

Curium Management Studies in France, Japan and USA

A Report by the WPFC Expert
Group on Chemical Partitioning
of the NEA Nuclear Science
Committee

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Foreword

Under the auspices of the NEA Nuclear Science Committee (NSC), the Working Party on Scientific Issues of the Fuel Cycle (WPFC) has been established to co-ordinate scientific activities regarding various existing and advanced nuclear fuel cycles, including advanced reactor systems, associated chemistry and flow sheets, development and performance of fuel and materials, and accelerators and spallation targets. The WPFC has different Expert Groups to cover a wide range of scientific fields in the nuclear fuel cycle.

The Expert Group on Chemical Partitioning was created in 2001 to (1) perform a thorough technical assessment of separations processes in application to a broad set of partitioning and transmutation (P&T) operating scenarios and (2) identify important research, development and demonstration necessary to bring preferred technologies to a deployable stage and (3) recommend collaborative international efforts to further technological development.

This report summarises recent studies on curium separation and management in France, Japan and USA.

Acknowledgement

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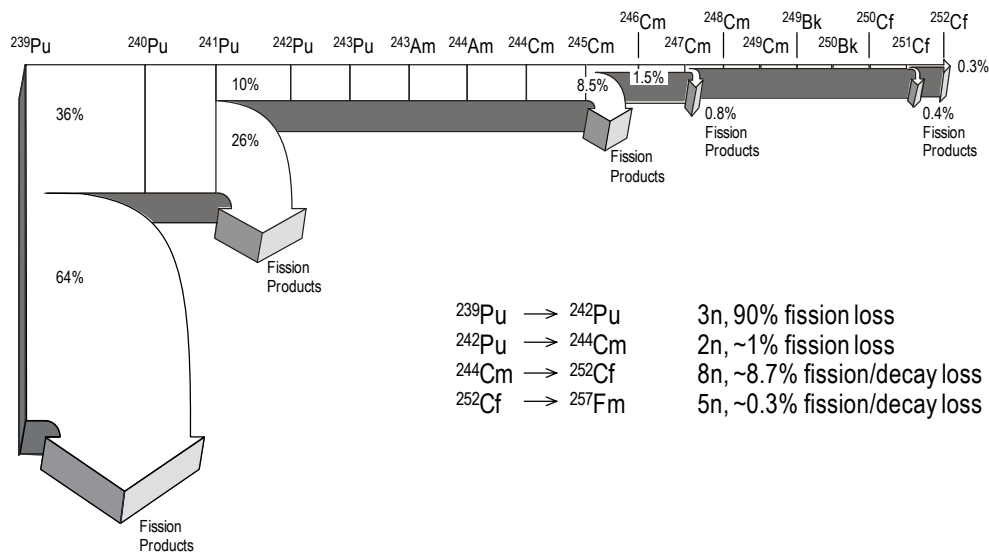
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Executive Summary

Curium is produced in thermal-spectrum irradiation of nuclear fuel in Light Water Reactors (LWR) used in many countries for the generation of electricity. The production path begins with ^{239}Pu , produced by neutron capture of ^{238}U , and then, continues through a series of transmutation neutron capture and beta decay reactions, as illustrated in the diagram below:



Transplutonium-element yield and fission loss during thermal neutron irradiation of plutonium

As indicated, the amount of curium produced is relatively small because many of the precursor isotopes undergo fission reactions such that only about 10% of the ^{239}Pu is converted into curium. Moreover, the odd isotopes of curium (^{245}Cm and ^{247}Cm) that are produced are largely destroyed by fission. The net production of curium in current LWR used fuels with burn-up ranging from 45-55 GWd/MT, is <0.01%. However, recycled used fuels can contain substantially increased (>10 fold) amounts of curium.

The curium isotopes, ^{244}Cm and ^{245}Cm , are the dominant mass of the curium produced, with typical isotopic portions of 85% and 15%, respectively. Even though the mass of curium produced in used nuclear fuels is small, the ^{244}Cm is the major source of spontaneous fission neutrons which requires special shielding and handling provisions. In addition, ^{244}Cm (half-life = 18.1 years) is a significant contributor to decay heat emission for several decades after irradiation. Furthermore, after decay for longer periods, the ^{245}Cm (half-life = 8265 years) becomes the predominant isotopic mass, leading to potential safeguards concerns.

Curium is closely associated with americium in irradiated fuels because of their chemical similarity with regard to potential separation requirements, and because americium also requires special shielding and handling requirements due to its gamma radiation emission. Americium is produced in greater mass than curium in irradiated nuclear fuels and the mass ratio can grow exponentially with decay time because of the simultaneous decay of ^{244}Cm and in-growth of ^{241}Am from decay of ^{241}Pu (half life = 14.4 years).

