

## **CLOSING THE NUCLEAR FUEL CYCLE: ISSUES AND PERSPECTIVES**

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### **Abstract**

Partitioning and transmutation (P&T) aims at making nuclear energy more sustainable from the viewpoint of the back-end of the fuel cycle by minimising the high-level waste with respect to its mass, radiotoxicity and (possibly) repository risk. P&T mainly deals with the management – i.e. transmutation and/or special conditioning and confinement – of minor actinides and fission products, but involves the closure of the fuel cycle for plutonium as a necessary first or parallel step. The conditions for a completely closed fuel cycle, the goals for transmutation, and the implications for the reactor and fuel cycle technology are overviewed and discussed, and the currently favoured transmutation strategies are compared with respect to achievable waste radiotoxicity reduction and impact on the releases of potentially troublesome actinides from a repository for vitrified high-level waste.

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## 1. From the open fuel cycle to the semi-closed fuel cycles for plutonium management

The LWR once-through fuel cycle at a mean burn-up of 50 GWd/t<sub>HM</sub> produces (a) spent fuel consisting of fission products (1.1 t/GWe-a), irradiated uranium, plutonium, and minor actinides (about 20 t/GWe-a, mostly uranium), and (b) depleted uranium from the enrichment process (about 170-190 t/GWe-a, depending on the <sup>235</sup>U concentration of the tails).

If these materials are not further utilised, they have to be considered as nuclear waste. The preferred option is currently to store the irradiated fuel elements after appropriate cooling in suitable geological formations. The LWR once-through fuel cycle has the advantage that it avoids the difficulties of reprocessing; however, it can only extract about half a percent of the energy content of the mined uranium.

Depleted uranium stored as UF<sub>6</sub> is a chemical hazard and becomes radiotoxic in the long-term. Therefore, it has to be transformed into a more stable form (e.g. UO<sub>2</sub> or U<sub>3</sub>O<sub>8</sub>) and appropriately stored for use in future breeder reactors, or adequately disposed. In the nuclear waste discussion, not much attention has yet been given to the management of the increasing stocks of depleted uranium.

### 1.1 Incentive for reprocessing

The incentive to reprocess irradiated LWR fuel arises primarily from the desire to improve the *uranium utilisation* and implies the recycling of the bred plutonium (about 250 kg/GWe-a) in MOX-LWRs or future high-conversion and fast-spectrum reactors. Multi-recycling of uranium and plutonium in LWRs in the so-called self-sufficient mode would allow the uranium utilisation to improve by about a factor of two, but involves a higher <sup>235</sup>U and plutonium enrichment due to the “degradation” of the uranium and plutonium isotopic composition. A much better (close to 100%) uranium utilisation could be achieved with fast reactors (FR).

Industrial reprocessing is currently based on the PUREX process which allows “clean” uranium and plutonium to be separated from the fuel and was initially developed for military applications. The remaining high-level waste (HLW), consisting mostly of fission products and minor actinides, is converted into a stable form for ultimate disposal, with the normal method being the storage of vitrified HLW in geologic repositories.

The separation of uranium and plutonium from the spent fuel has the advantages of reducing the actinide mass and the plutonium content of the HLW. In combination with vitrification, it minimises the risk of a clandestine recovery of fissile material from a waste repository; however, it does not significantly reduce the long-term toxicity of the HLW. Drawbacks are the extra investment in complex technology for reprocessing and  $\alpha$ -active fuel fabrication, and the potential proliferation risk associated with the handling of pure fissile materials. Since the balance of advantages and drawbacks depends on regional boundary conditions and political factors, there are currently contradicting policies regarding the recycling of plutonium.

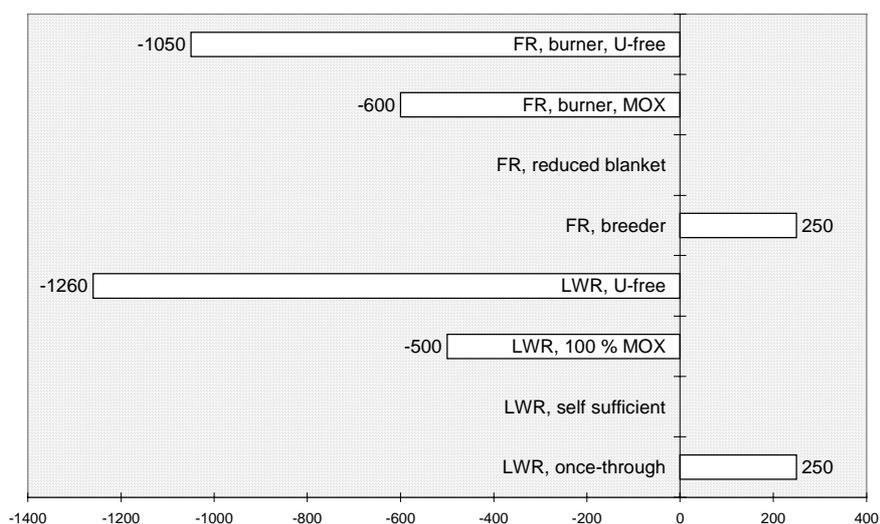
### 1.2 Plutonium stock management

In some countries, an early commitment to industrial reprocessing, combined with the delayed introduction of fast reactors, has led to large stocks of separated plutonium. The available fabrication capacity for LWR-MOX fuel of about 300 t<sub>HM</sub>/a, however, will now allow some 25 tPu/a to be recycled, which balances the current output of the reprocessing facilities [1].

Relative to the self-sufficient recycling mode, which has a zero plutonium balance, the plutonium consumption of an LWR can be enhanced by increasing the number of MOX fuel elements in the core, up to a full MOX core. An even higher plutonium consumption – up to the theoretical limit indicated in Figure 1 for uranium-free concepts – could be achieved by reducing the uranium content of the fuel. However, it should be noticed that LWRs alone cannot completely burn plutonium because the buildup of the even isotopes in a thermal neutron spectrum constrains the number of recycles to two or three at most. The remaining degraded plutonium has to be disposed, or stored until it can be utilised in fast reactors, which offer similar net consumption rates as the LWRs (cf. Figure 1).

It is obvious that plutonium stocks can be managed effectively with LWRs and fast reactors; new types of burner reactors or reprocessing methods are not needed, but plutonium could, of course, also be managed with other types of reactors. The respective issues, including fuel developments, have been discussed in the framework of working parties and workshops of OECD/NEA [2,3] and in many international conferences. It should be emphasised that the closure of the fuel cycle for plutonium is a prerequisite for, but not a direct issue of P&T. Therefore, plutonium management as such is not in the focus of this paper.

