



Contents lists available at ScienceDirect

# Progress in Particle and Nuclear Physics

journal homepage: [www.elsevier.com/locate/ppnp](http://www.elsevier.com/locate/ppnp)

## Review

# Radioactive waste partitioning and transmutation within advanced fuel cycles: Achievements and challenges

M. Salvatores<sup>a,b,\*</sup>, G. Palmiotti<sup>b</sup><sup>a</sup>CEA-Cadarache, DEN-Dir, 13115 Saint-Paul-Lez-Durance, France<sup>b</sup>Idaho National Laboratory, 2525 Fremont Ave., P.O. Box 1625, Idaho Falls, ID 83415, USA

## ARTICLE INFO

### Keywords:

Nuclear reactor  
Partitioning  
Transmutation  
Advanced fuel cycle  
High-level waste

## ABSTRACT

If nuclear power becomes a sustainable source of energy, a safe, robust, and acceptable solution must be pursued for existing and projected inventories of high-activity, long-lived radioactive waste. Remarkable progress in the field of geological disposal has been made in the last two decades. Some countries have reached important milestones, and geological disposal (of spent fuel) is expected to start in 2020 in Finland and in 2022 in Sweden. In fact, the licensing of the geological repositories in both countries is now entering into its final phase. In France, disposal of intermediate-level waste (ILW) and vitrified high-level waste (HLW) is expected to start around 2025, according to the roadmap defined by an Act of Parliament in 2006. In this context, transmutation of part of the waste through use of advanced fuel cycles, probably feasible in the coming decades, can reduce the burden on the geological repository. This article presents the physical principle of transmutation and reviews several strategies of partitioning and transmutation (P&T). Many recent studies have demonstrated that the impact of P&T on geological disposal concepts is not overwhelmingly high. However, by reducing waste heat production, a more efficient utilization of repository space is likely. Moreover, even if radionuclide release from the waste to the environment and related calculated doses to the population are only partially reduced by P&T, it is important to point out that a clear reduction of the actinide inventory in the HLW definitely reduces risks arising from less probable evolutions of a repository (i.e., an increase of actinide mobility in certain geochemical situations and radiological impact by human intrusion).

© 2010 Elsevier B.V. All rights reserved.

## Contents

1. Introduction.....	145
1.1. Spent nuclear fuel and radioactive wastes.....	145
1.2. Features and potential role of P&T.....	147
1.3. P&T objectives.....	148
2. Fundamentals of transmutation.....	148
2.1. Physics of transmutation.....	148
2.1.1. The competition between capture and fission reactions.....	148
2.1.2. The neutron consumption per fission.....	149
2.1.3. Neutron balance ( <i>D</i> -factor) intercomparison.....	150
2.2. Implementation in different types of reactor.....	150
2.2.1. Transmutation performances of thermal reactors.....	150

\* Corresponding author at: CEA-Cadarache, DEN-Dir, 13115 Saint-Paul-Lez-Durance, France. Tel.: +33 442253365; fax: +33 442254142.  
E-mail address: [Massimo.salvatores@cea.fr](mailto:Massimo.salvatores@cea.fr) (M. Salvatores).

2.2.2.	Transmutation in FRs. Implementation and performance .....	152
2.2.3.	Transmutation in dedicated transmuters: external source-driven subcritical reactors and critical low conversion ratio FRs .....	155
2.2.4.	Transmutation of long-lived FPs (LLFPs) .....	156
2.2.5.	An essential challenge: transmutation consequences on the fuel cycle .....	158
2.3.	Summary of technological options .....	158
3.	Status of research and development .....	159
3.1.	Transmutation physics .....	159
3.2.	Technology .....	160
3.3.	Separation R&D [32] .....	160
3.4.	Fuels for transmutation .....	161
4.	Potential benefits and impact on deep geological repositories of advanced fuel cycles with P&T .....	162
4.1.	Status of some deep geological repository programs .....	162
4.2.	P&T potential benefits .....	162
5.	P&T implementation in an EU perspective .....	164
6.	Conclusions .....	165
	References .....	165

## 1. Introduction

By 2050, the global electricity demand is expected to have increased by about a factor 2.5 [1]. At the same time, the world faces environmental threats caused by anthropogenic polluting emissions. Nuclear energy offers the opportunity to meet a significant part of the anticipated increase in electricity demand in combination with a reduction of the polluting emissions. Therefore, a strongly increasing demand for nuclear power can be expected.

- Nuclear installed capacity could be multiplied by a factor of 3 to 4 by 2050 (1500–2000 gigawatt electrical, GWe). This will be made possible with the next generation of light water reactors (LWRs) using uranium fuel, which are expected to dominate the world market for the first half of the 21st century.
- These reactors will have a minimum lifetime of 60 years. The countries that will build these reactors by 2050 and plan to operate them until 2110 will have to take into consideration uranium supply issues.
- There is a definite need for a clear and proven vision of waste management.

In such a scenario, sustainability becomes a predominant concern, which means that preservation of natural resources, waste minimization, and proliferation resistance criteria are as important as economy and safety.

### 1.1. Spent nuclear fuel and radioactive wastes

Most of the hazards in dealing with spent fuel stem from some of the following chemical elements: plutonium, neptunium, americium, curium, and some long-lived fission products (FPs) such as iodine and technetium at concentration levels of kilograms per ton. At present, approximately 2500 tons of spent fuel containing about 25 tons of plutonium and 3.5 tons of the “minor actinides” (MAs) neptunium, americium, and curium, as well as 3 tons of long-lived FPs (out of a total of about 100 tons of FPs) are produced annually in the European Union. The contents of typical spent fuel from a pressurized water reactor (PWR) are shown in Fig. 1.

These radioactive by-products, although present at relatively low concentrations in the spent fuel, are hazardous to life forms when released into the environment. As such, their final disposal requires isolation from the biosphere in stable, deep geological formations for long periods of time. Table 1 shows the radioactive characteristics of most spent fuel constituents.

A measure of the hazards of these elements is provided by the *toxicity*, and in particular the *radiotoxicity* arising from their radioactive nature rather than their chemical form. A reference point is the radiotoxicity associated with the raw material used to fabricate 1 ton of enriched uranium, including not only the uranium isotopes, but also all of their radioactive progenies. The radiotoxicity of the FPs dominates the total radiotoxicity during the first 100 years. Long-term radiotoxicity is dominated solely by actinides, mainly plutonium and americium isotopes (see Fig. 2).

The reference radiotoxicity level is reached by spent nuclear fuel only after periods of more than 100,000 years. In more detail, the radiotoxicity of FPs dominates the first 100 years after discharge and decreases to the natural reference level in about 300 years. However, in the longer term, the radiotoxicity is mainly dominated by transuranics (TRUs), particularly plutonium isotopes and decay products of Pu-241. Approximately 100–1000 years after fuel discharge, the radiotoxicity is dominated by Am-241, the radioactive daughter of Pu-241, with a level of about  $3 \times 10^7$  Sv/ton U, i.e., about 300 times as large as the natural reference. Between 1000 and 10,000 years, radiotoxicity is dominated by Pu-240, with a value of about  $4 \times 10^6$  Sv/ton U. Thereafter, Pu-239 is the main contributor to radiotoxicity with a value of  $2 \times 10^6$  Sv/ton U. Beyond 100,000 years, the total radiotoxicity decays to the level of  $10^5$  Sv/ton U. After that, the main sources of radiotoxicity come from the descendants of Am-241.

A detailed analysis of FP radiotoxicity reveals that a few long-lived radionuclides (e.g., I-129, Tc-99, etc.; see Table 1) contribute to very-long-term radiotoxicity. However, their absolute magnitude remains below the TRU radiotoxicity, and

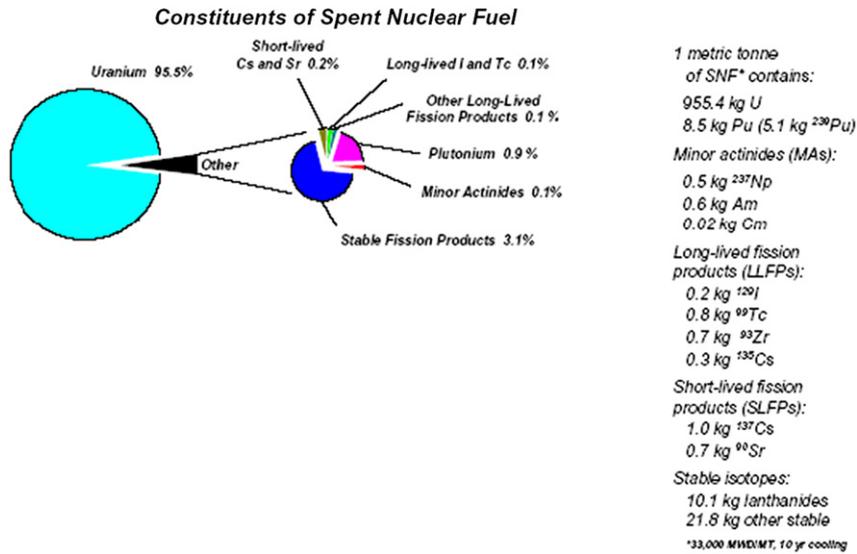


Fig. 1. Spent nuclear fuel composition (PWR-UOX, BU = 33 Gigawatt days per ton, GwD/ton, 10 y cooling).

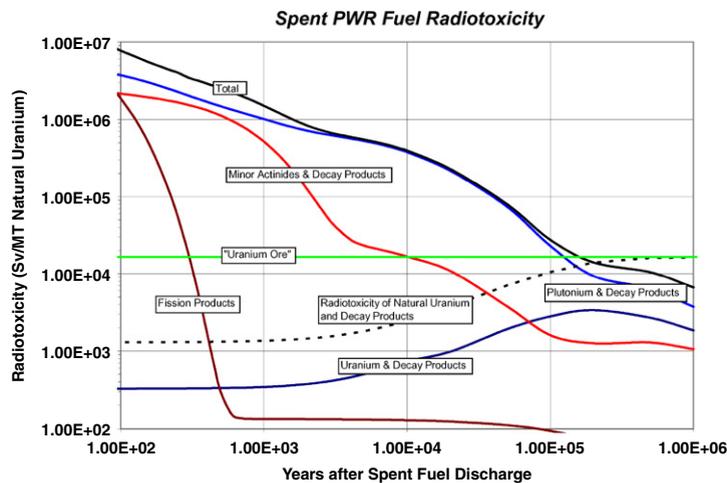


Fig. 2. Spent PWR fuel radiotoxicity and its components.

even below the radiotoxicity of natural ore materials used to fabricate enriched uranium. The total radiotoxicity of FPs is about  $1.4 \times 10^7$  Sv/ton U (enriched) 100 years after discharge, but decreases to 875 Sv/ton U (enriched) after 1000 years. Thereafter, it is stabilized at that level for a long time ( $\sim 100,000$  years) at a level much lower than our reference level for natural ore.

Many fuel cycle options have been discussed and presented in the literature, mainly using an open or closed fuel cycle as well as the so-called P&T strategy that will be described in the next section.

No sustainability is guaranteed with the open-cycle approach due to uranium availability and cost issues. Historically, this option has been associated with LWRs, which effectively use only  $\sim 1\%$  of the mined uranium.

The closed fuel cycle approach has historically been associated with enhanced resource utilization, fuel reprocessing, and Pu recovery, while P&T has been associated with the waste minimization goal and has been discussed mostly in the last two decades as another option.

Recently, the Generation-IV (Gen-IV) initiative [2] defined a set of general goals for future systems in four broad areas: (1) sustainability (more efficient use of the available U resources and waste minimization), (2) enhanced economics, (3) safety and reliability, and (4) proliferation resistance and physical protection. Gen-IV objectives include P&T (waste minimization activities). P&T is seen as consistent with sustainability and non-proliferation objectives (i.e., as an integrated part of “advanced fuel cycles”).

