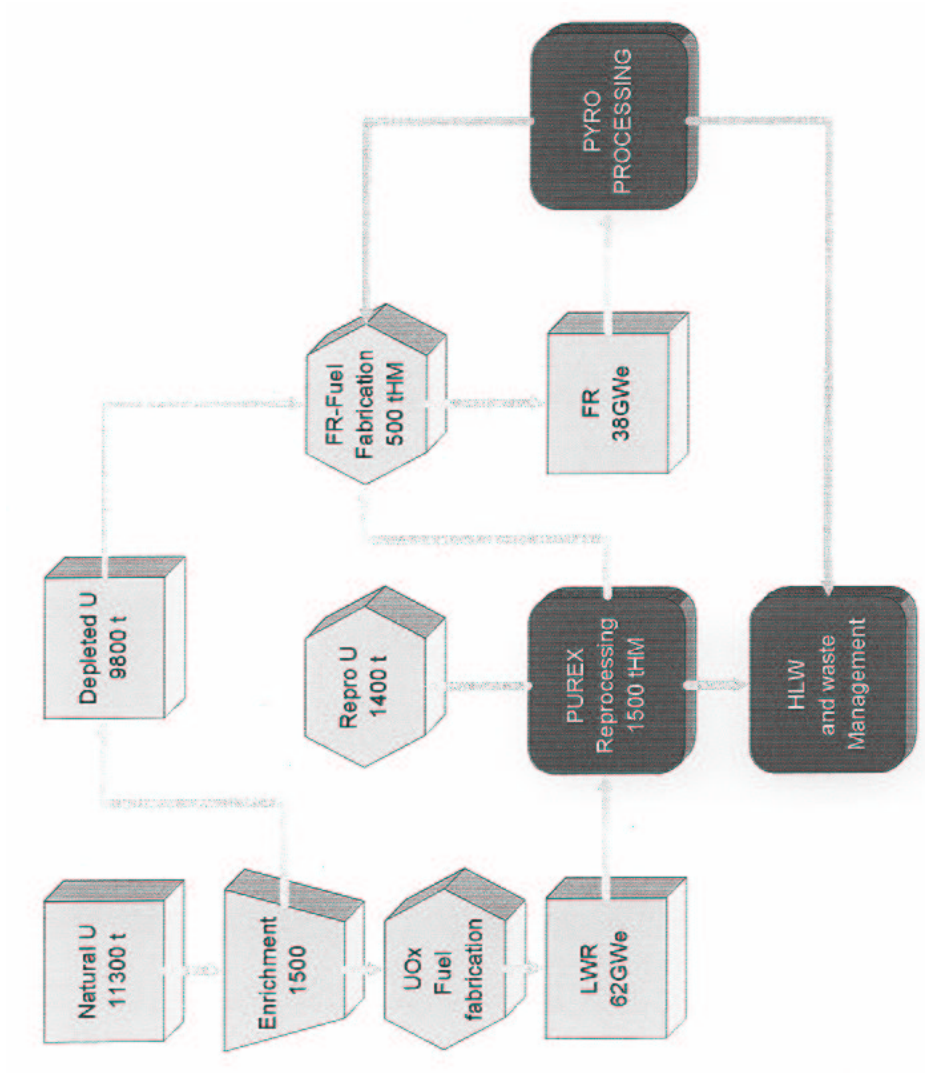


Fig 10 Schematic reactor park in a LWR-FR reactor strategy

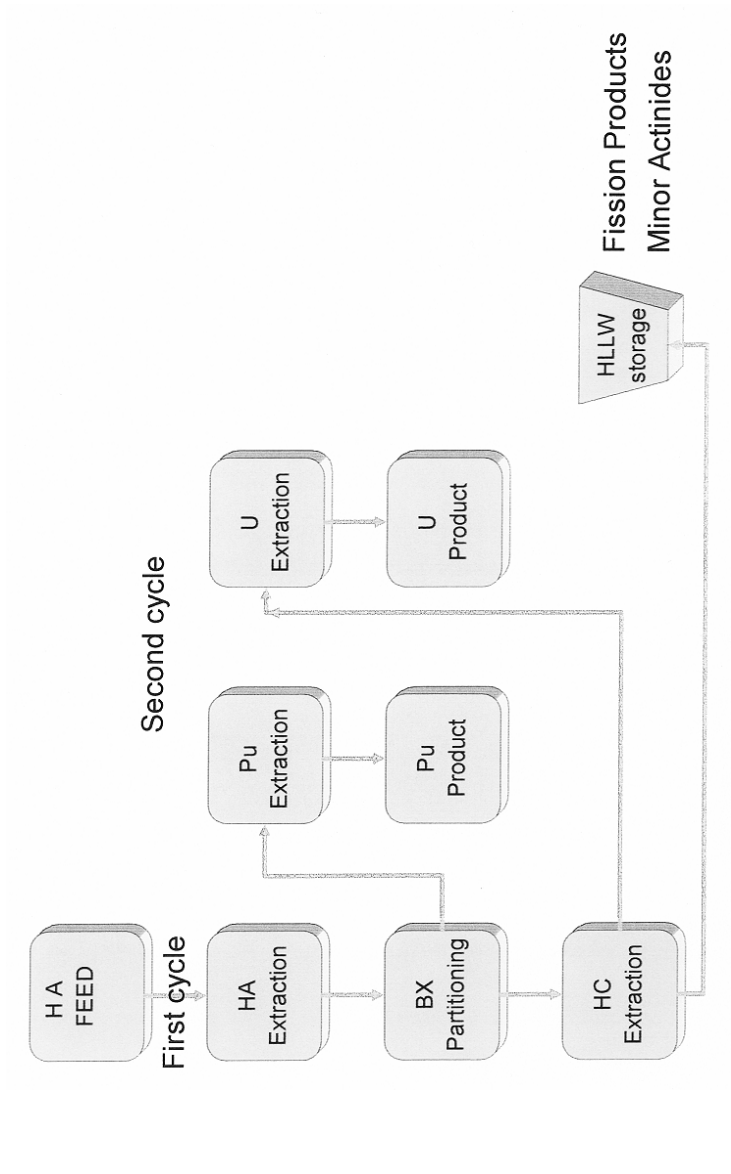


Fig 12 Schematic conventional