For these reasons, curium management is challenging. Countries that are now engaged in or planning future fuel recycle operations, are considering methods to manage the curium produced and minimise the shielding and handling requirements, as well as the reprocessing requirements for separation of curium from americium. France, Japan, and the USA have begun curium management studies. Curium management methods under consideration include (1) separation of curium from americium and storage of curium for several decades to allow ^{244}Cm to decay substantially to ^{240}Pu , while moving ahead to recycle americium; (2) recycling of americium and curium without separation; and (3) waiting several decades to reprocess used nuclear fuels, allowing decay minimisation of curium emissions and the requirement for separation of curium from americium, and allowing an alteration of the subsequent transmutation path to reduce the production of curium in recycled used fuels.

Curium management in France, Japan and the USA

In this report, recent curium management studies in France, Japan, and the USA have been described. The French studies included scenarios that compared the recycle of all minor actinides (neptunium, americium, and curium) with the recycle of only neptunium and americium in radial blankets of sodium-cooled fast reactors (SFR). In the latter scenario, curium is separated from americium during used fuel reprocessing and stored for 5000 years to allow ^{244}Cm to decay to ^{240}Pu which is then recycled. Even though the studies showed that the recycle of all MAs provides the most efficient decreases of radiotoxic inventory and decay heat in the high level waste, the removal and decay storage of curium allows overall reductions while minimising the neutron shielding and handling requirements during fuel recycle irradiations. Thus, French separations studies have been directed toward americium separation from curium and fission products in a single process. The development of the ExAm process can accomplish the needed recovery of americium.

The Japanese studies compare Minor Actinide (MA) isotope production in (1) low-enriched uranium LWR used fuel, (2) in U-Pu mixed oxide (MOX) used fuel, (3) in Fast Breeder Reactor (FBR) MOX used fuel, and (4) in FBR U-Transuranium (TRU) MOX used fuel. The studies show the effects of increasing decay time following irradiation on decay heat generation from both fission products and actinide isotopes. The decay time effects of decreasing ^{244}Cm and fission product inventory with a simultaneous increase in the ^{241}Am inventory are shown. Also, the Japanese studies describe curium separation process development by means of extraction chromatography methods.

The USA studies describe the radioactive decay effects and indications on mass, heat, and radio toxicity amounts and ratios of curium and americium. Also shown is the decreased curium production during transmutation of actinides after several decades of decay storage, prior to used fuel reprocessing. Curium inventory is compared for recycle scenarios using LWRs or fast reactors with either 10-year or 35-year fuel cycles.

Chapter 1: France

1.1 Introduction

The French CEA (Commissariat à l'énergie atomique et aux énergies alternatives) is currently studying different solutions to manage plutonium and minor actinides within the framework of the French law on waste management. Therefore, scenario and process development studies are in progress in France to assess different options of the separation and transmutation of plutonium and minor actinides in the French fleet of reactors.

The option chosen for the generation IV reactors in France is the Sodium cooled Fast Reactor (SFR). Scenarios studies evaluate the consequences of waste management on the SFR deployment from the plutonium inventory point of view, the possibility to transmute minor actinides, the capacity of cycle facilities and the waste production.

The transmutation of minor actinides can be obtained in two different modes: minor actinides can be directly mixed with standard fuel (homogeneous mode) or separated from the fuel and added in radial blankets (heterogeneous mode). Only the heterogeneous mode will be considered here.

Among the minor actinides, curium represents a special challenge as it contains radioisotopes that are high neutron emitters and have a high thermal capacity. These issues impose many constraints on fuel cycle facilities and on the implementation of treatment and recycling operations. On the other hand, americium is the main contributor to the long term radiotoxicity and heat generation of glasses used today for High Level Waste (HLW) storage. Until now, the question of its recycling still remains open and the accompanying advantages and drawbacks must be carefully weighed.

1.2 Scenario studies

Scenario studies are carried out with the simulation software COSI and the objective is to assess the impact of curium recycling on HLW radiotoxicity and decay heat.

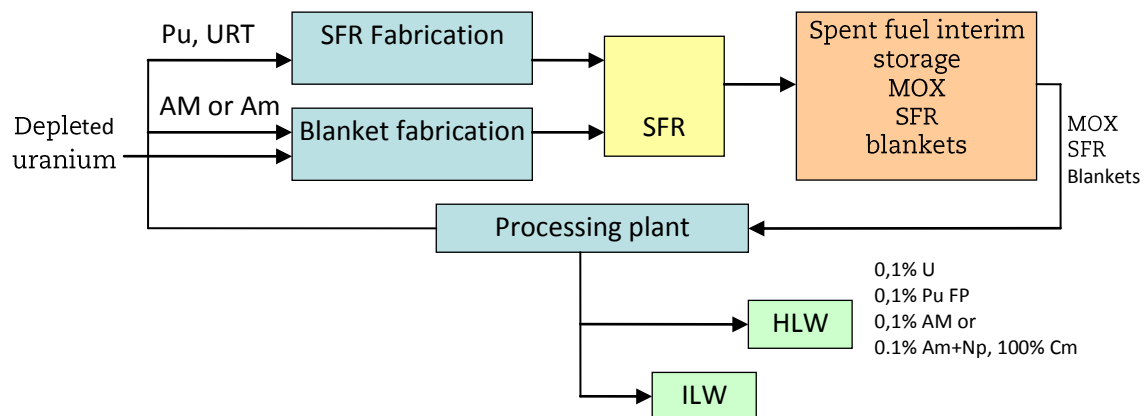
To reach this objective, two scenario studies are considered as shown in figure 1.1:

- Scenario 1 (S1) considers a fleet of SFR recycling plutonium in the fissile part of the core and minor actinides in the radial blankets. Two cases are considered: an ambitious one with 40% of MA in the blankets and a more realistic case with 10% of MA.
- Scenario 2 (S2) considers a fleet of SFR recycling plutonium in the fissile part of the core and 40% of americium + neptunium in radial blankets (curium is sent to the waste).

1.2.1 Scenario Assumptions

The scenario assumptions consider a fleet of Sodium-cooled Fast Reactors (SFR) recycling plutonium and its associated nuclear facilities: reprocessing plant; fuel fabrication plant; blanket fabrication plant; interim storage for spent fuel; and HLW and ILW storage.

Figure 1.1. Scenarios S1 and S2: Scheme of the fleet after 2100



The SFR core concept employed was developed by the CEA and French partners in 2008 and is known as SFR Version V2B [1]. The main neutronic data of this SFR are listed in table 1.1. Basically, this SFR core has a breeding gain close to zero, even without any radial and axial blankets. This SFR concept is studied in scenario 1 and has an iso-generation objective.

Radial blankets are added to the core to transmute minor actinides on a depleted uranium support. Consequently, the breeding gain of SFR becomes positive thanks to plutonium production in radial blankets.

Table 1.1. SFR: Assembly and core characteristics

Assembly and core characteristics	SFR V2B
Thermal power	3600 MWth
Net electrical power	1450 MWe
Core management	5 × 410 EFPD
Number of fissile assemblies	453
Heavy nuclide volume fraction (%vol) ¹	43.7
Na volume fraction (%vol)	27.6
Average core power density (W/cm ³) ²	207
Reloading frequency	5
Average burnup (GWd/tons)	~100
Maximum burn-up (GWd/tons)	139
Average Pu content (%vol fuel)	15.8
Equivalent ²³⁹ Pu weight content (%)	11.0
Initial Pu mass (tons) ³	12.0
Total heavy nuclides mass (tons)	74.0
Average instantaneous breeding gain BOC/EOC	+0.01
Void effect (\$) EOC	4.9

The fuel cycle assumptions are listed in Table 1.2.

¹ Fuel porosity : 0,96

² Volume is calculated at T = 20°C

³ Pu vector : ²³⁸Pu=3.57 / ²³⁹Pu=47.38 / ²⁴⁰Pu=29.66 / ²⁴¹Pu=8.23 / ²⁴²Pu=10.38 / ²⁴¹Am=0.78

Table 1.2. Fuel cycle assumptions

Fabrication	
Fuel fabrication time	2 years
Reprocessing	
Cooling time before reprocessing	SFR blankets: 5 years SFR fuel: 5 years,
Reprocessing losses	Scenario 1 : 0.1% U, Pu, Am, Np, Cm Scenario 2: 0.1% U, Pu, Am, 100% Np, Cm
Priorities for reprocessing	Chronologic order

1.2.2 Impact of Curium recycling on radiotoxicity and thermal power of the HLW

The complete calculation of High Level Waste isotopic composition allows to determine the evolution of waste during long time storage, up to ten million years. Radiotoxicity and thermal power of waste are calculated during long time storage. The contributions of both heavy nuclei and fission products are taken into account.

In scenario 1, Pu and all the minor actinides are recycled. Thus, the fission products, and U, Pu, MA losses are sent in the HLW.

In scenario 2, Pu, Am and Np are recycled. Thus, the fission products, and U, Pu, Am, Np losses and all Cm are sent in the HLW. Compared to scenario 1, the recycling of Am in the SFR also induces an increase in Cm production.