**Table 1**  
Properties of transuranium nuclides.

Nuclide	Half-life	Energies of primary emissions (MeV)		Specific activity			Dose coefficients (Sv/Bq)
		$\alpha$	$\beta$	(Ci/g)	(W/g)	(Neutron $\text{min}^{-1} \text{mg}^{-1}$ )	
<sup>237</sup> Np	$2.14 \times 10^6$ y	4.78		$7.07 \times 10^{-4}$	$2.07 \times 10^{-5}$	$<7 \times 10^{-6}$	$1.1 \times 10^{-7}$
<sup>238</sup> Np	2.10 d		0.25 1.24	$2.61 \times 10^5$	$1.27 \times 10^3$		
<sup>239</sup> Np	2.359 d		0.332 0.427	$2.32 \times 10^5$	$5.86 \times 10^2$		$8.0 \times 10^{10}$
<sup>238</sup> Pu	87.404 y	5.49		17.2	0.570	155	$2.3 \times 10^{-7}$
<sup>239</sup> Pu	$2.4413 \times 10^4$ y	5.15		$6.13 \times 10^{-2}$	$1.913 \times 10^{-3}$	$1.35 \times 10^{-3}$	$2.5 \times 10^{-7}$
<sup>240</sup> Pu	6580 y	5.16		0.227	$7.097 \times 10^{-3}$	53.7	$2.5 \times 10^{-7}$
<sup>241</sup> Pu	14.98 y	4.9	0.02	99.1	$4.06 \times 10^{-3}$		$4.7 \times 10^{-7}$
<sup>242</sup> Pu	$3.869 \times 10^5$ y	4.90		$3.82 \times 10^{-3}$	$1.13 \times 10^{-4}$	95.3	$2.4 \times 10^{-7}$
<sup>241</sup> Am	432.7 y	5.48		3.43	0.1145	$3.55 \times 10^{-2}$	$2.0 \times 10^{-7}$
<sup>242</sup> Am	16.01 h		0.63–0.67	$8.11 \times 10^5$	$2.08 \times 10^3$		
<sup>242m</sup> Am	144 y	5.207	I.T.	10.3	$3.08 \times 10^{-2}$		$1.9 \times 10^{-7}$
<sup>243</sup> Am	7370 y	5.27		0.200	$6.42 \times 10^{-3}$		$2.0 \times 10^{-7}$
<sup>242</sup> Cm	162.7 d	6.11		$3.32 \times 10^3$	122	$1.21 \times 10^6$	$1.3 \times 10^{-8}$
<sup>243</sup> Cm	32 y	5.79		45.9	1.677		$2.0 \times 10^{-7}$
<sup>244</sup> Cm	18.099 y	5.81		80.94	2.832	$6.87 \times 10^5$	$1.6 \times 10^{-7}$
<sup>245</sup> Cm	8265 y	5.36		0.177	$5.89 \times 10^{-3}$		$3.0 \times 10^{-7}$
<sup>246</sup> Cm	4655 y	5.39		0.312	$1.01 \times 10^{-2}$	$5.58 \times 10^5$	$2.9 \times 10^{-7}$
<sup>247</sup> Cm	$1.56 \times 10^7$ y	4.87		$9.28 \times 10^{-5}$	$2.94 \times 10^{-6}$		$2.7 \times 10^{-7}$
<sup>248</sup> Cm	$3.397 \times 10^5$ y	5.05		$4.24 \times 10^{-3}$	$5.34 \times 10^{-4}$	$2.58 \times 10^6$	$1.1 \times 10^{-6}$
<sup>249</sup> Cm	64 m		0.9	$1.18 \times 10^7$	$2.06 \times 10^4$		
<sup>250</sup> Cm	$1.74 \times 10^4$ y			$8.20 \times 10^{-2}$	$\sim 0.1$	$6.49 \times 10^8$	$2.9 \times 10^{-7}$

## 1.2. Features and potential role of P&T

During the last decade, numerous studies were performed in order to identify appropriate P&T strategies aimed at reducing the burden on geological storage (see, among many others, [3]). P&T strategies are very powerful and unique tools that can drastically reduce the radiotoxicity level of the wastes as well as the time needed to reach the reference level (from  $\sim 100,000$  years to a few hundred years (i.e., comparable to the period in which technological and engineering means allow for reasonable control of the radioactivity confinement)). Moreover, P&T allows, in principle, the reduction of the residual heat in a geological repository, with the potential for significant impact on repository size and characteristics.

The first requirement of P&T strategies is the deployment of aqueous or dry spent fuel reprocessing techniques, which are both in the continuity of today's technologies (e.g., as implemented at La Hague in France, where Pu is separated up to 99.9%) or which represent innovative, adapted approaches (e.g., pyrochemistry). The requirement is to extend the performance of Pu separation to 99.9%, separating Np, Am, and Cm, either as a group or individually, and in any case removal of lanthanide contaminants to the extent possible.

Separated TRUs should then be “transmuted” (or “burned”) in a neutron field. The essential mechanism is to transform them via fission into much shorter-lived or stable FPs. However, the fission process is always in competition with other processes and, in particular, with neutron capture, which eliminates isotope atomic mass A, but transforms it into isotope  $A + 1$ , which can still be radioactive. Isotope  $A + 1$  can in turn be fissioned or transmuted into isotope  $A + 2$ , and so on.

The neutron field has to be provided by a fission reactor. The requirement for this (dedicated) reactor is for the fission process to be preferential with respect to the capture process and be loaded with fuels with potentially very different mixtures of Pu and MAs, according to the chosen approach and the objective of the P&T strategy, all while preventing any negative affect on its safety or penalizing its operability.

Partitioning and transmutation is considered, in principle, a means for reducing the burden of a geological disposal. Spent fuel from nuclear power plants must be managed in a safe, environmentally sound manner that is acceptable for the public. At present, two management options are considered worldwide: (1) direct disposal of the spent fuel (associated with the open fuel cycle) and (2) reprocessing. The two options are illustrated in Fig. 3.

The management of the spent fuel is a major challenge for all countries where nuclear energy has been developed, and whatever perspective is applied to its future use, from further development to progressive phase out.

As indicated above, most of the hazards from spent fuel stems from only a few chemical elements—plutonium, neptunium, americium, curium, and some long-lived FPs such as iodine and technetium at concentration levels of kilograms per ton.

These radioactive by-products, although present at relatively low concentrations in spent fuel, are hazardous to life forms when released to the environment. As such, their disposal requires isolation from the biosphere in stable deep geological formations for long periods of time.

P&T has been considered as a way of reducing the burden on a geological disposal. Since plutonium and MAs are mainly responsible for the long-term radiotoxicity, when these nuclides are removed from the waste (partitioning) and fissioned

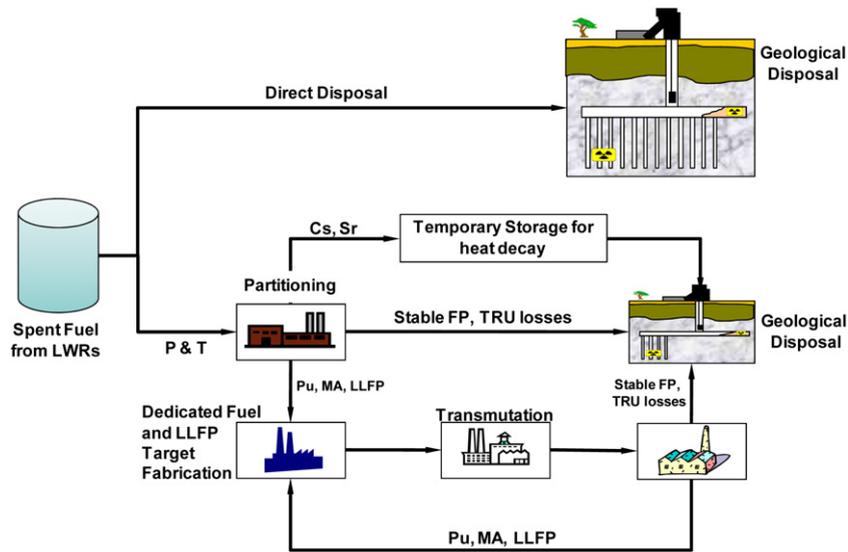


Fig. 3. Open fuel cycle and advanced fuel cycles with P&T.

(transmutation), the remaining waste loses most of its long-term radiotoxicity. However, P&T has little to no impact on the inventory of FPs in the radioactive waste.

Many studies have shown that the radiotoxicity inventory can be reduced up to a factor of 10 if all of the Pu is recycled and fissioned. Reduction factors higher than 100 can be obtained if, in addition, the MAs are burned. A prerequisite for these reduction figures is a nearly complete fissioning of the actinides, for which multi-recycling is a requirement. Losses during reprocessing and refabrication must be well below 1% and probably in the region of 0.1%.

Moreover, the P&T strategy allows, in principle, a combined reduction of the amount of radioactive waste to be stored along with the associated residual heat. These features and their potential impact will be discussed in later sections.

### 1.3. P&T objectives

The P&T approach has been developed within radioactive waste management strategy studies in terms of the reduction of potential source of radiotoxicity as a potential mitigation to the consequences of accident scenarios (e.g., human intrusion) in the repository evolution with time, and of reduction of heat load in the repository, as indicated above. However, despite this common generic interest for P&T, different objectives are pursued that can be gathered into three categories [4].

*Sustainable development of nuclear energy and waste minimization.* For this objective, multi-recycle in fast reactors (FRs) is needed as the TRU is unloaded from LWRs and, successively, as it is unloaded from FRs, if a transition from an LWR fleet to an FR fleet is foreseen. This objective could also be compatible with an increased proliferation resistance of the fuel cycle.

*Reduction of MA inventory.* This objective is compatible both with use of Pu as a resource in LWRs for a limited period of time, in the hypothesis of a delayed deployment of FRs, and with a sustainable development of nuclear energy, based on the deployment of FRs.

*Reduction of TRU inventory as unloaded from LWRs.* This objective is related to the management of spent fuel inventories, as a legacy of previous operation of nuclear power plants.

## 2. Fundamentals of transmutation

### 2.1. Physics of transmutation

The “transmutation” concept in a neutron field applies to the physical phenomena that transform a fresh fuel into an irradiated fuel. The description of such phenomena is obtained by the solution of the set of Bateman equations (see Fig. 4) from which the vector of the nuclei densities  $\bar{n}$  at a time  $t = t_F$ , starting from an initial value  $\bar{n}_{t=t_0}$ , are obtained.

#### 2.1.1. The competition between capture and fission reactions

Any type of transmutation is a function of the neutron cross sections and their spectral dependence. In the transmutation of nuclear wastes, the preferential physics process is obviously fission. The competition between the capture and fission processes is then highly relevant.

$$\frac{dn_j}{dt} = n_j(-\sigma_{aj}\phi - \lambda_j) + \sum_K (\sigma_K^j\phi + \lambda_K^j)n_K$$

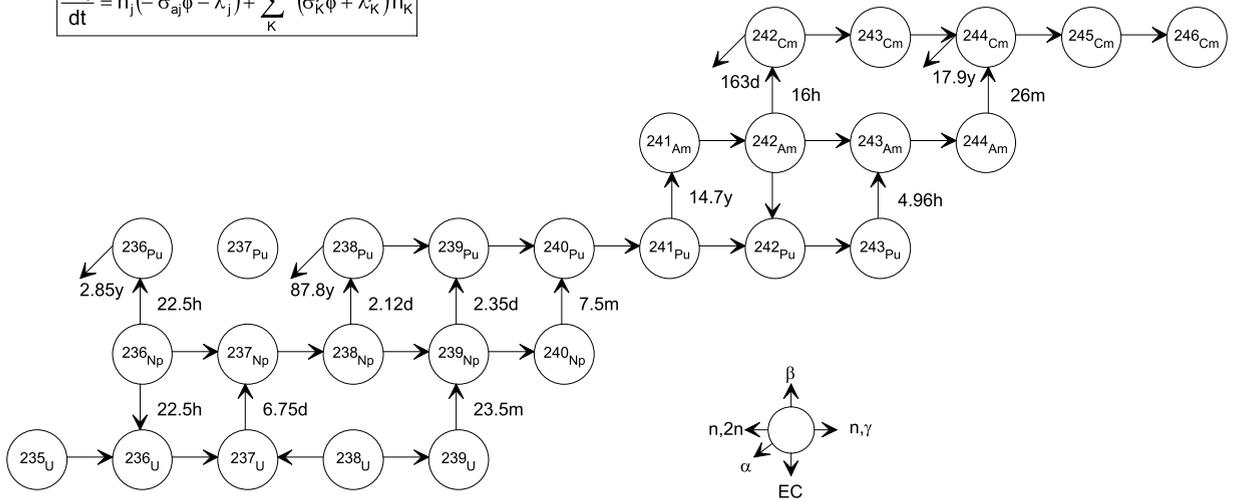


Fig. 4. Actinide transmutation chain and nuclei time-evolution equations.

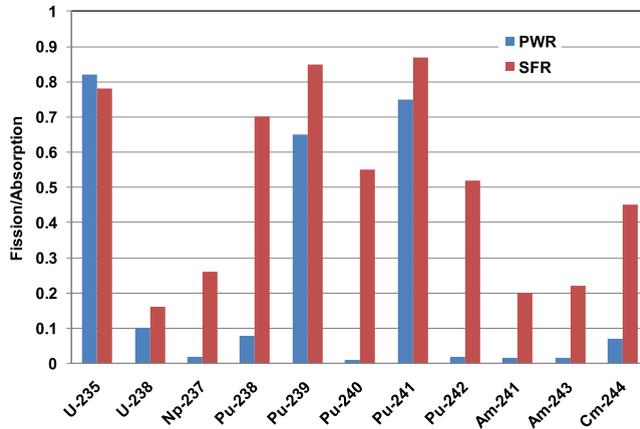


Fig. 5. Comparison of fission/absorption ratio for PWR and SFR [5].