Figure 1. Net plutonium production of different reactor types (kg/GWe-a)



## 2. Fully closed fuel cycles with P&T

P&T aims at making nuclear energy more sustainable from the viewpoint of the *back-end of the fuel cycle* and implies the separation and further utilisation of valuable materials as well as the minimisation of the remaining HLW with respect to its mass, radiotoxicity and (perhaps) repository risk. It thus responds to current concerns of the public and politicians who are not satisfied with a radiological hazard which extends over millions of years, although the associated long-term risk in terms of annual individual dose is very small.

Figures 2 and 3 show the radiotoxicity of the HLW produced for a 120 GWe-a scenario after reprocessing and the resulting annual individual dose, estimated for vitrified waste emplaced in crystalline host rock [4]. Evaluations for other scenarios and repository concepts give comparable results [5,6] and confirm the following general observations:

- From the viewpoint of the *radiotoxicity*, which plays a role in accidental intrusion scenarios, P&T must first be concerned with the actinides, particularly the minor actinides americium and neptunium, the toxicity of the fission products (shaded in Figure 2) laying at least two orders of magnitude below that of the actinides after  $10^3$  years.
- From the viewpoint of the *long-term risk* of a geologic repository, the relatively mobile fission products are more important than the actinides, the fission products  $^{135}\text{Cs}$ ,  $^{79}\text{Se}$ ,  $^{99}\text{Tc}$  and  $^{126}\text{Sn}$  being dominant dose contributors in vitrified HLW scenarios<sup>3</sup>.
- The fission product risk peaks in the time span  $10^4$  to  $10^6$  years after the closure of a repository, whereas an actinide risk arises “only” after one million years.

## 2.1 Goal for minor actinide mass reduction

Figure 2 shows that the radiotoxicity of the actinides requires more than ten-thousand years to decay to the toxicity level,  $U_{\text{nat}}(\text{LWR})$ , given by the consumed natural uranium. With a hundred-fold reduction in the actinide content, this goal could be reached already after a few hundred years. The fact that the “natural toxicity” level for a pure fast reactor strategy,  $U_{\text{nat}}(\text{FR})$ , is about hundred times smaller than that for an LWR strategy speaks also for the goal of a reduction in the actinide content of the HLW by a *factor of 100*.

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

## 2.2 Goal for actinide recovery

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

$$M^{\text{W}} = \delta L M^{\text{F}}$$

where  $M^{\text{F}}$  is the total mass of actinides fissioned,  $L$  is the actinide loss fraction during reprocessing and fuel fabrication, and the burn-up factor,  $\delta$ , can be evaluated from the fraction of heavy metal fissioned,  $B$ , as  $(1 - B) / B$ . Under equilibrium conditions,  $M^{\text{F}}$  equals the top-up fuel mass,  $M^{\text{T}}$ , which, in general, can be divided into the mass,  $M^{\text{B}}$ , of transuranic or minor actinides to be burnt (i.e. transmuted and ultimately fissioned), and a diluent, usually consisting of fertile uranium (normal critical burner cores are not suited for fertile-free top-up fuel).

<sup>3</sup> The long-lived fission product  $^{129}\text{I}$ , which is known to dominate the repository risk in direct storage scenarios, is not present in vitrified HLW because it is discharged to the sea during reprocessing. Since sea disposal will not be a desirable feature of a “clean” nuclear energy,  $^{129}\text{I}$  is also a candidate for P&T.

Figure 2. Radiotoxicity of LWR spent fuel after uranium and plutonium separation

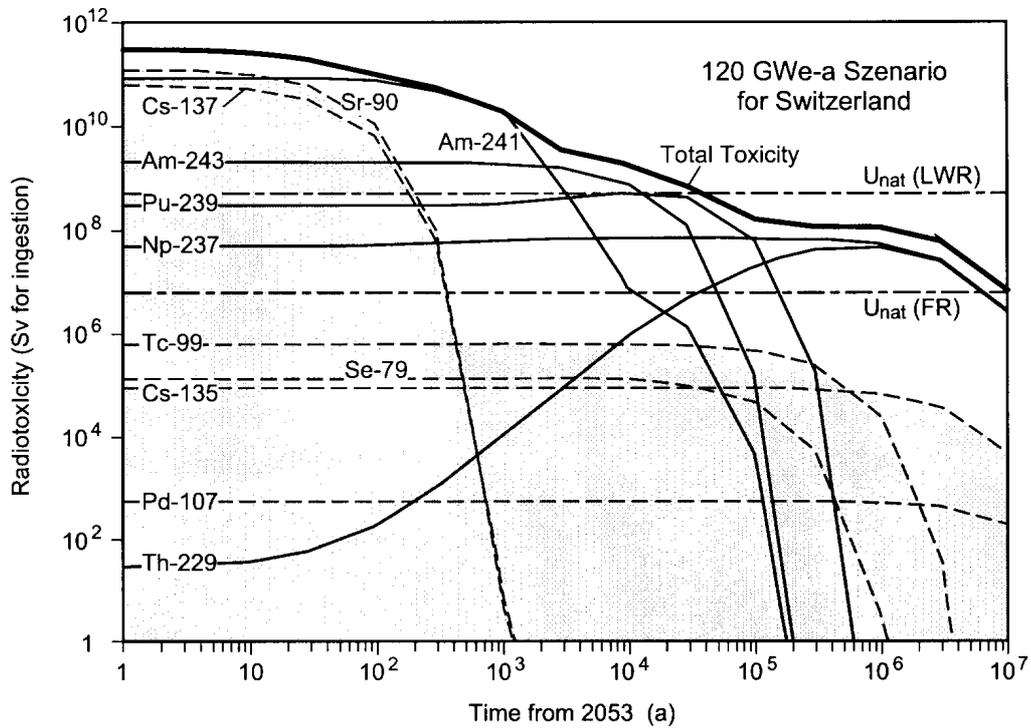
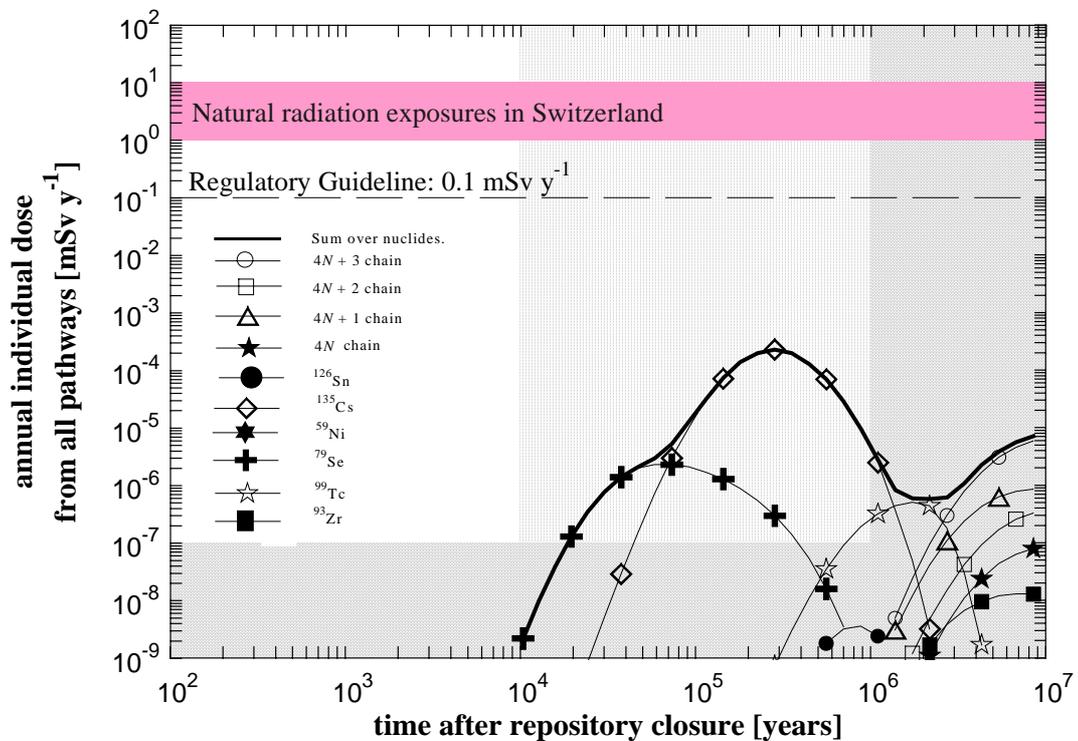