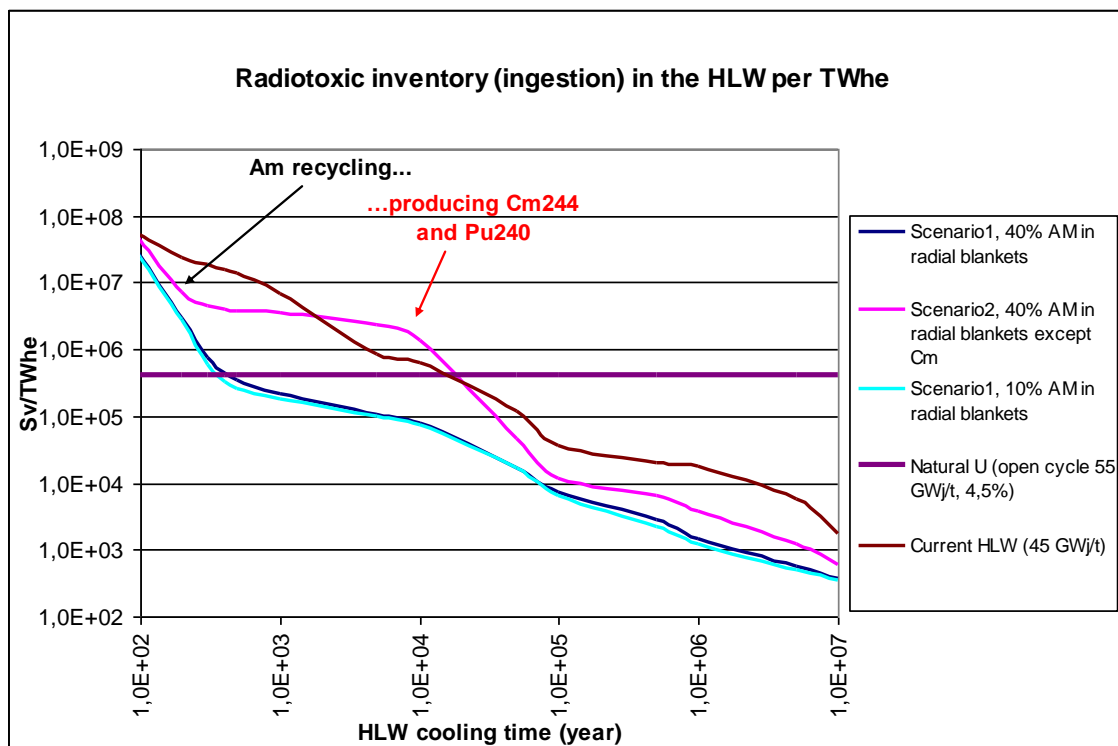
The results are compared with the current HLW produced in France, coming from UOX reprocessing and containing the fission products, all the minor actinides, U and Pu losses.

The radiotoxic inventory of the HLW is presented in Figure 1.2. In the case of Am+Np recycling (Cm in the HLW), the radiotoxic inventory of the HLW:

- Is lower than current HLW until 5000 years thanks to Am recycling (^{244}Cm decay, $T = 18$ years)
- Is higher than current HLW from 5000 to 50 000 years : impact of ^{240}Pu produced by ^{244}Cm decay
- After 50 000 years : lower than current HLW (decay of Am)

The radiotoxic inventory of the HLW of the scenario 1 is lower than the current HLW after 100 years of cooling, i.e., after the decay of the fission products.

Figure 1.2. Radiotoxic inventory of the HLW



The decay heat of the HLW is presented in Figure 1.3. In the case of Am+Np recycling (Cm in the HLW), the decay heat of the HLW :

- is higher than current HLW until 100 years : FP and Cm dominate
- is lower between 100 and 5000 years, thanks to Am recycling (almost all ^{244}Cm has disappeared because of ^{244}Cm decay, $T = 18$ years)
- is higher between 5000 and 50000 years (^{240}Pu coming from ^{244}Cm)
- is lower than current HLW (decay of Am) because of Am decay

The decay heat of the HLW of the scenario 1 is lower than the current HLW after 100 years of cooling, i.e., after the decay of the fission products.