It is useful to look closely at the ratios  $\alpha = \bar{\sigma}_c / \bar{\sigma}_f$  of the average capture and fission cross section of the different isotopes. The neutron fission/absorption cross-section ratio for dominant actinides in PWR and sodium fast reactor (SFR) spectra is illustrated in Fig. 5. The fission/absorption ratios are consistently higher for the fast-spectrum SFR. For fissile isotopes ( $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$ ), over 80% of fast neutron absorptions result in fission, as compared to 60–80% in the PWR spectrum. In addition, the fast-spectrum fission fraction can rise to 50% for fertile isotopes as observed for  $^{240}\text{Pu}$  in Fig. 5, while remaining low (<5%) in a thermal spectrum. Thus, in a fast spectrum, actinides are preferentially fissioned, not transmuted into higher actinides.

This implies that fast systems are more “efficient” (from the point of view of neutron economy) in destroying actinides because fewer neutrons are lost to capture reactions before eventual fission. Furthermore, higher actinides (americium, curium, etc.) continue to build up with LWR recycle. These higher actinides tend to be more radioactive and can be problematic for fuel handling and fabrication in a closed fuel cycle, as will be discussed later.

The hardest spectra are the most suitable, if, as we have indicated, fission is preferred. In fact, the fission cross sections of most of the isotopes of Am and Cm are of the threshold type.

### 2.1.2. The neutron consumption per fission

For a full understanding of the transmutation potential of different neutron fields, the notion of neutron consumption/fission of an isotope  $J$ ,  $D_J$ , has been introduced [6]. In fact, the total number of neutrons  $D_J$  consumed by the given  $J$ -family can serve as an indicator of the capability of a core to achieve destruction of a given  $J$ -feed if neither “parasitic” neutron consumption nor neutron leakage exists. In the case of a negative  $D$  (i.e., when the  $J$ -family produces more neutrons than it consumes), the core fed by  $J$ -nuclides produces enough neutrons to destroy the source material at

**Table 2**  
*D* (neutron consumption/fission) value by isotope in different systems.

Isotope	MOX-LWR <sup>b</sup> $r^a = 2$	VHTR	He-cooled carbide fuel GFR <sup>c</sup>	SUPER-PHENIX <sup>c</sup>	Lead-cooled nitride fuel LFR <sup>c</sup>	Na-cooled oxide fuel SFR <sup>c</sup>	Na-cooled metal fuel SFR <sup>c</sup>
<sup>235</sup> U	−0.43	−0.53	−0.84	−0.86	−0.92	−0.95	−1.04
<sup>238</sup> U	−0.06	0.23	−0.63	−0.62	−0.71	−0.79	−0.90
<sup>237</sup> Np	0.75	1.11	−0.51	−0.56	−0.65	−0.73	−0.88
<sup>238</sup> Pu	−0.16	0.12	−1.25	−1.33	−1.36	−1.41	−1.50
<sup>239</sup> Pu	−0.79	−0.72	−1.44	−1.46	−1.58	−1.61	−1.71
<sup>240</sup> Pu	0.14	0.12	−0.93	−0.91	−1.02	−1.13	−1.27
<sup>241</sup> Pu	−0.80	−0.88	−1.25	−1.21	−1.26	−1.33	−1.39
<sup>242</sup> Pu	0.73	0.79	−0.65	−0.48	−0.73	−0.92	−1.13
<sup>241</sup> Am	0.71	0.90	−0.56	−0.54	−0.65	−0.77	−0.91
<sup>242m</sup> Am	−1.66	−	−2.03	−1.87	−2.08	−2.10	−2.16
<sup>243</sup> Am	−0.15	−0.20	−0.84	−0.65	−0.85	−1.01	−1.15
<sup>242</sup> Cm	−0.18	−	−1.26	−1.34	−1.37	−1.41	−1.51
<sup>244</sup> Cm	−1.12	−1.19	−1.54	−1.44	−1.53	−1.64	−1.71
<sup>245</sup> Cm	2.44	−	−2.70	−2.69	−2.71	−2.74	−2.77

<sup>a</sup>  $r$  = moderator-to-fuel ratio.

<sup>b</sup>  $\phi = 2.5 \times 10^{14}$  n/cm<sup>2</sup> s.

<sup>c</sup>  $\phi = 1 \times 10^{15}$  n/cm<sup>2</sup> s—Sodium-cooled burner configuration SFR with oxide or metal fuel, sodium-cooled SUPERPHENIX FR, He-cooled fast reactor GFR, and lead-cooled fast reactor LFR.

equilibrium if the neutron excess compensates for parasitic captures (e.g., by structures, FPs, etc.) and for neutron leakage. In the case of a positive *D*, neutron consumption in the fuel dominates over neutron production, and the core requires a supplementary neutron source to support transmutation.

### 2.1.3. Neutron balance (*D*-factor) intercomparison

The *D*-values defined above can be used to compare the transmutation potential of a large variety of reactors, both with thermal and fast spectra [5,7]. In Table 2, the following systems are compared: a standard PWR (moderator-to-fuel ratio  $\sim 2$ ) with mixed oxide (MOX) fuel along with a variation in the LWR moderator-to-fuel ratio,  $r$ , a VHTR, and FRs with differing fuels and coolants. In the case of LWRs, *D<sub>i</sub>* depends significantly on the level of the neutron flux  $\phi$ .

The results allow comparison of the feasibility of transmutation of the different isotopes in each reactor concept. As an example, in the case of the Am isotopes, the <sup>241</sup>Am transmutation is a neutron-consuming process in any LWR concept, relatively independent from the moderator-to-fuel ratio ( $r$ ).

Significant variations in the *D*-factor are observed between the FR concepts. In general, a harder neutron spectrum leads to a more favorable neutron balance; thus, the metal-fueled SFR provides the most excess neutrons for every actinide isotope. However, all of the FR systems exhibit a significantly more favorable neutron balance compared to the MOX-LWR results.

In summary, these results indicate that fast neutron spectrum systems have a marked advantage in terms of neutron balance over thermal neutron systems.

## 2.2. Implementation in different types of reactor

The fundamental principles of transmutation, as discussed previously, have been applied in the analysis and performance assessment of different types of reactor: thermal neutron reactors, critical FRs, and subcritical source-driven reactors. The analysis has been mostly applied to the TRU transmutation, and it will be summarized in the next three sections (2.2.1–2.2.3). Section 2.2.4 will be devoted to long-lived fission-product transmutation. Finally, the issue of the impact of transmutation on the fuel cycle will be discussed in Section 2.2.5.

### 2.2.1. Transmutation performances of thermal reactors

The transmutation performances of different thermal neutron reactors have been widely investigated in the last decade. Here, we will focus on PWRs, high-temperature reactors (HTRs), and on the use of inert matrix fuels (i.e., without U matrix) in PWRs. As indicated above, the crucial issues related to the fuel cycle characteristics will be discussed in Section 2.2.5.

#### *PWR transmutation performance: Pu and MA multi-recycling in PWRs with “MOX-UE” assemblies*

In [7], the multi-recycling of the MA species, in addition to the Pu, was considered at equilibrium in a PWR concept (called MOX-UE, a French acronym for “Oxydes Mixtes avec Uranium Enrichi”) developed in order to allow multiple recycle of TRUs. Successive cases were analyzed with Pu-only, Pu + Np, Pu + Np + Am, and Pu + Np + Am + Cm recycle. In each case, the average content of Pu + MA in the core was set at 10% (e.g., 7.7% Pu and 2.3% MA when all MA are recycled: 0.3% Np, 0.6% Am, and 1.4% Cm). The results are shown in Table 3, where masses in Kg are normalized to the power produced and expressed in Terawatt hours electrical (Kg/Twhe).

**Table 3**  
Pu + MA multi-recycling in MOX-UE PWRs [7].

Inventory type	Closed cycle for Pu-only. Average Pu content ~10%	Closed cycle for Pu + Np. Av. Pu + Np content ~10 %	Closed cycle for Pu + Np + Am. Av. Pu + Np + Am content ~10%	Closed cycle for Pu + MA. Average Pu + MA content ~10%
$^{235}\text{U}/\text{U}$ in feed	4.9%	6.45%	9.0 %	6.45 %
$M(\text{Pu})^{\text{in}}$ (kg/TWhe)	226	211	190	163
$M(\text{Pu})^{\text{out}}$ (kg/TWhe)	170	168	159	142
$\Delta M(\text{Pu})$ (kg/TWhe)	56	43	31	21
$\text{MA}^{\text{(in)}}$ (kg/TWhe)	0	10.5	26.2	47.7
$\text{MA}^{\text{(out)}}$ (kg/TWhe)	17.1	20.4	30.4	44.5
$\text{Np}^{\text{(in)}}$ (kg/TWhe)	0	10.5	10.1	7.2
$\text{Np}^{\text{(out)}}$ (kg/TWhe)	1.38	7.1	7.7	5.6
$\text{Am}^{\text{(in)}}$ (kg/TWhe)	0	0.0	16.2	13.1
$\text{Am}^{\text{(out)}}$ (kg/TWhe)	9.89	8.4	14.2	11.8
$\text{Cm}^{\text{(in)}}$ (kg/TWhe)	0	0.0	0.0	27.3
$\text{Cm}^{\text{(out)}}$ (kg/TWhe)	5.87	4.9	8.5	27.1

To counterbalance the “poison” effect of MA, a higher percentage of  $^{235}\text{U}$  is needed with Np and Am recycle; the enrichment steadily increases from 4.9% with Pu-only recycle to 6.45% (Pu + Np) to 9.0% (Pu + Np + Am). A reverse trend is observed (down to 6.45%  $^{235}\text{U}$  enrichment) when the Cm is also recycled. However, note that the results shown in Table 3 indicate that the MA inventory is dominated by Cm when all MA elements are recycled; and this fuel would be very difficult to handle. In fact, only with full MA recycle can the MA inventory be stabilized, but the high Cm content of this case will severely complicate fuel handling (see Section 2.2.5). The TRU destruction in the case of full TRU multi-recycling will then be  $\sim 0.2$  g/MWd (i.e. Megawatt day) [5] to be compared to the data relevant to FR (see Section 2.2.3).

However, recycling of Np, while providing additional mass reduction for the repository, has minimal effect on the radiotoxicity, dose rate, and long-term heating rate, compared to the Pu-only case. Additionally, recycling Am with Pu + Np provides significant benefits to the fuel cycle, repository performance, and the proliferation resistance of the fuel.

The benefit provided by this approach is, however, limited, if  $^{244}\text{Cm}$  is buried in the repository because of the decay of this nuclide to  $^{240}\text{Pu}$ , which dictates the intermediate-term radiotoxicity and heat loads. The partial separation of Pu and Am by isotopic separation helps in this regard, as the amount of  $^{244}\text{Cm}$  to be sent to the repository would be decreased. The separation of Am from Cm, however, presents technological problems that have to be addressed, in particular, for the implementation at an industrial level of candidate processes that have been demonstrated at laboratory scale. Additionally, isotopic separation on the scale that would support the transmutation mission could be quite expensive and make the partial separation option unattractive. Finally, issues related to the increased  $^{238}\text{Pu}$  content, high helium build-up, potential local voiding positive reactivity effects, and cost of increased uranium enrichment, should be carefully evaluated in a detailed feasibility study.

TRU recycling provides the most benefit to the repository, both in terms of the radiotoxicity and dose rate, and the heat load. While this option is also beneficial from a proliferation-resistance viewpoint, its application would very much complicate fuel handling and makes it most expensive because of the shielding requirements (see Section 2.2.5). The impact of helium build-up (due to  $\alpha$  emissions) with TRU recycle on fuel performance is another issue that requires additional investigation.

#### Transmutation performance of VHTRs and IMF PWRs

To incinerate plutonium, neptunium, and americium nuclides, General Atomics (GA) has proposed the deep-burn modular helium-cooled reactor (DB-MHR) concept [8]. A study [9] has been performed to investigate the feasibility of this deep-burn transmutation concept by confirming the TRU consumption. This indicates that the overall TRU consumption of the total heavy metal is 58%. The consumption of plutonium is 62%, including 97%  $^{239}\text{Pu}$  depletion. About 55% of the  $^{237}\text{Np}$  is destroyed in the DB-MHR core.

In the same study [9], the use of inert matrix fuel (IMF) has been considered for the transmutation of TRUs in LWRs. This concept is very similar to the DB-MHR concept because non-uranium fuels are utilized. The primary differences are the reactor type and the fuel form, as the LWR IMF concept uses an LWR core and an inert-matrix (oxide solid solution) fuel. For a comparison of the two systems' performance, the LWR IMF concept has been evaluated using the same initial TRU vector utilized in the DB-MHR. It is noted that this is purely a neutronic comparison that has not evaluated the feasibility of the systems from a safety viewpoint. The specific power density of the DB-MHR core is about 670 W/g, while that for the LWR IMF concept is 360 W/g.

It was found that the discharge burnup of the DB-MHR core is very close to that of the IMF core (546 GWd/t versus 545 GWd/t), but the TRU consumption of the DB-MHR core is slightly higher than that of the IMF concept. It is worthwhile

**Table 4**

Comparison of fast reactor transmutation performance (CR = 0.25).