Figure 3. Annual individual dose for vitrified HLW emplaced in cristalline host rock



Denoting the transuranic (TRU) or minor actinide (MA) fraction of the top-up fuel,  $M^B/M^T$ , by  $\tau$  and the “waste mass reduction factor”,  $M^B/M^W$ , by  $R^M$ , one obtains the simple expression

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

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

### 3. Neutronic requirements for fully closed fuel cycles and role of the ADS

For neutronic reasons, not all reactors can operate with a fully closed fuel cycle. To assess the suitability of a reactor in terms of neutron multiplication, the production-to-absorption ratio of the actinides in the equilibrium core,  $\eta_{ec}$ , is a useful parameter. Alternatively, the overall neutron balance for the complete fissioning of actinides can be measured in terms of the “fuel neutron production parameter” –  $D$  [7].

An  $\eta_{ec}$  value smaller than 1 means that the fuel of the equilibrium core cannot maintain a chain reaction; a negative  $-D$  value indicates that an actinide or an actinide mixture cannot be completely fissioned. It can be shown that the parameters are mainly influenced by the neutron spectrum and flux of the system and that the two approaches lead to the same conclusions.

The  $\eta_{ec}$  and  $-D$  values in Table 1 refer to realistic reactor concepts including an ATW-type sub-critical thermal transmuter, a CAPRA-type fast plutonium and MA burner, a critical (sodium-cooled) fast TRU burner, a sub-critical (LBE-cooled) fast TRU burner, and a dedicated sub-critical MA burner. For the actinide feed, plutonium and TRU mixtures separated from PWR spent fuel and the MA mixture from the first stratum of a typical “double strata” fuel cycle strategy [8] are assumed. The values demonstrate that minor actinides cannot be completely burnt in thermal systems and that fast plutonium and TRU burners offer more surplus neutrons than the respective thermal systems. The surplus neutrons could be used for burning fission products.

Table 1. Neutronic performance of plutonium, minor actinide and transuranics burners

Actinide feed <sup>a</sup>	Thermal (ADS)		Fast (critical)		Fast ADS	
	$\eta_{ec}$	$-D$	$\eta_{ec}$	$-D$	$\eta_{ec}$	$-D$
Plutonium	1.15 <sup>b</sup>	0.40 <sup>b</sup>	1.64	1.18	1.80	1.34
Minor actinides	0.89	-0.37	1.28	0.71	1.33	0.79
Transuranics	1.11	0.30	2.00	1.52	1.75	1.29

<sup>a</sup> Plutonium and TRU from PWR spent fuel with a burn-up of 50 GWd/t<sub>HM</sub>, minor actinides from a park with PWR-UOX reactors (70%), PWR-MOX reactors (10%), and CAPRA reactors (20%).

<sup>b</sup> A MOX-PWR with self-sufficient plutonium recycling has a similar neutron economy.

### 3.1 Core design constraints

In practice, the design of a TRU or MA burner core, like that of any reactor core, is not only constrained by the above-mentioned basic neutronic criterion, but also by performance and safety parameters, such as the reactivity swing during burn-up, coolant void reactivity effect, Doppler coefficient, effective delayed-neutron fraction, etc. In particular, for a sodium-cooled fast reactor core, the substitution of normal MOX fuel by TRU- or MA-dominated fuel has an unfavourable influence on several of these parameters, and this is one of the reasons for the recent revival of various fast and thermal reactor concepts which were studied in the past, but have not been introduced as commercial systems. For example, the (positive) coolant void effect in sodium-cooled fast reactors, which deteriorates in a minor actinide burning regime, can be mitigated by substituting the sodium by *lead*, or even eliminated by substituting the liquid metal by a *gas coolant*.

To ensure that a critical burner core performs satisfactorily and has acceptable safety parameters, it is usually necessary to blend the TRU or minor actinides with the fertile materials uranium or thorium. However, blending reduces the transmutation effectiveness of the system. In this context, accelerator-driven systems offer interesting *additional parameters of freedom* by removing the criticality constraint and increasing the safety margin to prompt criticality. The latter feature is particularly important for MA burners, which are difficult to control as critical systems because the effective delayed-neutron fraction is only about half of that of a normal fast reactor.

In response to the new core design issues raised by P&T and the increased interest in advanced reactor technology in general, government and industry funded design teams in Europe, the Far East and the USA are currently spending a considerable effort on the optimisation of a broad range of advanced reactor designs featuring both critical and accelerator-driven cores.