Figure 1.3. Decay Heat of the HLW

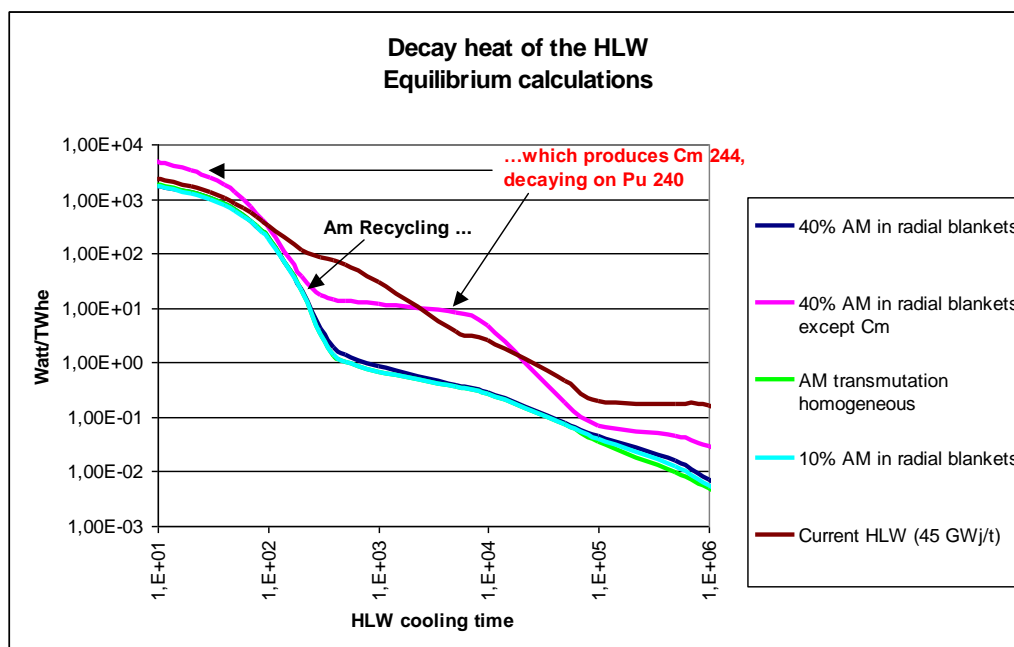


Table 1.3. Comparison of HLW decay heat in %. Reference = Current glasses

Cooling time	50	70	100	200	300
Current glasses	1	1	1	1	1
40% AM in radial blankets	-24.7	-28.6	-38.7	-81.4	-95.3
40% AM in radial blankets except CM	57.3	30.8	-3.1	-67.8	-80.4
AM transmutation homogeneous	-23.7	-27.8	-38.4	-82.3	-96.3
10% AM in radial blankets	-27.3	-31.1	-41.1	-82.7	-96.1

In the case of MA transmutation (Am + Np + Cm), a reduction of HLW decay heat can be obtained whatever the cooling time of the HLW. If Cm is not recycled, a reduction of the decay heat compared to the current glasses is obtained after more than 100 years of cooling.

Concerning the footprint of the HLW in deep geological storage, the different evaluations show that it depends not only on the heat generation rate of these wastes but also on the decay periods of the radionuclides involved. As shown in table 1.4, the fraction of thermal power due to americium decay heat has a bigger impact on the thermal footprint than the decay heat of the fission products. It is due to the intermediate decay period of americium.

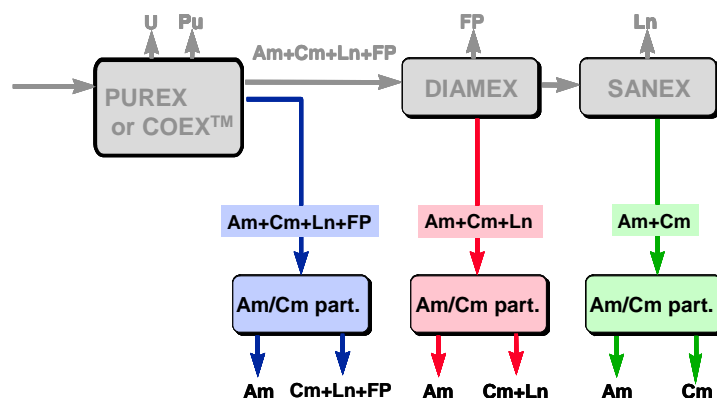
Table 1.4. Impact of Am on HLW thermal footprint

	Footprint (m/W)	P_{tot} (W)	$P_{\text{Am}}/P_{\text{tot}}$
C0	0.161	155	4.50%
C1	0.172	465	18.50%
C2	0.172	489	17.60%

Furthermore, these evaluations show then that Am+Cm recycling compared to only Am recycling has no significant reduction of the thermal footprint.

1.3 Americium separation process studies

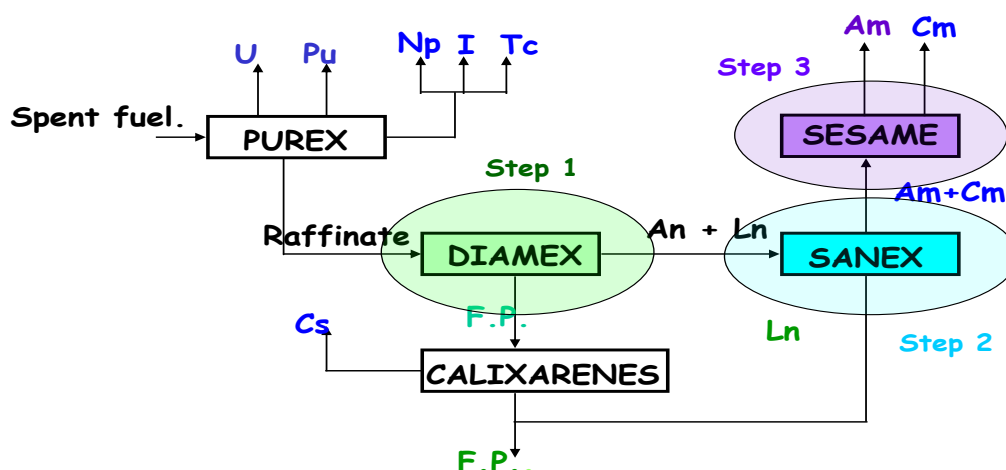
Selective americium separation becomes more difficult as this process continues (see Figure 1.4): i.e., after separation from all the fission products (for instance after the SANEX process); or after co -decontamination of minor actinides plus lanthanides from the other fission products (after the DIAMEX process); or directly from PUREX or COEX™ raffinates.