System	SFR	GFR	LFR
Net TRU destruction, g/MWt-day	0.74	0.76	0.75
System power, MWt	840	600	840
Outlet temperature, °C	510	850	560
Thermal efficiency, %	38	45	43
Power density, W/cc	300	103	77
TRU inventory, kg	2250	3420	4078
Fuel volume fraction, %	22	10	12
TRU enrichment, % TRU/HM	44–56	57	46–59
Fuel burnup, GWd/t	177	221	180

**Table 5**

MA transmutation performance of different fuel-type cores (loaded MA: 5 wt% of total fuel).

Element	Oxide core			Nitride core			Metal core		
	Transmutation			Transmutation			Transmutation		
	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)
Np	764	112	14.7	756	114	15.1	728	105	14.4
Am	746	72	9.7	728	83	11.4	695	78	11.3
Cm	150	–38	–25.4	148	–36	–24.4	139	–32	–23.1
MA total	1659	147	8.8	1631	161	9.9	1562	151	9.7

to note, that, despite issues that must still be clarified (e.g., the impact on the repository of the spent fuel composition if no recycle is envisioned), the so-called “deep burn” option is still the object of study, particularly in the US.

### 2.2.2. Transmutation in FRs. Implementation and performance

The recycling of plutonium and MA in fast-spectrum critical systems can be carried out both in homogeneous and heterogeneous modes [10–12].

In the homogeneous recycling mode, unseparated TRUs are recycled together, while, in the heterogeneous mode, the minor actinides are separated from plutonium, placed as targets in specific subassemblies, and managed independently from the standard fuel, which contains plutonium.

The effects of different fuel types and different coolants have also been studied and will be compared in terms of transmutation performances. Moreover, critical or subcritical fast-neutron transmutation systems have been envisioned, and both systems’ performances will be recalled in what follows.

#### Homogeneous recycling: transmutation and mass balance in critical FRs

The FR systems are designed to operate on TRU-based fuels with continuous recycle. Recycle technologies avoid direct disposal of the spent fuel; in particular, the TRUs are removed from the spent fuel (reducing the long-term heat, dose, and radiotoxicity) and recycled in advanced reactors for consumption.

As regards the difference in TRU destruction rates among different FR concepts based on different coolants (SFR: Na-cooled; GFR: gas-cooled; LFR: lead-cooled), Table 4 shows, as an example [13], a few parameters relevant to different particular concepts (for a conversion ratio equal to 0.25). Of course, the TRU consumption rate depends on the FR core conversion ratio, CR, and the maximum theoretical consumption rate,  $\sim 1$  g/MWt-day, is obtained for a CR = 0 (i.e., for a fuel that does not contain U). For example, in an SFR with CR = 0.5, the net TRU destruction will be 0.47 g/MWt-day, to be compared to the 0.74 g/MWt-day given in Table 4.

Note that, since the transmutation physics behaviour is similar for the fast burner concepts, for a given conversion ratio, the TRU destruction rate and compositions are very similar. However, variations in other fuel-cycle performance parameters are observed because of design differences, as shown in Table 4.

As regards fuel type (oxide, metal, nitride, carbide), each type of FR system has its specific requirement. However, the choice of the chemical form of fuel (oxide, nitride, carbide, etc.) has only limited effects on the transmutation performances, as was shown in Table 2, where the  $D$ -factors were compared for different FR concepts, which differ for fuel type and spectrum hardness, these last characteristics being associated with the conversion ratio value. Dense fuels, like metal and nitride fuels, and low conversion ratios allow potentially better transmutation performance.

A comparison of MA transmutation effectiveness in different fuels and coolant systems has also been reported in [14]. In that work, three types of FR fuel – oxide, nitride and metal – were considered. Table 5 shows the evaluated MA transmutation effectiveness of each fuel-type core where MA was loaded at 5 wt% of total fuel. The total transmutation effectiveness of nitride and metal-fueled cores is 9.9% and 9.7% per year, respectively, both a little better than that of the oxide-fueled core. The difference can be attributed to the harder neutron spectrum of the new fuel-type cores.

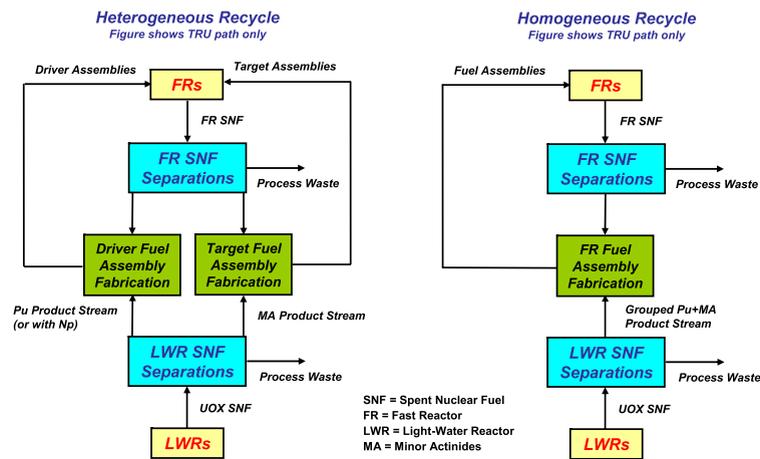


Fig. 6. Heterogeneous and homogeneous recycle schemes.

Once more, three types of coolant (sodium, lead, and gas) were compared in the same reference with respect to their impact on MA transmutation effectiveness. Although there are many differences in design parameters, including thermal power, operation cycle length and fuel-type etc., a rough evaluation of MA transmutation characteristics may be possible by normalizing the results with respect to thermal power and operating period.

Table 6 summarizes the MA transmutation effectiveness of each coolant-type core. After normalization, the MA transmutation effectiveness of these cores is almost identical to a value of 7.5%–7.7% per year. The ratio of transmuted MA to loading is a little worse in the case of lead coolant, but the difference is rather small compared to the dependence on other core parameters, like core-fuel inventory. As a conclusion, the effect of coolant choice in FR design will be negligible from the viewpoint of the MA transmutation.

*Homogeneous recycling: impact on reactivity coefficients.* The impact of MA on reactivity coefficients varies according to the reactor size and the type of coolant. However, in general, the introduction of Am and Np in the core has a beneficial effect on the reactivity variation with burnup (which becomes less negative, allowing, in principle, longer irradiation times) but can have undesired effects on some other reactivity coefficients. In fact, the Doppler reactivity coefficient becomes less negative and, in the case of SFRs, the coolant void reactivity coefficient becomes more positive. Moreover, the delayed neutron fraction will become smaller. However, the impact of MA loading increase on reactivity coefficients is very much related to the approach taken in core and fuel design, and it can be optimized according to predefined objectives and constraints [15].

*Heterogeneous recycling.* The homogeneous multi-recycle of TRU fuels (i.e. with unseparated TRUs) in an FR will end up with the production and the slow accumulation of Cm isotopes before reaching equilibrium. This build-up will be present whatever the Pu/MA ratio in the fuel, but will be somewhat mitigated by a high Pu/MA ratio. One significant consequence can be the strong increase of the neutron sources at fuel fabrication, with respect to the case of a Pu-only multi-recycle. In fact, if no specific measure is taken (cooling time optimization, blending of fresh and irradiated fuels, etc.), the neutron sources can increase by a factor of 100 or more, essentially due to the build-up of Cm-244, a relatively strong neutron emitter from spontaneous fission. A more detailed description of this point will be given below.

In this respect, an alternative to the homogeneous recycle in FRs could be to separate out the less radioactive component of the LWR spent nuclear fuel TRUs (e.g., Pu and Np) in order to make driver fuels and use the remaining MAs (primarily Am and Cm) in target fuels/assemblies (see, e.g., [10–12]) or stored as waste or for use in homogeneous fuels in the future. Consequently, the driver and target fuels can be managed separately in the fuel cycle. Heterogeneous recycling makes it possible to disconnect the MAs cycle from the conventional fuel cycle. In particular, the fabrication of the assemblies with MA can be done in a separated plant, and the specific constraints due to MAs handling (heat, neutron sources) are not applied to the conventional fuel fabrication plant. This separate management and recycle of the Pu–Np driver and MA target fuels constitutes the so-called heterogeneous recycle, as shown in Fig. 6.

The concept of heterogeneous recycle implies that the plutonium and MAs are managed separately in the core and fuel cycle. In that case, core driver and target zones are defined. In the driver fuel, the traditional [U, Pu] fuel is typically assumed. Potential difficulties associated with the heterogeneous recycle include the following.

- Difficulty of recycling, handling, and fabricating *target* assemblies.
- Immature state of target technology (fabrication, irradiation performance, etc.).
- Fuel behaviour under irradiation (swelling and helium production, mainly due to the  $\alpha$  decay of  $^{242}\text{Cm}$ , built up from the neutron capture in  $^{241}\text{Am}$ ).
- High specific heat for assembly manufacturing.
- High decay heat level for in-core and out-of-core assembly handling.
- High neutron source level at the fuel treatment step.

**Table 6**  
MA transmmutation performance of different coolant-type cores (loaded with 5 wt% MA in fuel).

Element	Sodium-cooled core				Lead-cooled core				CO <sub>2</sub> gas-cooled core			
	Transmutation				Transmutation				Transmutation			
	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)	Initial amount at BOEC (kg)	Amount (kg)	Effectiveness (%)
Np	1058	201	19.0	326	28	8.5	961	118	12.3			
Am	1262	148	11.7	364	25	6.8	1176	81	6.9			
Cm	321	-52	-16.2	64	-8	-12.1	228	-34	-15.0			
MA total	2641	297	11.3	754	45	5.9	2366	165	7.0			
Normalized MA transmmutation	695 kg/GWth	53 kg/GWth/year	7.6%/year	1077 kg/GWth	83 kg/GWth/year	7.7%/year	657 kg/GWth	49 kg/GWth/year	7.5%/year			

**Table 7**  
Effective delayed-neutron  
fraction for selected ac-  
tinides.

Nuclide	$\beta$
$^{238}\text{U}$	0.0172
$^{237}\text{Np}$	0.00388
$^{238}\text{Pu}$	0.00137
$^{239}\text{Pu}$	0.00214
$^{240}\text{Pu}$	0.00304
$^{241}\text{Pu}$	0.00535
$^{242}\text{Pu}$	0.00664
$^{241}\text{Am}$	0.00127
$^{243}\text{Am}$	0.00233
$^{242}\text{Cm}$	0.000377

### 2.2.3. Transmutation in dedicated transmuters: external source-driven subcritical reactors and critical low conversion ratio FRs

Potential safety-related problems were indicated in early transmutation studies in the case of an FR core loaded with only TRUs (i.e., the conversion ratio equals zero) and with a high content of MAs. In fact, in these types of FR core, the absence of uranium produces as a consequence both a very low fraction of delayed neutrons (see Table 7) and a very low Doppler reactivity coefficient (in general, mostly due to  $^{238}\text{U}$  capture). Moreover, we have seen that a high content of MAs like Am isotopes and Np induces, in principle, a deterioration of the void reactivity coefficient (in the case of liquid metal coolants; in particular, in the case of Na coolant).

Subcritical systems (or accelerator-driven systems, ADSs; see, e.g., [16]) were “rediscovered” (~1985) because they provide, in principle, a way out from these potential difficulties. More recently, fission–fusion hybrid systems [17,18] have also been “rediscovered”, and their potential as dedicated waste transmuters has been pointed out. A dedicated transmuter should be able to burn as much TRUs or MAs as possible according to the chosen strategy. “Transmutation” in this case means essentially “fission”. This is a very important point because, to compare the “transmutation” effectiveness of different systems, one has to compare the system performance at the same power (i.e., accounting for the same number of fissions). Then, what really matters is the fuel loaded in the “transmuter” because it determines which isotopes will be fissioned. A pure MA-fueled core (if feasible) obviously maximizes the MA destruction if the power density can be kept high enough, just as a pure TRU (no U)-fueled core maximizes TRU destruction if the power density can be kept high enough. In principle, since for each actinide  $\sim 1$  g is burnt (by fission) for 1 MWT-day, the total mass  $M_{F,i}$  (in kg) of isotope  $i$  burnt by fission in a year is given by

$$M_{F,i} = \frac{y}{f_i} \times W \times 365 \times 10^{-3}$$

where  $y$  is the load factor and  $f_i$  is the ratio of the total number of fissions in the system (all isotopes, all regions) to the fissions in the core due to isotope  $i$  and  $W$  is the power.

$M_{\text{Tot},i}$  is the total mass of isotope  $i$ , consumed both by fission and by capture:

$$M_{\text{Tot},i} = M_{F,i} \times (1 + \alpha_i),$$

where  $\alpha_i$  is the average capture-to-fission ratio of isotope  $i$ .

This formulation indicates that, in principle, the “burning” potential of a core is related to its power—i.e., to the fission rate in that particular core. In this sense, any core with the same power shows a comparable transmutation potential. The real difference is related by the amount and the “quality” (e.g., the ratio MA/Pu) that a particular core type does allow. In this sense, we will see that, when discussing dedicated fuel issues, a theoretical maximum burning capability should be evaluated in terms of appropriate fuel type development.