### 3.2 Transmutation effectiveness

Various, sometimes conflicting definitions for transmutation effectiveness, usually based on the minor actinide balance of a particular core, can be found in the literature (see, for instance, [6]). For a system with fully closed fuel cycle, the difficulty of defining a core-specific transmutation effectiveness is circumvented by defining an “overall transmutation effectiveness” as the relative content of the top-up fuel in transuranic and minor actinides,  $M^B/M^T$ , i.e. the already discussed quantity  $\tau$ .

It is interesting to notice that the overall transmutation effectiveness does not depend directly on the choice of the neutron spectrum, the fuel type and the coolant, but is governed by the above-mentioned performance and safety constraints. For a critical TRU burner based on liquid metal technology,  $\tau$  is smaller than about 0.5, and in the case of homogeneous MA recycling in a EFR-type fast reactor  $\tau$  is less than 0.1. The possibility to operate sub-critical MA and TRU burner cores with a fertile-free top-up fuel and hence 100% overall transmutation effectiveness, i.e.  $\tau = 1$ , is probably the *most important advantage of accelerator-driven systems*.

### 3.3 Neutronic transmutation effect

Using the same notation as before, the radiotoxicity reduction relative to the top-up fuel,  $R^T(t)$ , is:

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

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

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

where  $R^N(t)$  is a time-dependent “neutronic transmutation factor”<sup>4</sup>. It should be noticed that the toxicity reduction relative to the actinides to be burnt, which is of direct relevance for the assessment of transmutation schemes, equals  $R^T$  for all practical purposes because the toxicity of the fertile component of the top-up fuel is negligible compared with that of the actinides to be burnt.

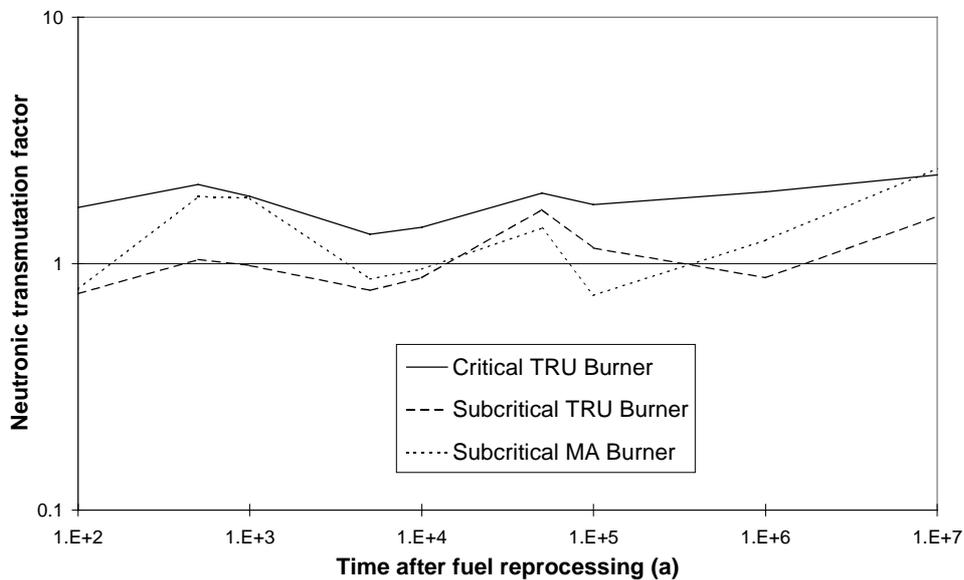
Figure 4 compares the neutronic transmutation factors for three systems: (a) an ALMR-type critical TRU burner, (b) an ATW-type sub-critical TRU burner with a fast-neutron spectrum, and (c) a minor actinide burner operating in the P&T cycle of a double strata fuel cycle. It can be seen that the critical burner has a small “neutronic” advantage over the sub-critical burners, but the difference is not significant compared with the goal for the total toxicity reduction by a factor of 100.

The general conclusions to be drawn from this discussion are that:

- Regarding the neutronic transmutation factor, no single transmutation system has a significant advantage over other systems.
- Radiotoxicity reduction has to be achieved primarily by an actinide mass reduction which implies the maximisation of fuel burn-up and the minimisation of reprocessing and fuel fabrication losses.

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

Figure 4. **Neutronic transmutation factors for transuranics and minor actinide burners**



<sup>4</sup>  $R^N(t)$  is a characteristic of the core and is sometimes called “neutronic toxicity reduction”.

#### 4. Fuel cycle problems and challenges

Current proposals for P&T technology rely on the aqueous (PUREX-type) reprocessing of spent fuel as a preliminary step preceding minor actinide partitioning. But, even in case of pyrometallurgical processing of TRUs, a mechanical head-end and an aqueous processing step (called UREX) for the prior removal of uranium as the main fertile element in any case precedes the sequence of pyrometallurgical separation steps.

The drastic increase of the plutonium content from 15% up to 43% and the fuel burn-up from 80 GWd/t<sub>HM</sub> up to 210 GWd/t<sub>HM</sub> for fast reactor concepts such as CAPRA make aqueous reprocessing more difficult because of the low solubility of plutonium and the radiation damage to the organic extractant (tributyl phosphate). Industrial “pilot” scale work at Dounreay and Marcoule has shown that, in specific technological conditions (pin-chopper, fast contactors), aqueous reprocessing can be considered as valid for fast reactor and future ADS fuel, if the decay heat can be mitigated by cooling or by dilution with LWR fuel. Special chopping or shearing systems have been developed for fast reactor (FR) fuel, e.g. for FFTF, in order to combine the single or multiple pin shearing with a more economical bundle-shear approach.

Spent fuel arising from a reactor park composed of 70% LWR-UOX reactors, 10% LWR-MOX reactors and 20% FR-MOX reactors has to be reprocessed in order to guarantee the plutonium recycle requirements in a stable scenario. The corresponding spent fuel discharges per 100 Gwe-a delivered by the reactor park are summarised in Table 2. Table 3 gives the residual decay heat for these fuel types, assuming the reprocessing operations to be carried out 5 years after the discharge of the fuel from the reactor.