Figure 1.4. Three possible strategies for Am separation

The physico-chemical properties of Am, Cm and Ln(III) are very similar. They are strongly hydrated and their ionic radius is similar. In addition, the acidity of the medium (PUREX raffinates) is very high so there is a competition between nitric acid and cation extraction. Moreover, the element inventory is very large.

It was in the framework of the French Act 1991 on high activity waste, that a three step strategy was conceived for the first time (see Figure 1.5) [2]:

Figure 1.5. Three step strategy for minor actinide separation



Step 1: The DIAMEX process with An(III) and Ln(III) coextraction from PUREX raffinate, thanks to a malonamide extractant, such as N,N'-dimethyl-N,N'-dioctyl-hexylethoxy-malonamide (DMDOHEMA). An advantage of this step is that it reduces the acidity of the "An(III)+Ln(III)" product solution, which is introduced in the second step process. In this process, the trivalent actinides and lanthanides would be separated.

Step 2: The SANEX process for An(III) from Ln(III) separation:

Several routes have been investigated and finally CEA has chosen An(III) selective back extraction. A hydrosoluble complexing agent (polyaminocarboxylic acid such as HEDTA) efficient at low acidity is used in a buffered solution (pH adjusted between 2 and 4). The solvent is a mixture of a malonamide (DMDOHEMA) co-extracting An(III)+Ln(III) and an acidic extractant (di-alkyl phosphoric acid such di-(2-ethyl-hexyl)-phosphoric acid acronym HDEHP) which maintains the Ln(III) in the organic phase whereas the An(III) are selectively back-extracted thanks to the HEDTA aqueous mixing agent in a buffered stripping solution.

This second step was validated in December 2005 in the C11/C12 hot cell in the ATALANTE facility (see Figure 1.7), using the recovered An(III)+Ln(III) flux issuing from the first DIAMEX step implemented just before, in November 2005, in another hot cell called CBP for Process hot cell (see Figure 1.6) [3].

It should be mentioned that a single-step version of this concept was also settled and validated thanks to a counter-current hot test performed on a genuine PUREX raffinate in 2000 in the C11/C12 hot cell of the ATALANTE facility [4].

Figure 1.6. DIAMEX hot test performed in November 2005 on a genuine PUREX raffinate in the ATALANTE facility