The work reported in [14] was devoted to the comparison of ADSs and critical FRs. In terms of transmutation performance, it was found that the two types of system are comparable. As regards the choice of coolant and fuel type, the indications given above for critical FRs will hold also for ADSs. Despite the fact that one could, in principle, reach in an ADS the theoretical maximum TRU consumption rate (i.e.,  $\sim 1$  g/MWT-d), in practice, lower TRU consumption rates on the order of  $\sim 0.75$  g/MWT-d have been found, very close to the performance of a critical burner FR with conversion ratio (CR) equal to 0.25.

Fertile-free fuel can be envisioned for ADSs in order to maximize the ADS support ratio in the power-producing energy complex. As indicated previously, the neutronic properties of fertile-free TRU or MA fuel (and in particular the associated low delayed neutron fraction), make it much more difficult to design a critical FR with this type of fuel. Therefore, the ADS concept can more readily accept U-free fuels and reach high transmutation performances. This means, in particular, that ADS-based transmutation will require a smaller number of transmuters to handle the TRU arising from the electricity-producing reactor fleet, and will allow for a separate stratum of the fuel cycle, leaving the fuel cycle stratum devoted to electricity production uncontaminated by the presence of MA.

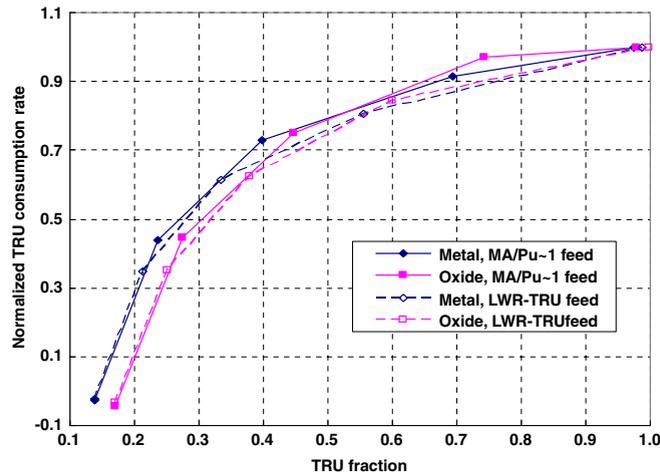


Fig. 7. TRU consumption versus TRU fraction in the fuel.

Subcritical systems (ADSs) can also provide an option, if critical FR deployment is delayed. This type of system (ADS) can provide a tool to decrease TRU or MA stocks within a wide range of fuel types and compositions. However, feasibility issues related to the development of U-free fuels (as of today, no definitive candidate has been found, even if some promising candidates have been pointed out; see Section 3.4) can suggest exploring ADSs with some uranium in the fuel, corresponding to very low conversion ratio values (e.g., less than 0.25), for which a critical core could still present safety difficulties.

The physics features of ADSs (e.g., relationship between the subcriticality level and accelerator current, relationship between the external source and the intrinsic fission source, etc.) allow for judgment on the interest, feasibility, and cost of transmutation in an ADS as compared to transmutation in a critical FR. As we will discuss in the following, low conversion ratio FRs can also offer an interesting alternative if the TRU “burning” is considered a priority objective.

The ADS concept, initially investigated in the US [19] and successively in Japan [20] and Europe, is still being actively investigated in Europe. In fact, after a first assessment and the proposal of a possible roadmap towards implementation [21], a large research and development (R&D) program sponsored by the European Union (EU) (EUROTRANS, [22]) has been recently completed. Several experimental programs have also been performed, e.g., to validate innovative components as the spallation target [23] or the physics of a subcritical system [24]. Some work is still being performed in Japan, but the US has practically no research activity left in that area.

#### Low conversion ratio FRs

The specific issue of U-free fuels has been advocated in order to maximize TRU consumption; studies devoted to Pu consumption in FRs [25] pointed out a gradual saturation of the Pu consumption with the reduction of the amount of uranium in the fuel. Using a 1000 MW advanced burner reactor (ABR) core design [26], the relation between TRU consumption rate and TRU conversion ratio has been investigated [27]. Both metal and oxide core designs were investigated for TRU conversion ratios of 1.0, 0.75, 0.50, 0.25, and 0.0, both with Pu/MA ratio  $\sim 9$  and with Pu/MA  $\sim 1$  in the feed fuel.

Fig. 7 shows the TRU consumption rates relative to the maximum theoretical value of uranium-free fuel as a function of TRU fraction in the charged fuel for both cases.

Similar trends are shown for both cases (i.e., makeup TRU feed from LWRs with Pu/MA  $\sim 9$  and makeup TRU feed with Pu/MA  $\sim 1$ ) and for both types of fuel. The slope of TRU consumption rate with respect to TRU fraction in the fuel decreases with increasing TRU fraction. The TRU consumption rate reaches  $\sim 80\%$  of the maximum theoretical value when the TRU fraction is  $\sim 60\%$ , which corresponds to TRU conversion ratio in the range  $\sim 0.25$ – $0.35$ .

The effective delayed neutron fraction, Doppler constant, radial expansion coefficient, axial expansion coefficient, and sodium void worth at the end-of-irradiation cycle (EOC) have been studied as a function of the TRU fraction in heavy metal (HM). As an example, as the TRU fraction in the HM increases, the effective delayed neutron fraction decreases monotonically because of the reduced fission of  $^{238}\text{U}$  (see Fig. 8).

However, from the results reported in [27], it is clear that cores with conversion ratio in the range 0.25–0.40 are, in principle, feasible, as parameters relevant for the safety performance of these cores become comparable to those of cores that have already proven to be feasible. Since these cores allow TRU consumption, whatever the Pu/MA ratio and fuel type, close to 80% of the maximum theoretical consumption, it seems that U-free fuels can be avoided for any scenario and specific P&T strategy.

#### 2.2.4. Transmutation of long-lived FPs (LLFPs)

In this section, we will discuss the potential of FRs in order to transmute long-lived fission products (LLFPs). A detailed analysis of FP radiotoxicity [28] reveals that a few long-lived radionuclides (e.g.,  $^{129}\text{I}$ ,  $^{99}\text{Tc}$ , etc.; see Table 8) contribute to the

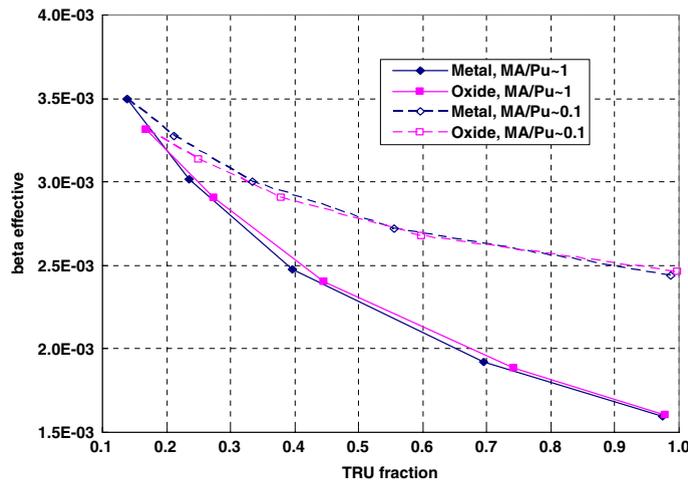


Fig. 8. Delayed neutron fraction versus TRU fraction in the fuel.

Table 8

Properties of main long-lived FPs (LLFP).

Isotope	Half-life (y)	Type of decay	Thermal power (W/Bq)	Dose (ingestion) (Sv/Bq)	Fraction in irradiated fuel (g/t) <sup>a</sup>
<sup>14</sup> C	$5.7 \times 10^3$	$\beta$	$1.6 \times 10^{-14}$	$5.7 \times 10^{-10}$	$1.3 \times 10^{-1}$
<sup>36</sup> Cl	$3.0 \times 10^5$	$\beta^-$ , $\beta^+$	$4.4 \times 10^{-14}$	$8.2 \times 10^{-10}$	$1.6 \times 10^0$
<sup>79</sup> Se	$6.5 \times 10^4$	$\beta$	$6.5 \times 10^{-15}$	$2.3 \times 10^{-9}$	$4.7 \times 10^0$
<sup>90</sup> Sr	$2.9 \times 10^1$	$\beta$	$2.8 \times 10^{-14}$	$3.9 \times 10^{-8}$	$5.0 \times 10^2$
<sup>90</sup> Y	$7.3 \times 10^{-3}$	$\beta$	$1.5 \times 10^{-13}$		$1.3 \times 10^1$
<sup>93</sup> Zr	$1.5 \times 10^6$	$\beta$	$2.6 \times 10^{-15}$	$4.2 \times 10^{-10}$	$9.8 \times 10^1$
<sup>99</sup> Tc	$2.1 \times 10^5$	$\beta$	$1.4 \times 10^{-14}$	$3.4 \times 10^{-10}$	$8.2 \times 10^2$
<sup>107</sup> Pd	$6.5 \times 10^6$	$\beta$	$1.4 \times 10^{-15}$	$3.7 \times 10^{-11}$	$2 \times 10^2$
<sup>126</sup> Sn	$1 \times 10^5$	$\beta$	$4.2 \times 10^{-14}$	$5.1 \times 10^{-9}$	$2.0 \times 10^1$
<sup>126</sup> Sb	$3.4 \times 10^{-2}$	$\beta$	$5.0 \times 10^{-13}$		$6.9 \times 10^{-6}$
<sup>129</sup> I	$1.6 \times 10^7$	$\beta$	$1.3 \times 10^{-14}$	$7.4 \times 10^{-8}$	$1.7 \times 10^2$
<sup>135</sup> Cs	$2.3 \times 10^6$	$\beta$	$9 \times 10^{-15}$	$1.9 \times 10^{-9}$	$1.3 \times 10^3$
<sup>137</sup> Cs	$3.0 \times 10^1$	$\beta$	$3.2 \times 10^{-14}$	$1.4 \times 10^{-8}$	$1.1 \times 10^3$
<sup>137m</sup> Ba	$4.9 \times 10^{-6}$	$\beta$	$1.1 \times 10^{-13}$		$1.7 \times 10^{-4}$
<sup>151</sup> Sm	$9.0 \times 10^1$	$\beta$	$3.2 \times 10^{-15}$	$9.1 \times 10^{-11}$	$1.6 \times 10^1$

<sup>a</sup> PWR-UOX (3.5 % U-235 enrichment, BU = 33 GWd/t).

very-long-term radiotoxicity. However, their absolute magnitude remains below the TRU radiotoxicity and even below the radiotoxicity of the natural ore materials removed to fabricate enriched uranium. The total radiotoxicity of FPs is about  $1.4 \times 10^7$  Sv/ton U (enriched) 100 years after discharge, but it decreases to 875 Sv/ton U (enriched) after 1000 years. Thereafter, it is stabilized at that level for a long time ( $\sim 100,000$  years), i.e., at a level much lower than our reference level for natural ore.

As for the possibility and limits to transmute FPs in order to reduce radiotoxicity, transmuting FPs is of very little interest. The majority of the FPs have decayed after about 250 years (see Table 8), and their contribution to the radiotoxicity of the spent fuel, which was very high during the first 100 years of storage, has become low. However, some FPs are very mobile in certain geological environments and can thus contribute significantly to the radiological effects of disposal in underground repositories. In addition, the treatment of spent fuel results in releases through gaseous and liquid effluents which also contribute to the long-term radiological effects of nuclear power generation. The FPs that deserve most attention in this respect are <sup>129</sup>I, <sup>135</sup>Cs, <sup>79</sup>Se, <sup>99</sup>Tc, and <sup>126</sup>Sn.

Unlike transuranics, FPs in a transmutation process produce no supplementary neutrons but are purely consumers. As seen in Section 1, neutron consumption is the most important parameter if one wants to assess the potential of transmutation in a given nuclear system.

The LLFP transmutation is related to a large neutron surplus availability (in units of neutrons/fission). In view of the application to a reactor fleet, it is necessary to evaluate the fraction  $f$  of reactors that are necessary within the fleet to perform the LLFP transmutation. It has been shown [28] that the fraction  $f$  of reactors needed varies between 8% and 15%, i.e., a very large (and unrealistic) fraction of the fleet.

Finally, it should be emphasized that heat production is essentially related to <sup>90</sup>Sr and <sup>137</sup>Cs, which are not candidates for transmutation (their short half-life is such that no transmutation process can provide a comparable “transmutation half-life”, evaluated as the product of the microscopic capture cross section of the isotope considered times the neutron flux

**Table 9**

Consequences at fuel fabrication in the case of recycling strategies in FRs and PWRs.

Reactor type	PWR		FR			ADS
	Fuel type		Pu only	Full TRU (Homogeneous recycle)		
Parameter	MOX (Pu only, reference)	Full TRU recycle	Pu only	Full TRU (Homogeneous recycle)		MA-dominated fuel
Decay heat ( $\text{W g}^{-1}$ HM)	1	$\times 3$	$\times 0.5$	$\times 2.5$		$\times 20\text{--}80$
Neutron source ( $\text{n s}^{-1} \text{g}^{-1}$ HM)	1	$\times 8000$	$\sim 1$	$\times 150$		$\times 20,000$

**Table 10**

Cm and Cf isotope properties.