PUREX reprocessing scheme

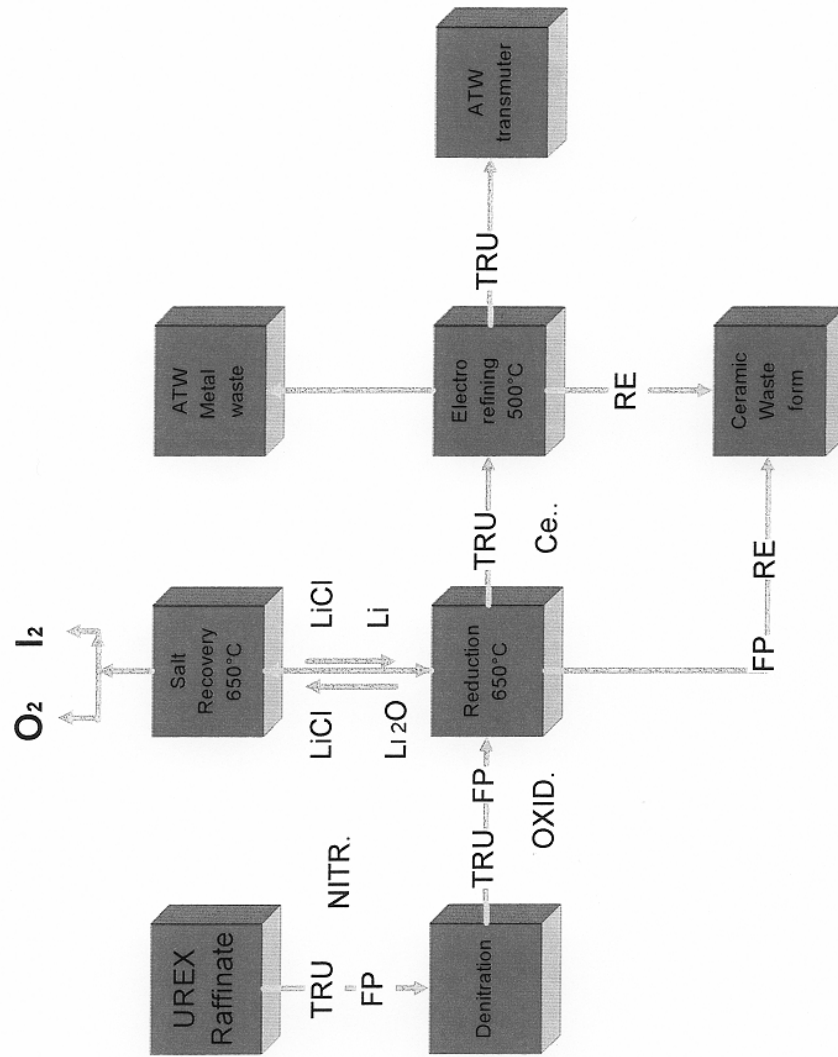


Fig 14 Pyrochemical reprocessing scheme

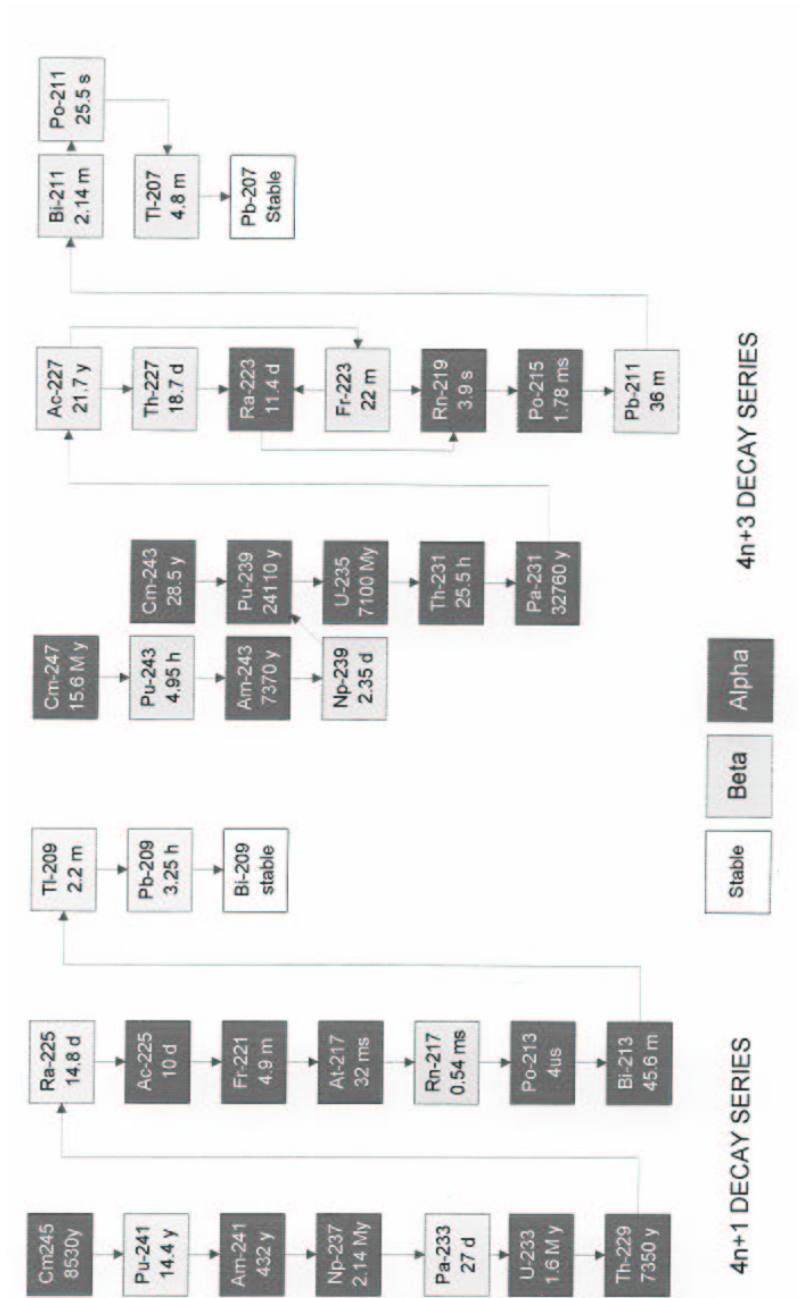