Table 2. Spent fuel arisings from a composite nuclear reactor park per 100 GWe-a

Fuel type	Burn-up (GWd/t <sub>HM</sub> )	Delivered energy (GWe)	Spent fuel discharge (t <sub>HM</sub> )
LWR-UOX	50	69.7	1 520
LWR-MOX	50	10.3	224
FR-MOX	150-200	20.0	124-153

Table 3. Decay heat of spent fuel 5 years after discharge from reactor

Fuel type	Burn-up (GWd/t <sub>HM</sub> )	Total decay heat (kW/t <sub>HM</sub> )	Fission products (kW/t <sub>HM</sub> )	Actinides (kW/t <sub>HM</sub> )
LWR-UOX	50	2.76	2.28	0.477
LWR-MOX	50	6.56	1.97	4.60
FR-MOX	150	30.3	6.60	23.7
FR-MOX	210	33.7	8.14	25.6

##### 4.1 Issues of aqueous reprocessing

The present state-of-the-art in aqueous reprocessing based on the PUREX process is almost satisfactory with regard to uranium and plutonium separation. Recovery yields of nearly 99.7% have been achieved and 99.9% is potentially achievable in a near future. However, reprocessing of industrial

quantities of LWR-MOX and FR-MOX within short cooling times will require gradual adaptations of the PUREX flow-sheet and involve the construction of additional extraction rigs.

In order to improve the plutonium dissolution yield and to avoid solvent radiation peaks during the extraction, a separate dissolver dedicated to FR-MOX treatment could be installed and connected to the main dissolver by a metering system. By connecting the dedicated FR-MOX dissolver to the main LWR dissolver, a constant radiation level can be kept throughout a given processing campaign. The second dissolver could also serve as “residue dissolver” by making use of highly oxidising compounds (e.g. electrochemically generated Ag(II)) to dissolve the insoluble fraction of the initial plutonium inventory and to reduce the transfer of insoluble plutonium residue to the waste stream.

The decay heat of the separated plutonium fraction enters the organic phase and remains in contact with the actinide fraction (U, Pu, Np) until separation and purification of the major actinides occurs. The  $^{238}\text{Pu}$  concentration in the plutonium stream is the main source of radiation damage to the tributyl phosphate extractant, a strong source of neutrons in the separation plant, and an additional radiolysis agent during the oxalate precipitation and conversion to  $\text{PuO}_2$ . On the other hand, the high  $^{238}\text{Pu}$  concentration in the separated  $\text{PuO}_2$  for the same reasons (heat, spontaneous neutrons) improves the non-proliferation resistance of the product during storage. The MOX fuel fabrication will have to adapt its handling technology to reduce the radiation dose to the workers in the plant and during the transport of MOX fuel assemblies.

The separation of the minor actinides is currently under intense investigation throughout the OECD/NEA countries. The separation of the bulk of  $^{237}\text{Np}$  from the aqueous product solution during the reprocessing operations is technically feasible but requires an adaptation of the first cycle extraction flow-sheet. The residual  $^{237}\text{Np}$  which goes directly to the HLW solution could, in principle, be recovered by recycling the HLW solution through a secondary recovery cycle after a valence adjustment. With liquid extraction processes, the separation yield can be increased up to the desired value of 99.9% by increasing the number of extraction stages.

The separation of americium and curium is much more difficult, but considerable progress has been made during the past decade. Bulk separation of minor actinides together with lanthanides has been demonstrated under hot-laboratory conditions with several new processes such as TRUEX, DIDPA, DIAMEX and TRPO [6,9]. The separation of minor actinides from lanthanides, however, remains an obstacle for the industrial up-scaling of Am-Cm separation from HLW. Several promising laboratory methods – e.g. the ALINA and CYANEX301 processes using DTPA [10] and BTP [11] as specific extractants for the separation of minor actinides from lanthanides – have been tested at the ITU of JRC Karlsruhe. Yields of 99% and 97.6% have been achieved for americium and curium, respectively. Further improvements are necessary in order to include these methods in a cycle of multiple reprocessing.

By progressively incorporating an additional extraction rig for minor actinides from HLW coupled to a conventional vitrification process, a much less toxic HLW could be obtained, keeping the actinides in the fuel cycle for ultimate fissioning, and keeping the fission products in the glass.

## **4.2 Pyrochemical reprocessing**

The proliferation risk potentially associated with the clean plutonium produced by the aqueous reprocessing has drawn the attention on the pyrochemical reprocessing which makes it difficult to separate individual TRU elements. Whereas the pioneering work was performed by ANL in the USA [12], most of the recent advances have been made in Russia [13] and Japan [14]. In the Russian

Institute of Atomic Research (RIAR), a pyrochemical process has been demonstrated with highly irradiated spent oxide fuel with burn-ups of 210-240 GWd/t<sub>HM</sub>. Good results were obtained during the demonstration. The recovered PuO<sub>2</sub> will be mixed with UO<sub>2</sub> and processed by vibropacking into fresh FR-MOX fuel. Pyrochemical reprocessing of metallic fuel was developed in the USA in the framework of the integral fast reactor (IFR) programme with capability for actinide burning. Recovery yields of 95% for uranium and 99% for mixtures of uranium and minor actinides have been obtained on laboratory and pilot-plant scales at the Idaho Argonne West Laboratories.

The advantages and disadvantages of the pyrochemical reprocessing can be summarised as follows:

- Highly concentrated TRU product streams can be handled without major radiation degradation of the reagents, allowing the facilities to be more compact than aqueous reprocessing facilities for the same TRU throughput. Because of the absence of water, much smaller criticality risks during purification and metallic fuel re-fabrication arise when processing industrial quantities of TRUs.
- Since all TRU elements remain together throughout the process, the proliferation risk is much reduced. The separation of the TRUs from the lanthanides, however, is difficult, requiring the development of multistage countercurrent extraction using highly corrosive reagents at high temperature. The most challenging issue is the selection and industrial manufacture of corrosion resistant equipment which must be designed for remote operation and maintenance.
- For spent LWR-UOX fuel, a genuine pyrochemical process has to cope with the problem of the elimination of the uranium, which is the major constituent of the LWR fuel. Therefore, a mixed approach using a (P)UREX-type process for uranium-neptunium elimination has been selected as the first step of the “ATW road map” project. Fluoride volatility has also been suggested as an alternative, but the mixed volatility of U-Pu and zirconium leads to complex waste streams which can be difficult to control.
- Starting from metal or nitride TRU fuel, a complete pyrometallurgical process involving a series of electro-refining steps in a wide range of molten salt baths has recently attracted much interest throughout the nuclear research communities in the USA, Russia, Japan and France. Most of the respective flow-sheets remain in the pre-conceptual phase and will require several decades of R&D to become a mature technology comparable to the present aqueous reprocessing.
- Whereas the aqueous reprocessing consists of an independent industrial process which supports a large reactor park and can operate independently on continental or even world scales, the pyrochemical process will predominantly be applied in small facilities installed in the immediate vicinity of the reactors.
- However, in the long-term, pyrochemical reprocessing will become indispensable, if very hot fuel has to be multi-recycled in fast reactor or ADS facilities on an industrial scale with a limited out-of-pile inventory.