Nuclide	Half-life	Neutrons/g/s		$E\gamma$ (keV)	$\gamma$	nSv/Bq (ICFR-72)
		Spontaneous	( $\alpha, n$ )			
$^{242}\text{Cm}$	163 d	$1.72 \times 10^7$	$4.18 \times 10^6$	1.4		12
$^{243}\text{Cm}$	30.0 y		$6.09 \times 10^4$	133.2	104 keV (24%) 228 keV (11%) 278 keV (14%)	150
$^{244}\text{Cm}$	18.1 y	$1.01 \times 10^7$	$8.84 \times 10^4$	1.3		120
$^{245}\text{Cm}$	$8.50 \times 10^3$ y			93.8	104 keV (30%) 100 keV (18%)	210
$^{246}\text{Cm}$	$4.73 \times 10^3$ y	$\approx 7 \times 10^6$	$\ll \text{SF}$	3.0		210
$^{247}\text{Cm}$	$1.60 \times 10^7$ y			302.8	403 keV (69%)	190
$^{248}\text{Cm}$	$3.40 \times 10^5$ y	$\approx 3 \times 10^7$	$\ll \text{SF}$	579.1	579 keV (100%)	770
$^{249}\text{Cf}$	351 y		n.a.	329.2		350
$^{250}\text{Cf}$	13.1 y	$\approx 8 \times 10^9$	$\ll \text{SF}$	6.3		160
$^{251}\text{Cf}$	898 y		n.a.	120.3		360
$^{252}\text{Cf}$	2.64 y	$\approx 10^{12}$	$\ll \text{SF}$	217.4		90

[units:  $\text{s}^{-1}$ ]). The best way to handle these two isotopes is probably to recover them with an appropriate chemical process, then store them and let them decay.

In summary, the transmutation of the so-called LLFPs is no longer envisioned by any major international program. The most important issue is generally recognized to be the impact on the repository of the heat produced by  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . We will come back to this issue in Section 4.

### 2.2.5. An essential challenge: transmutation consequences on the fuel cycle

As indicated above, the consequences of the TRU multi-recycle can significantly affect the fuel cycle. Table 9, taken from [29], summarizes the consequences at the fuel fabrication step for some of the FR concepts considered previously, with a comparison to the impact of TRU recycle in PWRs.

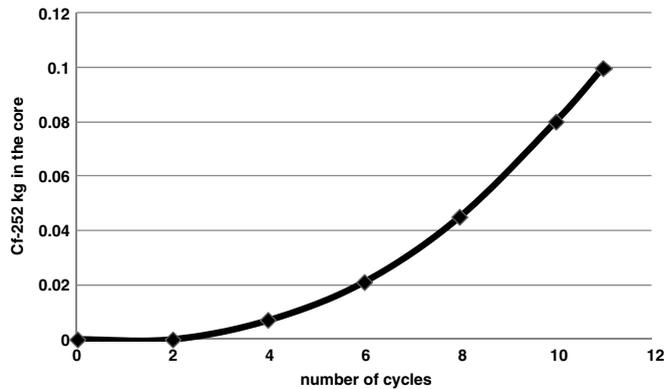
In the case of homogeneous recycling in PWRs and FRs, the large difference in the neutron source at fabrication is essentially due to the impact of the  $^{252}\text{Cf}$  spontaneous neutron fission ( $\sim 10^{12}$  n/g/s) contribution, due to the different mechanisms of its build-up in the two different types of spectrum (see Fig. 9).

The  $^{252}\text{Cf}$ , which is a very powerful neutron emitter (see Table 10), results in an unacceptably high neutron source at fuel fabrication. This is one of the important points that suggests avoiding full TRU transmutation in thermal reactors [30].

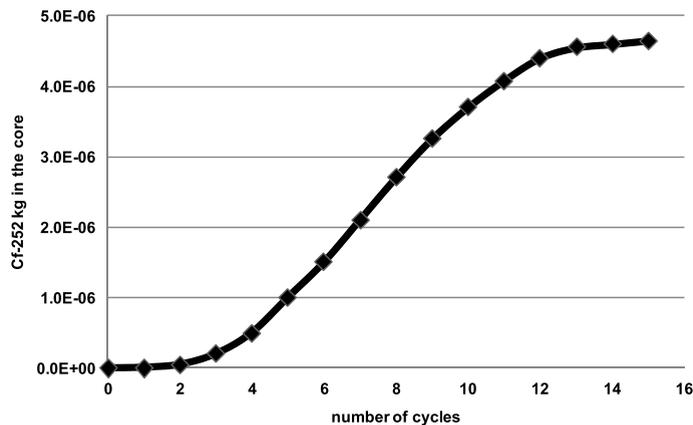
### 2.3. Summary of technological options

In summary, it can be said that, if from the point of view of the core feasibility, most options seem to be viable, the consequences on the fuel cycle (cost, feasibility, doses to the workers, etc.) seem to be more crucial. For example, apart from an increase of cost due to the need to over-enrich the core fuel of an LWR and some limitation in the amount to be loaded to avoid a deterioration of the reactivity coefficients, the full recycle of TRUs in this type of reactor is practically excluded due to the increase of neutron doses at fuel fabrication.

In the case of FRs, the constraints of the full TRU recycle on the neutronics and the safety of the core seem to be even more manageable. However, even if much less dramatic, the consequences of the TRU loading in the fuel can be significant and often justify the industry's reluctance to acknowledge the potential benefits of P&T; in this respect, the case of dedicated FRs (critical or subcritical, i.e. ADSs) can be even more affected by fuel cycle penalties. This can well be the case of the heterogeneous recycle too.



(a) Cf-252 inventory in the core. Case of full TRU multi-recycling in an LWR.



(b) Cf-252 inventory in the core. Case of full TRU multi-recycling in an FR.

**Fig. 9.** Evolution with time of the Cf-252 inventory in the irradiated fuel.

Finally, the development of the appropriate fuels, more or less loaded with TRU, has proven to be also a challenging task as we will discuss in Section 3.4. In this respect, the most challenging issue is the development of viable U-free fuels (IMF), i.e., the fuels that offer, in principle, the greatest benefits in terms of transmutation performance. This problem will also be discussed in Section 3.4; however, the potential difficulty associated with assessing an appropriate U-free fuel or target indicates the systematic use of U-based fuels, since they offer the possibility to develop more standard fuels, which are possibly much easier to reprocess with the less-exotic aqueous process, allowing for transmutation rates not too far from the maximum theoretical ones, as indicated in the previous sections.

Finally, as noted above, the transmutation of LLFPs does not seem to be realistic in view of the limited transmutation rates and the burden, in terms of reactivity loss during the cycle, on the core performances.

### 3. Status of research and development

In the last 20 years, remarkable progress has been made in key areas of interest for P&T. However, the process of implementation of advanced fuel cycles requires (1) a down-selection of options and the definition of priorities, (2) the deployment of large pre-industrial installations, and (3) a coordinated R&D effort in the key areas: physics, advanced reprocessing methods with actinide separations, innovative MA-loaded fuels, and the technologies of the different fast neutron systems (critical FR and subcritical ADS), including new materials, thermo-hydraulics, simulation tools, and nuclear data and, in the case of ADSs, the coupling of an accelerator and a subcritical core. In the following subsections, we will summarize the state of the art in the most crucial areas.

#### 3.1. Transmutation physics

The physics of transmutation, as presented in Section 2, is well understood. However, despite major progress made in the quantification of nuclear data of Pu isotopes and MAs uncertainties, R&D is still needed in order to reduce these uncertainties, and collaborative projects are under way. For example, an uncertainty analysis has been performed on two systems devoted

**Table 11**

Some features of the SFR and ADMAB systems.

System	Fuel	Coolant	TRU/(U + TRU)	MA/(U + TRU)	Power (MWth)
SFR	Metal	Na	0.605	0.106	840
ADMAB-ADS	Nitride	Pb–Bi	1.0	0.680	380

**Table 12**

Fast neutron systems with high MA content: total uncertainties (%).

Reactor	$k_{eff}$	Power peak	Doppler	Void	Burnup (pcm)	Decay heat	Dose	Neutron source
SFR	1.82	0.4	5.6	17.1	272	0.4	0.3	1.0
ADMAB	2.94	21.4	–	15.5	1044	0.7	1.0	2.5

to transmutation (one critical, called SFR, and one subcritical, called the “accelerator-driven MA burner”, ADMAB), where the fuel is heavily loaded in MA [31] (see Table 11).

For these systems, the uncertainty, due to nuclear data uncertainties, of the most important integral parameters has been investigated, and the results are summarized in Table 12.

The level of these uncertainties is significant, in particular for  $k_{eff}$ , burn-up reactivity, and void reactivity coefficients. However, these uncertainties do not prevent performance of meaningful preconceptual design studies. Successive phases of design optimization and of reduction of margins (for economical reasons) will require the reduction of these uncertainties—in particular by means of integral experiments in critical facilities.

### 3.2. Technology

The development of fast neutron spectrum reactors will require materials testing of reactors and hot cells. Additional required infrastructures are testing and qualification facilities for system technologies, components, and coolant quality control (specific liquid metal loops, gas loops, and hot cells), as well as code qualification and validation, which are mandatory for safety analyses. In a European concerted action [32], the need for experimental facilities in support of Na-cooled, heavy liquid metal (HLM)-cooled and gas-cooled fast (critical or subcritical) reactors has been pointed out.

### 3.3. Separation R&D [32]

As regards isotope chemical separation, two major technologies have been explored so far.

- Hydrometallurgical processes that benefit from more than 60 years of R&D and a proven experience of reprocessing at the industrial level, mainly focusing on the recovery of U and Pu.
- Pyro-chemical processes first studied in the 1950s and 1960s for the treatment of spent fuel from molten salt reactors and fast breeder reactors and, more recently, with a renewed interest at the end of the 1980s, for specific applications, but without reaching the industrial development level.

Considering hydrochemistry technology (the most developed technology in Europe), the processes differ from the extracting systems involved. Most of the partitioning strategies rely on a three-step approach:

- (1) Separation of U (and sometimes also Pu, or/and Np) from spent fuel dissolution liquors;
- (2) An(III) + Ln(III) co-extraction;
- (3) An(III)/Ln(III) separation, this latter step being the most difficult because of the similar chemical properties of these element groups. It is, however, mandatory for the following reasons.
  - Neutron poisoning: lanthanides (especially Sm, Gd, and Eu) have very high neutron capture cross sections, e.g., >250,000 barn for  $^{157}\text{Gd}$ .
  - Material burden: in spent LWR fuels, the lanthanide content is up to 50 times that of Am/Cm.

The processes developed around the world differ for the extracting systems involved and for the possibility of combining two of the three steps into a single one.

- For the first step, TBP (tri-butyl phosphate) is the basis of the PUREX (currently commercially implemented), UREX, and COEX<sup>TM</sup> processes, developed in Europe and the US.
- For the second step, malonamides, CMPOs (carbamoyl-methylene phosphine oxides), and TODGAs (diglycolamides) are used in the processes, respectively developed in Europe, the US, and Japan.
- Finally, for the third step, among the processes aiming at separating An(III) from Ln(III), the process developed in France, based on a selective stripping of An(III), is today the most promising route towards potential future implementation at a larger scale.

**Table 13**

Fuel types according to different transmutation strategies.

Strategy	Reactor	Fuel composition	MA concentration
Homogenous recycling (multi-recycle)	SFR	(U, Pu, MA)O <sub>2-x</sub> (U, Pu, MA)C (U, Pu, MA)N  Metal U–Pu–Zr–MA	≤5 wt% in HM
Heterogeneous recycling (multi-recycle)	SFR	IMF: (Pu, MA)O <sub>2-x</sub> + MgO or (Pu, MA)O <sub>2-x</sub> + Mo  (U, MA)O <sub>2-x</sub>	Up to ~20%  Up to ~20%
Recycling in dedicated reactors	ADS	IMF: (Pu, MA)O <sub>2-x</sub> + MgO or  (Pu, MA)O <sub>2-x</sub> + Mo (Pu, MA, Zr)N	~50–60 wt% in HM

Finally, the TRU actinide group separation (i.e., Np, Pu, Am–Cf) related to the homogeneous recycle in FRs, discussed previously, has only been scarcely investigated up until now, and many challenging technological obstacles will have to be overcome before demonstration tests can successfully be carried out. However, France is currently developing the GANEX process [33].

If they have shown potential viability at the laboratory scale, the different processes have reached various levels of laboratory-scale demonstration and are more or less suitable for industrial implementation with respect to process development requirements (number of cycles, amount of secondary wastes generated, scale-up of equipments, process control, sturdiness, safety analysis, etc.) or to solvent management (treatment for recycling, by-products management, etc.).