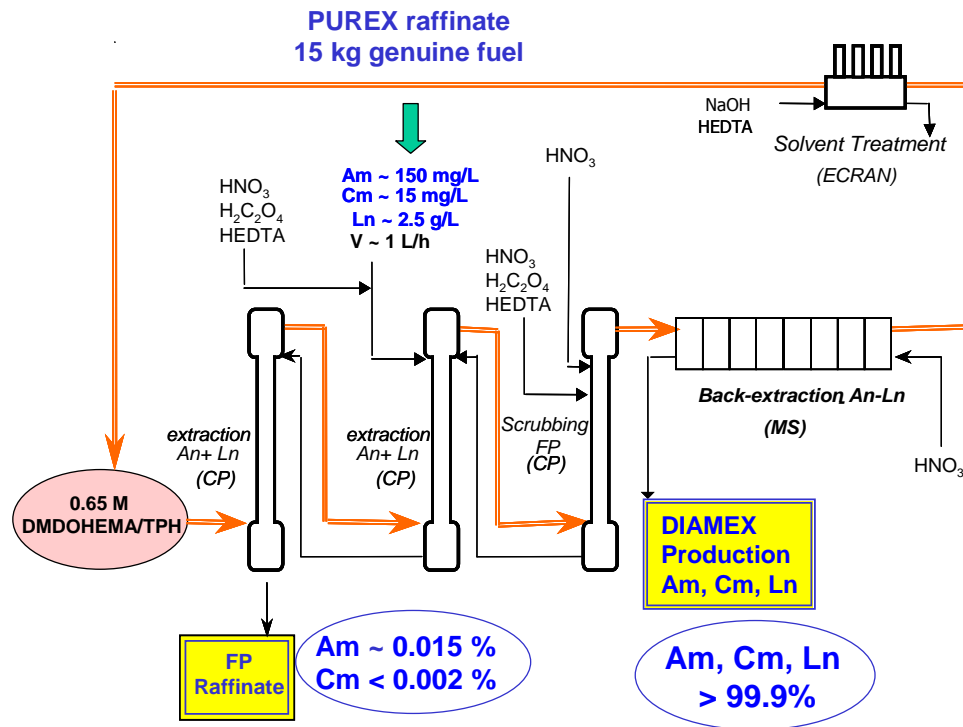
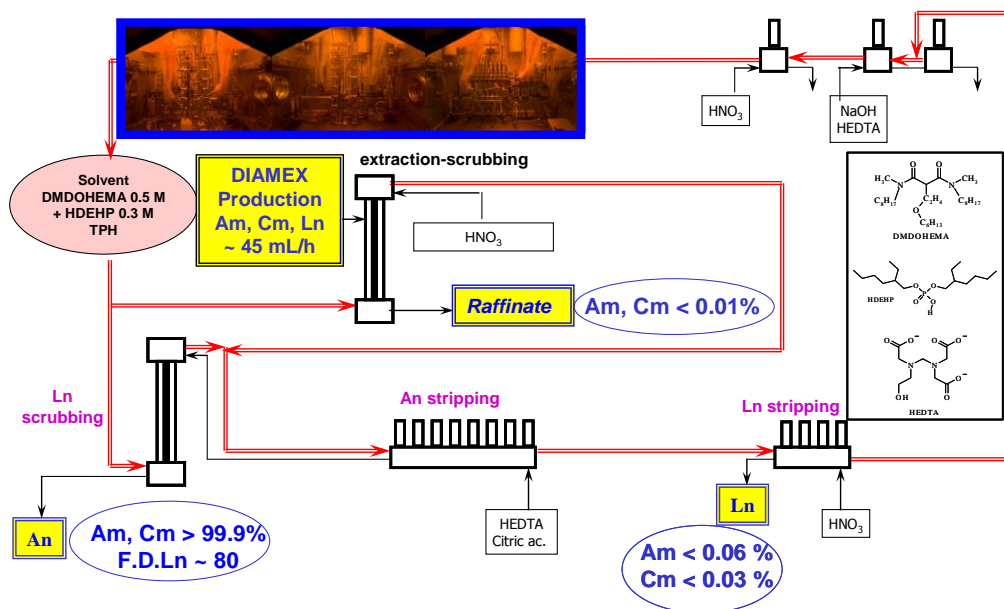


Figure 1.7. SANEX hot test performed in December 2005 on a DIAMEX stripping solution in the ATALANTE facility



Step 3: Am(III) from Cm(III) separation

Two options were investigated in France.

The SESAME process

During a first phase (1992-2001), the SESAME process (Partitioning by selective extraction of americium by electrochemical methods) was studied as a priority.

This choice is easily explained because SESAME provides additional flexibility. This process permits the reprocessing of the flows arising either from SANEX or DIAMEX, with the idea being to directly extract the americium from this effluent, with the curium not passing through the fission products. SESAME is based on the selective oxidation of americium (III), previously oxidised to oxidation state (IV) and then (VI) by electrolysis; a lacunar heteropolyanion is used to stabilise the intermediate state by complexation. The americium (VI) is then easily extracted by the TBP extractant from the PUREX process.

Between 1994 and 2001, research permitted the process to be refined, notably by:

- acquiring the basic data relating to the electrochemical oxidation phenomena of americium, as well as its extraction by solvents;
- validating, by four high-level trials, the different unitary operations of the process;
- selecting the reagents.

All of these advances allowed, at the end of 2001, the implementation of a scientific feasibility trial (SESAME 5) in shielded cells at ATALANTE. This test, performed under real conditions, implemented the following steps:

- previous oxidation of the incoming flow to produce americium (VI) with complexation of the valency (IV), by means of a continuously running electrolyzer by recirculation until it is sent to extraction;

- extraction of americium (VI) using the TBP solvent;
- reoxidation of the fraction of americium remaining in the aqueous solution after three extraction steps. This is performed by an online electrolysis step complemented by additional americium (VI) extraction.

The results of the SESAME 5 experiment are not consistent with the performance predictions calculated with the aid of a model developed from the data acquired during the previous trials. Thus, for americium, the following statistics were measured:

- a 78% recovery rate of the initial americium, with a purity of 99.5%;
- a loss of 13% of americium in the catholyte during electrolysis of the incoming flow; this fraction would still be potentially recyclable;
- a leakage of 7 to 8% of the americium in the extraction phase due to limited oxidation into Am (VI) in the incoming flow, which is no doubt attributable to the presence of impurities.

For curium, the recovery rate is 88% in the solution arising from the second extraction and the loss of 14% in the catholyte of the first oxidation. Thus, in 2001, the SESAME process could not demonstrate its efficiency for reprocessing true high-level effluents.