## 5. Principal actinide transmutation strategies

Depending on regional boundary conditions and political factors, countries with P&T programmes have developed different transmutation strategies. As to the transmutation of actinides, Table 4 provides an overview of the principal approaches and indicates respective driving forces. In

view of the historic development, the table distinguishes between evolutionary (right) and innovative (left) approaches.

The *evolutionary approach*, adopted mainly in Europe and Japan, aims at closing the fuel cycle in successive steps, starting with the recycling of plutonium in LWRs and later in fast reactors using conventional reprocessing and MOX fuel technology, and finally complementing the system with a dedicated P&T cycle which features MA burners with fast neutron spectra. This approach has the advantage that it can respond flexibly to changes in the priorities for plutonium and minor actinide management, and that new technologies have to be developed only for a comparatively small number of MA burners which support a large system of conventional LWRs and fast reactors.

Table 4. **Principal actinide transmutation strategies**

<b>TRU burning</b>		<b>Pu recycling</b>	
Separation of uranium and TRU from spent LWR fuel, TRU remain together. TRU recycled in thermal or fast critical or sub-critical reactors with fully closed fuel cycles. Dry reprocessing particularly suited for closed fuel cycles and highly active fuels.  Principal driving force: non-proliferation		Separation of uranium and Pu from spent LWR fuel. Pu recycled in thermal and later in fast reactors (limited number of “thermal” recycles). PUREX-type wet reprocessing methods are appropriate. Flexible thermal: fast reactor ratio from about 4 to zero. Possibility to move to a pure fast reactor strategy.  Principal driving force: resource management	
<b>With thermal neutrons</b>	<b>With fast neutrons</b>	<b>Without transmutation</b>	<b>With MA transmutation</b>
TRU burning in thermal reactor.	<b>TRU burning in FR</b>  Flexible thermal: fast reactor ratio from about 2 to zero.  Possibility to move to a pure FR strategy (IFR system).	<b>Pu burning</b>  Fully closed fuel cycle for Pu.  MA and FP conditioning by vitrification and/or dedicated insolubilisation with ceramics technology.	<b>Double strata fuel cycle</b>  MAs (and FPs) burnt in a fully closed P&T cycle.  Fast spectrum required for MA transmutation, ADS has safety advantages.  Dry reprocessing particularly suited for fully closed cycle.  One MA burner supports some 15 “normal” reactors.
<b>TRU burning in thermal ADS</b>  Pure burner strategy. (thermal ATW system)	<b>TRU burning in fast ADS</b>  Thermal: fast reactor ratio of about 3.  Possibility to move to a pure ADS strategy by adding fertile fuel (Energy Amplifier).	Principal driving force: waste management.	Principal driving force: waste management.

The *innovative approach*, first suggested in the USA, aims at co-processing plutonium and minor actinides to avoid the use of technologies with a potentially high proliferation risk. After the initial

separation of the uranium from the LWR spent fuel, the unseparated transuranic actinides are recycled in a transuranic burner with a closed fuel cycle based on pyrochemical reprocessing. Compared with the double strata strategy, the number of burners in the equivalent system is four to six times larger, but the burners are not subjected to a (fast) neutron-spectrum condition. Nevertheless, most of the currently evaluated critical and sub-critical transuranic burners feature a fast neutron spectrum.

### 5.1 Strategies studied by OECD/NEA

An expert group of the OECD/NEA Nuclear Development Committee is currently comparing the principal actinide burning and transmutation strategies in more detail. The investigated strategies comprise:

- a) Plutonium burning in LWRs and CAPRA-type fast reactors.
- b) The double strata strategy with LWRs and CAPRA reactors in the first stratum and accelerator-driven MA burners in the second stratum.
- c) TRU burning in critical fast reactors (IFR concept).
- d) TRU burning in sub-critical fast reactors (ADS).
- e) A heterogeneous strategy in which americium and curium are recycled in targets.

Table 5 summarises the most important assumptions for strategies a to d, and Figure 5 gives preliminary results for the achievable actinide waste radiotoxicity reduction relative to the open fuel cycle. The figure shows that transmutation strategies b, c and d all meet the goal of a hundred-fold reduction, and confirms that plutonium recycling alone is not effective in reducing the actinide waste radiotoxicity.

Figure 5. Actinide waste radiotoxicity reduction relative to open fuel cycle (Preliminary results of OECD/NEA study)

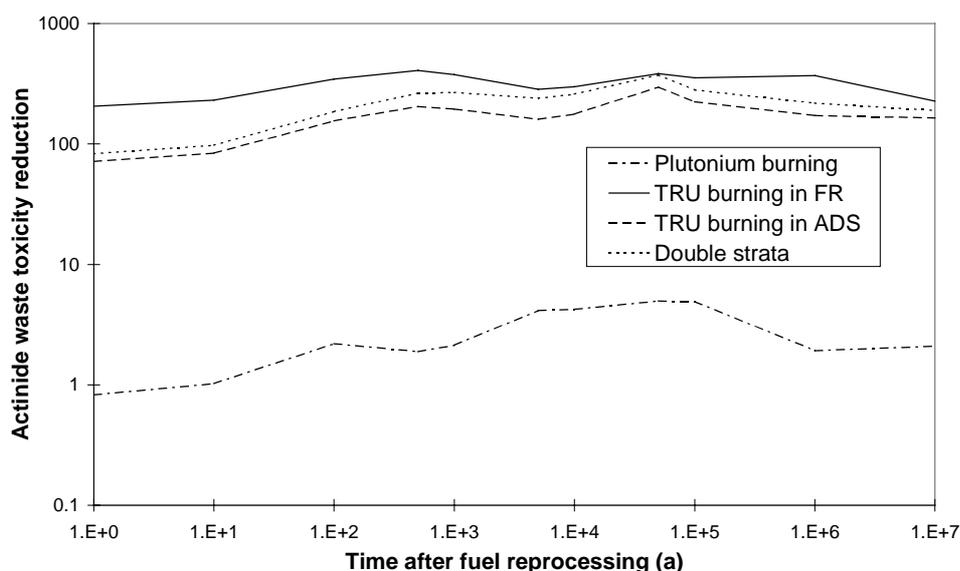


Table 5. Assumptions for transmutation strategies compared by OECD/NEA

Strategies	Reactor/ADS	Fuel	Av. burn-up (GWd/t <sub>HM</sub> )	Reprocessing method	Recovery yield (%)
All	LWR (N4)	UOX/MOX	50	wet	99.9
a, b	FR (CAPRA)	MOX	185	wet	99.9
b	MA Burner, ADS	AcN-ZrN	140	dry	99.9
c	TRU Burner, IFR	Ac-Zr	140	dry	99.9
d	TRU Burner, ADS	Ac-Zr	140	dry	99.9

## 6. Fission product transmutation

The neutron capture process is currently the only promising nuclear reaction for transmuting fission products. Other processes which have been proposed in the past rely on technologies which are still at a very early stage of development (e.g. fusion neutron sources) and generally suffer from a poor energy balance. The capture process consumes neutrons but, theoretically, fast reactors could deliver *enough surplus neutrons* to allow the potentially troublesome long-lived fission products to be completely transmuted to shorter-lived or stable species (cf. Table 1).