Finally, considering pyrochemistry technology, crucial technological issues should still be explored: material corrosion, online monitoring, and salt motion in pipes. Beyond the goal of developing separation processes, basic research, research infrastructures, and tools also represent a major stake to sustain these developments and improve European knowledge as well as the predictive power of simulation tools.

### 3.4. Fuels for transmutation

This section presents a short summary of the present state of the art as regards transmutation fuels according to the major strategies presented in Section 2. Once more, all references to specific literature have been omitted, since they can be found in [34]. The development of the fuels for the advanced fuel cycles is a crucial and challenging issue. The different types of fuels envisioned, according to the different strategies and scenarios described in Section 2, are summarized in Table 13.

The following remarks for each class of fuel can be made.

(1) Fuels for MA transmutation in FRs in homogeneous mode.

*Oxide fuels.* This type of fuel can be considered an extended version of the existing MOX when a small amount of MA (few times less than amount of Pu) is added. These fuels will contain on the order of 5% MA in HM. In order to assure an effective transmutation, they should operate up to a very high burnup (~200 MWd/kg HM) and use new cladding materials resistant to high damage doses (~200 dpa). In the past experiment SUPERFACT [35], some of these fuels (loaded with Pu, Am, and Np) were fabricated and irradiated in PHENIX.

*Ceramic fuels of high density: carbides and nitrides.* Nitride fuels are extensively investigated because of their high thermal conductivity and chemical compatibility with liquid Na. The possibility of using <sup>15</sup>N isotope in nitride fuels, in the place of <sup>14</sup>N, has been analyzed to avoid the production of environmentally hazardous <sup>14</sup>C. The technology of nitride fuel production was developed only at the laboratory scale. Irradiation experiments were performed in a fast-spectrum sodium-cooled irradiation reactor in Russia (BOR-60), and some irradiations have been performed in France (PHENIX) and in Japan (JOYO). Despite advancements in the development of nitride fuels, uncertainties still exist, e.g., on their stability at high temperature. Moreover, in case of incorporation of MA in the fuel, MA volatilization is observed and should be minimized. Finally, carbide fuels are studied for GFRs only by France.

*Metallic fuels.* The advantages of these fuels are their high density and thermal conductivity. The higher density results in a harder neutron spectrum, and the high thermal conductivity allows the lowest fuel operation temperature. The best-known metallic fuel is based on the Pu–U–Zr system, which was extensively studied in the 1980s in the US. Such fuel has been loaded and extensively irradiated in the experimental breeder reactor (EBR-II) reactor in the US. For this type of fuel, an important, innovative experiment [36] has been performed in Europe, since the fuel consisted of Zr-based alloys containing a low percentage of MA (including Cm). The fuel has been successfully irradiated in PHENIX (METAPHIX experiment), and presently the post-irradiation examination (PIE) is under way.

*Fuels with inert matrix support (i.e., without fertile isotopes – IMFs), [37–39].* The main advantage of IMFs is, in principle, the possibility to destroy Pu and MA more effectively than with the fuels containing U (i.e., avoiding further Pu production).

These fuels have been envisioned both as ADS fuels and for the targets of the heterogeneous recycle mode (see Sections 2.2.2 and 2.2.3).

Different kinds of IMF have been studied during recent years in the framework of different European projects. The matrices under consideration have been Mo, MgO, and  $ZrO_2\text{-}Y_2O_3$ .

Several irradiation programs have either been completed or are under way. However, with the final shutdown of PHENIX, no irradiation facility with a fast neutron spectrum is available. This is a crucial difficulty in the development and validation of appropriate fuels for any future advanced fuel cycle.

Finally, note that no industrial-scale facility exists yet for the fabrication of fuels loaded with significant amounts of MA, and the impact of the presence of MA on fuel fabrication plants has undergone only preliminary investigation.

In summary, for the feasibility of the different fuel concepts, a sufficient level of knowledge is available only for the fuels foreseen in the homogeneous recycling in FRs. All other types of fuel still need development, either through experiments at the pin scale or through analytical irradiations devoted to the understanding of fuel behaviour under irradiation in specific temperature and neutron dose ranges.

#### 4. Potential benefits and impact on deep geological repositories of advanced fuel cycles with P&T

##### 4.1. Status of some deep geological repository programs

Remarkable progress in the last two decades has been made in the field of geological disposal, and some countries have reached important milestones. In the US, a license application for a geological repository at Yucca Mountain was submitted to the regulator in 2008. However, the current administration has removed support for this repository, so its future is uncertain. In Finland and Sweden, a repository design has been developed and a site selected for the geological disposal of spent fuel in a granite formation. In Finland, the final disposal of spent nuclear fuel at Olkiluoto is planned to start in 2020. In Sweden, it is expected that the geological repository at Forsmark will become operational in 2023. In France and Switzerland, a safety case on the geological disposal of various high-level and intermediate-level waste types in a clay formation has been submitted and reviewed by national and international committees. In Switzerland, the first phase of the site selection process was initiated in 2008. In France, studies are under way to select a site and develop a design for a disposal facility. A safety case for a license application will be submitted to the authorities in 2015. Depending on the decision of the authorities, it is expected that the facility will become operational in 2025.

In the UK, the Radioactive Waste Management Directorate (RWMD) is responsible for managing the delivery of geological disposal for higher activity radioactive wastes, as required under UK Government policy. This policy also states that the choice of a site for a geological disposal facility will be based on a volunteerism and the partnership approach. The development of the implementation program is in the early stages.

In Japan, according to the Final Disposal of Designated Radioactive Waste Program, issued in 2000 under the Law on Final Disposal of Designated Radioactive Waste, final disposal will start sometime in the latter half of the 2030s. The geological disposal is to be performed in four stages:

- Selection of acceptable geological formations (first stage)
- Selection of the candidate disposal sites (second stage)
- Demonstration of disposal technology at the candidate disposal site (third stage)
- Construction, operation, and closure of the disposal facilities (fourth stage).

##### 4.2. P&T potential benefits

In this context, P&T still offers some significant extra advantage towards the solution for managing existing wastes. However, the incentive to implement any significant P&T technology can only be seen in the context of future innovative fuel cycles in view of the demonstrations still to be made and the potential additional cost (separations, remote fuel fabrication, fuel handling, and potential impact on reactor availability, etc.). In fact, studies have shown that advanced fuel cycles with P&T offer significant potential benefits to deep geological storage:

- Reduction of the potential source of radiotoxicity in a deep geological storage (of relevance, e.g., in the so-called “intrusion” scenarios);
- Reduction of the heat load: larger amounts of waste can be stored in the same repository (depending on the host formations).

As regards the first point (i.e., radiotoxicity reduction), while P&T will not replace the need for appropriate geological disposal of high-level waste, several studies have confirmed that different transmutation strategies could significantly reduce – i.e., by a hundred-fold – the long-term radiotoxicity [3].

With P&T, a reduction factor larger than 100 on the mass of these transuranium elements can be achieved, and the same reduction factor can be achieved on radiotoxicity. The large reduction on the inventories provides significant reduction of the consequences of low-probability accidents, like human intrusion, and drastically reduces the potential proliferation interest of the repository. The inventory reduction also implies that radiotoxicity reaches the level corresponding to the uranium

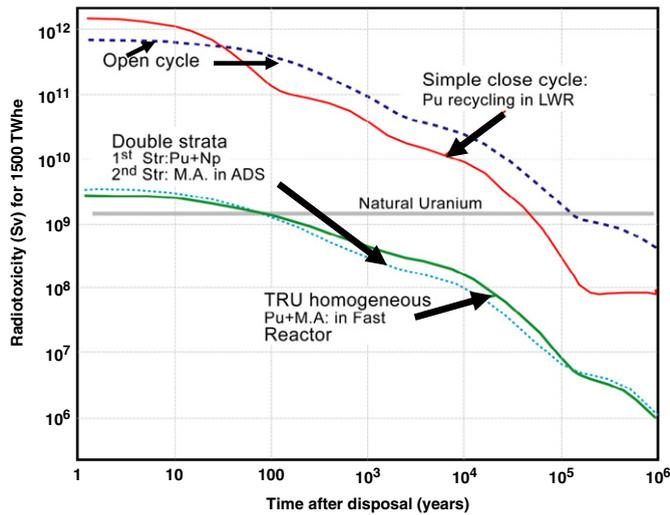


Fig. 10. Radiotoxicity reductions according to different strategies.

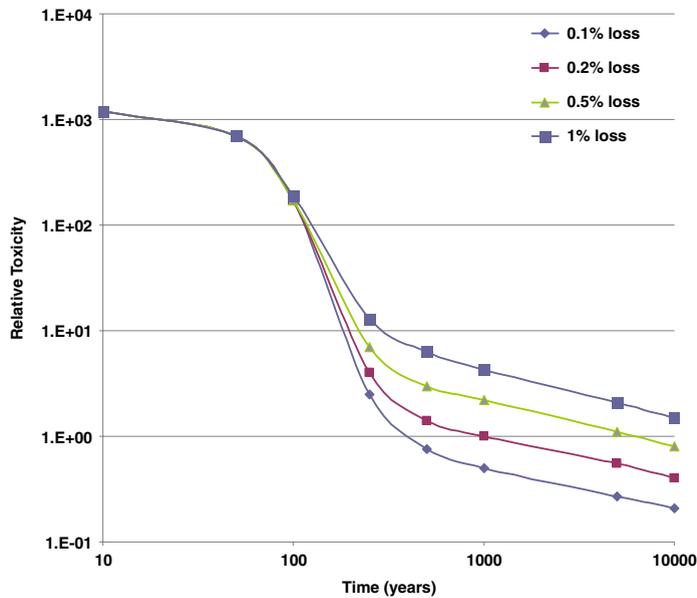


Fig. 11. Reduction of radiotoxicity for different % of losses at reprocessing.

ore mined for the fabrication of the fuel in less than 1000 years, whereas the spent fuel in the open cycle will take several times that (100,000 years) to reach the same level (see Fig. 10).

Radiotoxicity reduction is comparable (i.e., higher than a factor 100) in fuel cycle scenarios (a) and (b), as discussed previously in Section 2, and depends on the level of losses during reprocessing. However, the goal to reduce it to the level of the initial ore after ~2–300 years cannot be reached if the losses at TRU reprocessing are higher than 0.2% (see Fig. 11).

As regards the second point (i.e., heat load reduction), in general the high-level radioactive waste arising from the advanced fuel cycle scenarios associated with P&T generate less heat than the LWR spent fuel. This is important, because, in the case of disposal in hard rock, clay, and tuff formations, the maximum allowable disposal density is determined by thermal limitations.

Several impact studies have been performed in the last few years that have underlined the role of heat load and its potential reduction.

This reduction shows the possibility, for scenarios with full Pu and MA recycling, of large gains in the reduction of the thermal load to the repository and on its associated capacity by delaying the disposal time 100–200 years more. Similar reduction on the HLW thermal power can be gained at shorter times by separating the Sr and Cs from the HLW [34].

Studies performed in Japan [40,41] have confirmed the possible benefit of P&T of MA and FP in the context of the waste management by varying several conditions of fuel cycle parametrically, i.e., the reactor type, the cooling time before reprocessing, and the scheme of reprocessing: PUREX, MA-recycling, FP partitioning, and full P&T for both MA and FP.

It was found that MA transmutation in FRs and FP partitioning reduced the repository area by a factor  $\sim 4$ –5, and that MA transmutation plus FP partitioning and long-term storage of Cs and Sr reduces the repository area by more than a factor of 100.

A similar study has been done within the framework of a European project [42]. An activity is currently under way by a task force at the OECD-Nuclear Energy Agency in order to gather and analyze results of different studies that assessed the potential impact of P&T on different types of repositories in different licensing and regulatory environments. This means that criteria, metrics, and impact measures will also be analysed and compared in order to give as far as possible an objective state of the art that can help to shape decisions on different options of future advanced fuel cycles.

As indicated above, P&T has referred to the separation and recovery of actinide elements for recycle, although the potential for treating some of the FPs, such as  $^{99}\text{Tc}$  and  $^{129}\text{I}$ , has been examined. The utility of treating actinides may depend on the specific repository environment being considered, since, for many of the options, the actinides do not appear to be very important for the normal evolution of the repository.

When the abnormal events are considered, the situation may be different. Actinides appear to dominate the risk since the radiotoxicity of the materials is more of a determining factor, depending on the details of the scenario. In this case, P&T of the actinides would significantly impact the associated risk as measured by changes in the radiotoxicity, and the candidate isotopes for P&T could be identified. Finally, it should be kept in mind that a key output from any repository safety assessment is the identification of uncertainties that can affect the safety of the repository.

What matters for overall repository performance is the inventory of hazardous radioactive materials. Then, one significant effect of P&T is that the inventory of the emplaced materials is much lower on an energy-generated basis for the actinide elements, which can make the uncertainty about repository performance less important. Moreover, P&T may reduce the importance of uncertainties about the disturbed condition scenarios, since these scenarios seem to be affected by the hazard (radiotoxicity) and not so much by the geology. P&T of the actinides does reduce the hazard of the emplaced materials.