Fig 15 Radiochemical decay chains 4n+1 and 4n+3

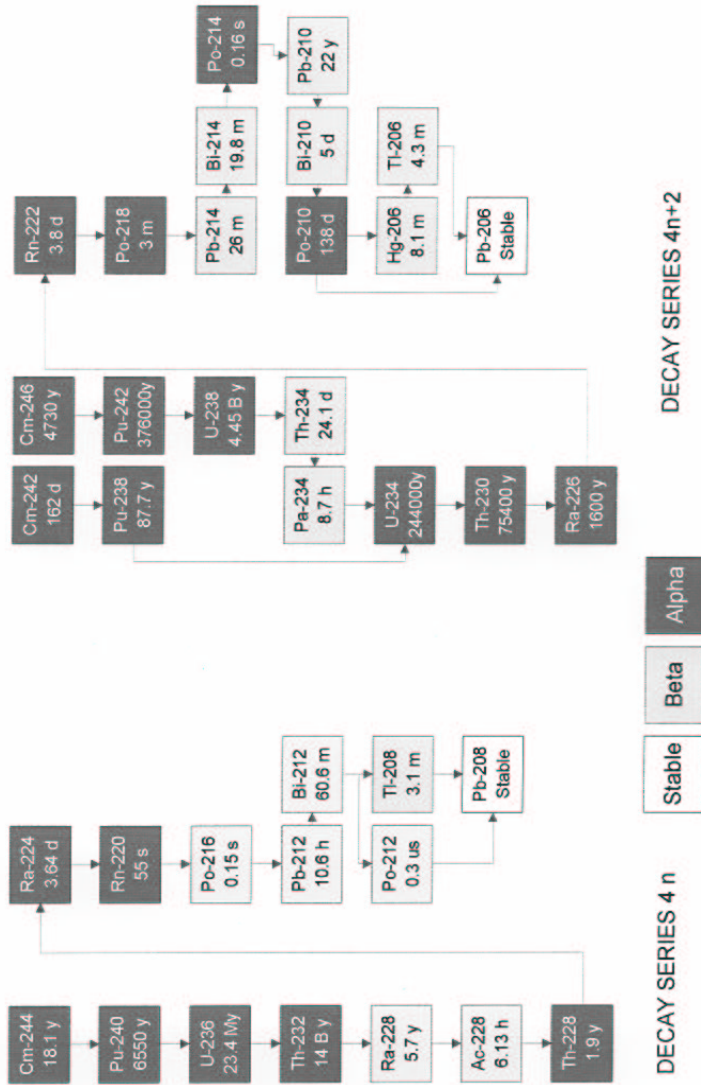


Fig 16 Radiochemical decay chains $4n+2$ and $4n$

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