The transmutation of a fission product makes sense only if the reaction rate (microscopic cross-section times neutron flux) is higher than the natural decay rate of the nuclide. With the practically achievable neutron fluxes, this condition cannot be met for the most abundant fission products <sup>137</sup>Cs and <sup>90</sup>Sr with half-lives of only about 30 years, i.e. these fission products are “non-transmutable”. However, since their radioactive life is limited to less than 300 years, they can be safely enclosed using engineered barriers only. On the other hand, the fission products determine the size of the vitrified waste repository which, consequently, *cannot be much reduced* by P&T operations.

The fission products which influence the long-term risk of a repository are, in order of radiologic importance, <sup>129</sup>I, <sup>99</sup>Tc, <sup>135</sup>Cs, <sup>93</sup>Zr, and <sup>126</sup>Sn. Activation products (<sup>14</sup>C and <sup>36</sup>Cl) can also contribute to the dose. Obviously, the relative contribution of these nuclides to the integral risk varies according to the type of repository host rock. In the following, the problems associated with the transmutation of these five fission products are discussed separately. It should be noted that the determination of the separation yield and the decontamination factor (DF) for HLW depends to a great extent on policy decisions and that, depending on the nuclide, special *conditioning and confinement* is an alternative to transmutation.

### 6.1 Iodine-129 ( $T_{1/2} = 1.6 \cdot 10^7$ a)

In most of the direct storage concepts for spent fuel, <sup>129</sup>I is the first nuclide to enter into the biosphere due to its very high mobility. During aqueous reprocessing, iodine is removed from the dissolver solution with a yield of 95-98%. To improve the separation yield, more complex chemical treatments are necessary. Better separation yields may also be achieved with high-temperature pyrochemical processes. Special methods for conditioning in the form of AgI, PbIO<sub>4</sub>, etc. have been developed but, because of the extremely long half-life, the eventual migration to the environment cannot be excluded.

Since the radiotoxicity of  $^{129}\text{I}$  is the highest of the fission products and equivalent to that of actinides, it would be advisable to increase the separation yield to achieve a DF of about 1 000. The necessity of an isotopic separation and the limited stability of the target material, however, make the transmutation of  $^{129}\text{I}$  difficult, and conditioning and confinement seems to be the best method to reduce its radiological impact. Nevertheless, the present method of diluting iodine in the sea may still be defensible because of the enormous dilution of  $^{129}\text{I}$  in the (natural)  $^{127}\text{I}$  present in seawater. The storage in a salt dome, consisting of evaporated seawater, is another alternative which still has its merits.

### **6.2 Technetium-99 ( $T_{1/2} = 2.1 \cdot 10^5 \text{ a}$ )**

The radiologic significance of  $^{99}\text{Tc}$  is important if the repository surroundings are slightly oxidic. In reducing conditions  $^{99}\text{Tc}$  is remarkably stable and insoluble as technetium metal or  $\text{TcO}_2$  suboxide. Partitioning of  $^{99}\text{Tc}$  is not an easy task because it occurs as insoluble metal and as soluble technetate ion in solution. Separation from aqueous effluents is possible in an advanced PUREX scheme, but recovery from insoluble residues is difficult, with the present recovery yield at best reaching 80%. Improving this yield significantly implies the development of new separation technologies such as the not yet proven conversion of the technetium into a single chemical species. Alternatively, a group separation together with the platinum metals may be carried out using pyrometallurgical processes. If separated in metallic form, transmutation appears to be feasible because of its stability and relatively large neutron capture cross-section.

### **6.3 Caesium-135 ( $T_{1/2} = 2.3 \cdot 10^6 \text{ a}$ ), Zirconium-93 ( $T_{1/2} = 1.5 \cdot 10^6 \text{ a}$ ) and Tin-126 ( $T_{1/2} = 1.0 \cdot 10^5 \text{ a}$ )**

Caesium occurs in the form of the isotopes 133 (stable), 135 and 137. In terms of radiologic significance,  $^{137}\text{Cs}$  is the major constituent of HLW (see above). The activity of the long-lived  $^{135}\text{Cs}$  in HLW is a million times lower. However, once released from a matrix as glass, caesium is very mobile. Transformation of  $^{135}\text{Cs}$  to stable  $^{136}\text{Ba}$  is possible from a neutronics point of view, but probably impracticable because a close to 100% isotopic separation efficiency would be required (traces of  $^{133}\text{Cs}$  in the target would generate new  $^{135}\text{Cs}$  during the irradiation).

Zirconium-93 is somewhat similar to  $^{135}\text{Cs}$ , it has also a very long half-life and a small isotopic abundance (about 14% of the total Zr). An isotopic separation would be necessary, and its transformation to stable  $^{94}\text{Zr}$  would be very slow because the thermal capture cross-section is about five times smaller than that of  $^{135}\text{Cs}$ .

Tin-126 is partly soluble in HLW from aqueous reprocessing but occurs also as an insoluble residue, similar to technetium. Isolation involves a special treatment of the HLW, and the use of isotopic separation techniques. Transmutation of  $^{126}\text{Sn}$  is questionable due to the very low neutron capture cross-section.