In any case, arguments for P&T inevitably require operation of reprocessing plants. The realization of P&T in combination with future Gen-IV reactors is advantageous with regard to an efficient use of nuclear fuel and related to the minimization of produced waste per generated electricity. At the same time, P&T provides the advantage of minimizing the lifetime of radiotoxic inventory in the waste. Realization of such concepts represents a huge step forward in the development of nuclear energy technology. Moreover, the public perception of nuclear energy can be definitely improved by recognizing the existence and viability of a sound technical option able to substantially reduce the amount of radioactive wastes.

## 5. P&T implementation in an EU perspective

As stated in the Strategic Research Agenda (SRA) of the SNETP [43], to increase the sustainability of nuclear energy, more efforts should be dedicated to the development of advanced fuel cycles. Consistent with that goal, a roadmap towards implementation has been proposed and integrated in the SRA.

To implement P&T at the 2040–2050 horizon, it is expected that by 2012 the following milestones will be achieved.

- Review of national positions, review of the potential of P&T to reduce the burden on the geological repository in terms of radiotoxicity, residual heat, and capacity.
- Review of ADS versus critical fast systems and different coolant technologies.
- Selection of technologies (chemical reprocessing and innovative fuels) for a closed fuel cycle based on technical and economic criteria.
- Decision on demonstration facilities to be built between  $\sim 2012$  and 2020.

As regards R&D, between 2009 and 2012, a common trunk (i.e., independently from the choices indicated above) of R&D is under way on the different technologies discussed in Sections 2 and 3 to support the reviews and the decisions to be made in 2012, as indicated above. In this respect, several projects have been implemented in the FP7 of the European Union (e.g., ACSEPT: Actinide Recycling by Separation and Transmutation; GETMAT: Gen-IV and Transmutation Materials; F-BRIDGE: Basic Research for Innovative Fuel Design for Gen-IV Systems; and FAIRFUELS: Fabrication and Irradiation and Reprocessing of Fuels and Targets for Transmutation).

A number of demonstration facilities are expected to be built between 2012 and 2025:

### A. A sodium FR (SFR) prototype

- The ASTRID (advanced sodium test reactor for industrial demonstration) has been proposed in France, and a decision on major design options is expected by 2012.

### B. Experimental reactor for the demonstration of the following:

- Alternative coolant technology with respect to Na (e.g., the ALLEGRO reactor, proposed by CEA, in support of the gas-cooled fast reactor, GFR).

- ADS concept, operation, and reactivity control and possibly to provide a fast spectrum irradiation facility (the MYRRHA facility developed at SCK-CEN).
- C. *Advanced reprocessing facility*
- In order to have available the MA necessary for a first demonstration irradiation, (e.g.,  $\sim 1$  subassembly to be loaded in a prototype after 2020), it is necessary to have an advanced processing pilot unit able to process the equivalent of 1 ton at a time horizon of 2020. In fact, today, the ATALANTE facility in Marcoule, France, allows applying advanced aqueous partitioning processes to irradiated fuel according to a 15 kg/batch, one batch/year.
- D. *Advanced fuel fabrication facility*
- Today the MALAB at the Institute for Transuranics ITU has limits—e.g., in the maximum amount of Am that can be handled in one year ( $\sim 500$  grams). Moreover, no curium-bearing fabrication has been made, with the exception of the METAPHIX experimental pins mentioned in Section 3.
  - In order to have the fuels for one or more subassemblies to be put in a prototype (beyond 2020) it is necessary to have an advanced fuel fabrication pilot unit with a capacity between 100 and 200 kg/year, at a time horizon of  $\sim 2025$ –2030. As regards fuel fabrication, it will also be necessary (by  $\sim 2018$ ) to have an FR fuel fabrication workshop first for the fuel (probably mixed oxide) to be initially loaded in a prototype FR ( $\sim 10$ –20 tons of MOX fuel for a prototype of several hundred of MWe) and then for the experimental reactor.

## 6. Conclusions

Whatever the future of nuclear power, it is universally recognized that a safe and acceptable final solution must be pursued for existing and projected inventories of high-activity, long-lived radioactive waste. Transmutation of part of the waste through use of advanced fuel cycles, although perhaps feasible in the coming decades, would not eliminate the need for managing the currently existing waste and residual quantities of high-activity, long-lived radioactive waste from future fuel cycles but can reduce the burden on the geological repository.

The physics principles of transmutation are well understood, and fast neutron spectrum systems appear to be the most adapted to support transmutation. However, several strategies can be envisioned, as has been summarized in this article. Major issues and challenges are found in transmutation fuel development and demonstration and in the field of separation chemistry. Significant progress has been made in the last two decades, but further crucial demonstrations are expected in the near future in order to allow decision-making in terms of practical implementation and deployment at the industrial level of P&T within advanced fuel cycles.

Many recent studies have demonstrated that the impact of P&T on geological disposal concepts is not overwhelmingly high, but that it can be significant, particularly considering the introduction of innovative fuel cycles. In fact, by reducing waste heat production, a more efficient utilization of repository space is likely. Moreover, even if radionuclide release from the waste to the environment and related calculated doses to the population are not significantly reduced by P&T, it is important to point out that a clear reduction of the actinide inventory in HLW reduces risks arising from less probable evolutions of a repository, i.e., increase of actinide mobility in certain geochemical situations and radiological impact by human intrusion. Finally, the public perception of nuclear energy can be definitely improved by the recognition of the existence and viability of a sound technical option able to substantially reduce radioactive waste.

## References

- [1] Nuclear Energy Outlook 2008, OECD/NEA, Paris, 2008.
- [2] Technical Roadmap for Generation IV Nuclear Energy Systems, US Department of Energy, Nuclear Energy Research Advisory Committee, Washington, Report GIF-002-00, 2003.
- [3] M. Salvatores, Nucl. Eng. Design 235 (2005) 805.
- [4] M. Salvatores, J. Knebel, Overview of advanced fuel cycles for the 21st century, Jahrestagung Kerntechnik 2008, Hamburg, May 27–29, 2008.
- [5] R.N. Hill, T.A. Taiwo, Transmutation impacts of generation-IV nuclear energy systems, in: Proc Int. Conf PHYSOR, 2006.
- [6] M. Salvatores, I. Slessarev, M. Uematsu, Nucl. Sci. Eng. 116 (1994) 1.
- [7] M. Salvatores, R. Hill, I. Slessarev, G. Youinou, The physics of TRU transmutation – a systematic approach to the intercomparison of systems, in: Proc. PHYSOR 2004 – The Physics of Fuel Cycles and Advanced Nuclear Systems: Global Developments, Chicago, Illinois, April 25–29, 2004.
- [8] F. Venneri, et al. Deep burn transmutation: a practical approach to the destruction of nuclear waste in the context of nuclear power sustainability, General Atomics, FDO-E00-N-TRT-X-000132, December 19, 2001.
- [9] T.K. Kim, T.A. Taiwo, W.S. Yang, R.N. Hill, F. Venneri, Assessment of deep burnup concept based on graphite moderated gas-cooled thermal reactor, in: Proc. PHYSOR 2006 – Advances in Nuclear Analysis and Simulation, Vancouver, British Columbia, Canada, September 10–14, on CD-ROM, American Nuclear Society, LaGrange Park, IL, 2006.
- [10] T. Wakabayashi, Progr. Nuclear Energy 40 (2002) 3–4.
- [11] L. Buiron, et al., Minor actinide transmutation in SFR depleted uranium radial blanket: neutronics and thermal-hydraulics evaluation, in: Proc. Int. Conf. GLOBAL 2007.
- [12] T. Taiwo, et al., US study on impacts of heterogeneous recycle in fast reactors on overall fuel cycle, in: Proc. Int. Conf. on Fast Reactors, FR09, Kyoto, Japan, December 2009.
- [13] M.A. Smith, et al., Physics and safety studies of low conversion ratio sodium cooled fast reactors, in: PHYSOR 2004, Chicago, USA.
- [14] Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Fuel Cycles. A comparative study, OECD-NEA Report 2002.
- [15] G. Palmiotti, M. Salvatores, M. Assawaroongruengchot, Innovative fast reactors: impact of fuel composition on reactivity coefficients, in: Proc. Int. Conf. on Fast Reactors, FR09, Kyoto, Japan, December 2009.
- [16] M. Salvatores, J. Phys. IV, France 9 (1999) 17–33.
- [17] H.A. Bethe, Nucl. News 21 (1978) 41.
- [18] W.M. Stacey, Fusion Eng. Design 82 (2007) 11–20.

- [19] A Roadmap for Developing Accelerator Transmutation of Wastes (ATW) Technology, Report to Congress DOE/RW-0519, October 1999.
- [20] T. Mukaiyama, et al., Importance of the double strata fuel cycle for minor actinide transmutation, in: Proc. 3rd OECD NEA Int. Inf. Exchange Mtg. on Partitioning and Transmutation, Aix-en-Provence, December 1994.
- [21] A European Roadmap for Developing Accelerator Driven Systems (ADS) for Nuclear Waste Incineration ETWG, April 2001.
- [22] J.U. Knebel, H. Ait Abderrahim, L. Cinotti, F. Delage, C. Fazio, M. Giot, B. Giraud, E. Gonzalez, G. Granget, S. Monti, A.C. Mueller, EUROTRANS, European research programme for the transmutation of high-level nuclear waste in an accelerator driven system, in: Proc. "FISA 2006 EU Research and Training in Reactor Systems", Luxemburg, 2006, pp. 353–372.
- [23] C. Fazio, et al., Nuclear Eng. Design 238 (6) (2008) 1471–1495.
- [24] M. Salvatores, et al., MUSE-1: A first experiment at MASURCA to validate the physics of subcritical multiplying systems, 2nd ADTT Conference Kalmar, Sweden, June 1996.
- [25] J. Rouault, M. Salvatores, The CAPRA Project: Status and Perspectives, Nuclear Europe Worldscan, 1995.
- [26] E.A. Hoffman, W.S. Yang, R.N. Hill, Trans. Am. Nucl. Soc. 96 (2007).
- [27] C. Fazio, M. Salvatores, W.S. Yang, Down-selection of partitioning routes and of transmutation fuels for P&T strategies implementation, in: Proc. Int. Conf. GLOBAL 2007, Boise, Idaho, September 2007.
- [28] M. Salvatores, I. Slessarev, A. Tchistiakov, Nucl. Sci. Eng. 130 (1998) 309–319.
- [29] M. Delpech, et al., Scenarios analysis of transition from Gen II/III to Gen IV systems, Case of the French fleet, GLOBAL 2005, Tsukuba, Japan, 2005.
- [30] T.A. Taiwo, et al., Nucl. Technol. 155 (1) (2006) 34–54.
- [31] M. Salvatores, Nuclear data for advanced fuel cycles, in: Proc. Int. Conf. IEMPT-10, Mito, Japan, October 2008.
- [32] Partitioning and Transmutation European Roadmap for Sustainable Nuclear Energy (PATEROS), FI6W-Contract Number 036418, Syntheses Report", 2008.
- [33] J.M. Adnet, et al., Development of new hydrometallurgical processes for actinide recovery: GANEX concept, in: Proc. Int. Conf. GLOBAL '05, Tsukuba, October 9–13, 2005.
- [34] R.A. Wigeland, et al., Nucl. Technol. 154 (2006) 95.
- [35] J.F. Babelot, N. Chauvin, Joint CEA/ITU synthesis report of the experiment SUPERFACT 1, Report No. JRCITU-TN-99/03, September 1999.
- [36] L. Breton, et al., METAPHIX-1 non destructive post-irradiation examinations in the irradiated element cells at PHENIX, in: Proc Int. Conf. Global 2007, 9–13 Sept. Boise USA. See also H. Otha et al. "Irradiation Experiment on Fast Reactor Metal Fuels Containing Minor Actinides up to 7% At. Burn-up", ibidem.
- [37] J.M. Bonnerot, et al., Progress on inert matrix fuels for minor actinide transmutation in fast reactor, in: Proceedings Global 2007, Boise, Idaho, USA, 2007.
- [38] D. Struwe, J. Somers, Overview of activities in Europe exploring options for transmutation, in: Proc. EURADWASTE, Luxemburg, October 2008.
- [39] F. Delage, et al., 1st Int. Workshop on the Technology and Components of ADS, Karlsruhe 2010.
- [40] H. Oigawa, et al., Partitioning and transmutation technology in japan and its benefit on high-level waste management, in: Proc. Int. Conf. GLOBAL'07, Boise, Idaho, September 2007.
- [41] K. Nishihara, et al., J. Nucl. Sci. Technol. 45 (1) (2008) 84–97.
- [42] W. Von Lensa, et al., (Eds) REDIMPACT: Impact of Partitioning, Transmutation and Waste Reduction Technologies on the Final Nuclear Waste Disposal, Schriften des Forschungszentrums Jülich, Volume 15, 2007.
- [43] Sustainable Nuclear Energy Technology Platform (SNETP), Strategic Research Agenda (SRA), Edition 2009.