## **7. Consequences for geologic disposal**

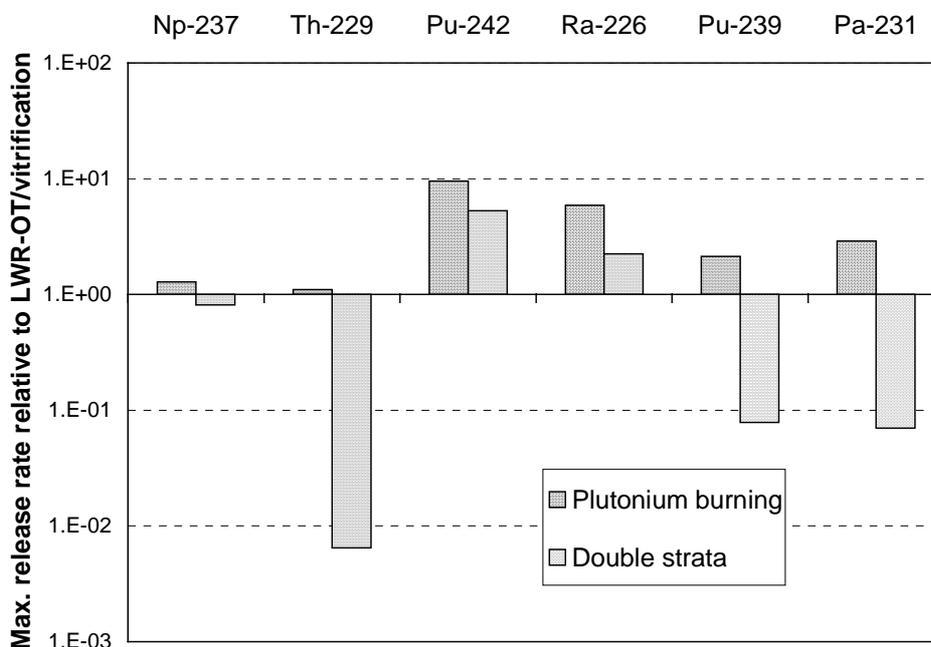
As mentioned before, the primary concern of geologic repositories are possible releases of the relatively mobile fission products. Since the fission product yields are not very sensitive to the fuel composition and the neutron spectrum of the reactor, the fission product risk depends primarily on the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

separating troublesome fission products from the waste. The choice of the reactor can, however, influence the in-situ transmutation of a fission product. For example, capture of thermal neutrons in the precursor  $^{135}\text{Xe}$  reduces the  $^{135}\text{Cs}$  production in an LWR by as much as 70% [15].

As to the influence of different transmutation strategies on the release of actinides from a repository, the usual approach is to perform complete nuclide transport calculations. Such an integral approach was chosen, for instance, by the authors of a recent study of the European Commission who compared nuclide fluxes at the clay-aquifer interface [16]. However, a difficulty of this approach is the strong dependence of the results on the host rock characteristics, which can vary considerably depending on the structure of the host rock and the storage concept (storage in salt, clay or granite). For generic studies, it may therefore be preferable to perform site-independent comparisons on the basis of releases of potentially troublesome actinides from the well-defined near-field of the repository<sup>5</sup>, allowing a subsequent folding with site-specific geosphere and biosphere responses to be performed independently and according to the needs of specific repository projects [15].

Adopting the latter approach, near-field release rates have been evaluated for the strategies with vitrification investigated in the framework of the afore-mentioned OECD/NEA study. The results in Figure 6 indicate a strongly non-linear relationship between release rates and actinide concentrations in the glass. Important conclusions are that the plutonium burning strategy generally increases the maximum release rates, and the addition of the P&T cycle does not reduce maximum release rates for all potentially troublesome actinides.

Figure 6. **Maximum release rates from near-field relative to LWR once-through/vitrification case (Strategies investigated in the framework of the OECD/NEA study)**



<sup>5</sup> Actinides to be considered in this context are  $^{231}\text{Pa}$ ,  $^{237}\text{Np}$  and their respective daughter products  $^{227}\text{Ac}$  and  $^{229}\text{Th}$ ,  $^{226}\text{Ra}$ , a decay product of  $^{234}\text{U}$ , and the long-lived plutonium isotopes 239 and 242.

## 8. Summary

The principal points and conclusions arising from this overview discussion of the nuclear fuel cycle and P&T are summarised as follows:

The closure of the nuclear fuel cycle with P&T will be a long-term endeavour and becomes a central issue in the development of a future sustainable nuclear energy system; the P&T strategy will directly influence the choices of new reactor and reprocessing technologies.

Plutonium recycling is a first step in this direction. Plutonium can be managed effectively with existing LWRs which, later on, should be complemented with fast reactors to burn the plutonium completely. The necessary extension of the proven, PUREX-type reprocessing technology to cope with the highly active fuels arising from plutonium-burning strategies appears to be feasible. The motivation for the utilisation of plutonium is its energy content, but not a mitigation of the long-term radiological hazard associated with the back-end of the fuel cycle.

Partitioning and transmutation aims at making the fuel cycle more sustainable from the viewpoint of the back-end by reducing the HLW radiotoxicity and (possibly) the migration of radiotoxic nuclides from HLW repositories to the biosphere. To this end, it introduces the separation and transmutation (or, alternatively, improved immobilisation) of minor actinides and fission products, assuming that the fuel cycle is already (or simultaneously) closed for plutonium.

Transmutation implies the development of advanced and innovative reactor and fuel cycle technologies, including ADS reactor technology, fuels with very high burn-up capability, and pyrochemical reprocessing methods. It can be shown that the goals of transmutation cannot be achieved without the implementation of fast-neutron-spectrum systems in some form, and that the overriding requirement is that for high fuel burn-ups and actinide recovery yields. Regarding neutronics, it appears that no single transmutation system has a significant advantage over other systems.

As to the separation of minor actinides from HLW using aqueous reprocessing, promising new processes have recently been developed. The results achieved at laboratory and pilot-plant scales give confidence that the required high recovery yields can ultimately be achieved on an industrial scale. Nevertheless, the long-term future appears to belong to the pyrochemical reprocessing method, which is not yet mature, but is intrinsically better suited for a fully closed fuel cycle with highly active fuels and may be more proliferation resistant because all actinides remain together, and the relatively compact plants can be collocated with the reactors.

A comparison of the double strata and the TRU burning strategy indicates that, under comparable assumptions, the two strategies are equivalent and that both have the potential of achieving a hundred-fold radiotoxicity reduction of the actinides in the HLW. These (or any other) actinide transmutation strategies, however, are not effective in reducing the total mass of the HLW which is dominated by the fission products and, hence, mainly depends on the total energy produced.

The primary risk of geologic repositories is related to the release of long-lived fission products. With the exception of  $^{99}\text{Tc}$ , however, the transmutation of long-lived fission products appears to be difficult because of low neutron reaction cross-sections and the necessity of isotopic separations. This means that, for most fission products, special conditioning and confinement is the only practical method to reduce the radiological impact. Finally, it is shown that, due to the non-linear relationship between release rates from a repository and actinide concentrations in the glass, P&T does not necessarily result in reduced release rates for all potentially troublesome actinides as one could expect.

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