Alternative process based on DIAMEX

The inadequate performance shown by the SESAME 5 experiment has led, since the end of 2001, to the development of an alternative process by liquid-liquid extraction based on the selectivity of certain extractants for americium (III) versus curium (III). Since they are selective, the extractants, which will be used later on the industrial scale, must be easy to implement and must not generate excessive quantities of secondary waste. In principle, two types of systems meet such constraints:

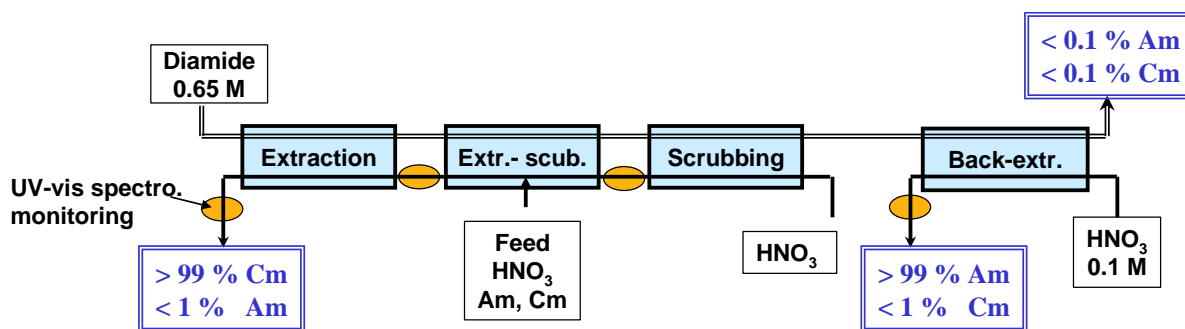
- the diamides, like DMDOHEMA already used in the DIAMEX and SANEX processes;
- the calixarenes, such as Calix[4]arene-CMPO (CMPO: acronym for Carbomoyl Methyl Phosphine Oxide). The latter, though the most selective, has not been adopted for the trial to demonstrate the scientific feasibility. In fact, its implementation proved to be more intricate than expected for the recovery of the extracted americium in the aqueous phase.

The experiment procedure, performed in the ATALANTE facility at the end of 2002, was as follows:

- the solvent is that used in the DIAMEX process, i.e. DMDOHEMA at a concentration of 0.65 mol/l diluted in TPH, with the nitric acid concentration of the incoming flow being fixed at 3 mol/l;
- the extraction-washing step comprises forty-eight stages and the stripping step has eight stages. The trial had to be conducted with great rigor due to its sensitivity to variations in temperature, flow rate and acidity. An online spectrophotometric monitoring of the americium at five strategic points in the experiment permitted the control of the trial and the attainment of the target performance;
- 99.9% of the two actinides were recovered;
- the rate of americium left in the raffinate with curium was only 0.6 % and so the recovery rate of purified americium reached 99.4% with only 0.7 % of the curium from the feed.

The above experiment procedure demonstrated the scientific feasibility of this alternative method based on liquid-liquid extraction (see figure 1.8) [5]. Due to its similarities to the DIAMEX process, this process was called DIAMEX 2.

Figure 1.8. Americium-curium partitioning hot test performed in December 2002 on surrogate solution in ATALANTE facility



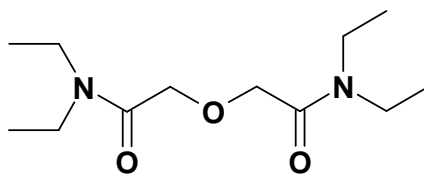
1.3.1 Innovative single step process: ExAm

More recently, in the frame of the 2006 act, the objective was to develop a single cycle process to recover the sole Am(III) directly from PUREX or COEX™ raffinates. The corresponding process was called ExAm (Extraction of Americium).

The ExAm process has been developed on the basis of the DIAMEX-SANEX process, which was developed for the co-recovery and separation of Americium and Curium from PUREX/COEX™ raffinate and improved by the progress accomplished during GANEX process developments [6-7].

The principle of the ExAm process is based on the use of DMDOHEMA in the organic phase; thanks to the little preferential extraction for Am versus Cm (separation factor is equal to 1.6). So it is possible to extract americium together with some light lanthanides with close values of distribution coefficients in high nitric acidity, while curium and other lanthanides remain in the aqueous phase. The Tetra Ethyl Di Glycol Amide (acronym TEDGA, see Figure 1.9) is added in the aqueous phase, as a hydrophilic ligand, in order to increase the selectivities Am/Cm and Am/heavy Lns, because of the preferential complexation of curium and heavy lanthanides by this amide: the global efficiency of the process is largely improved, with a corresponding decrease in the number of the necessary separation stages. Then the americium is selectively stripped from light lanthanides like in the DIAMEX-SANEX/HDEHP process [8].

Figure 1.9. Structure of the N,N,N',N'-tetra-ethyl-diglycolamide

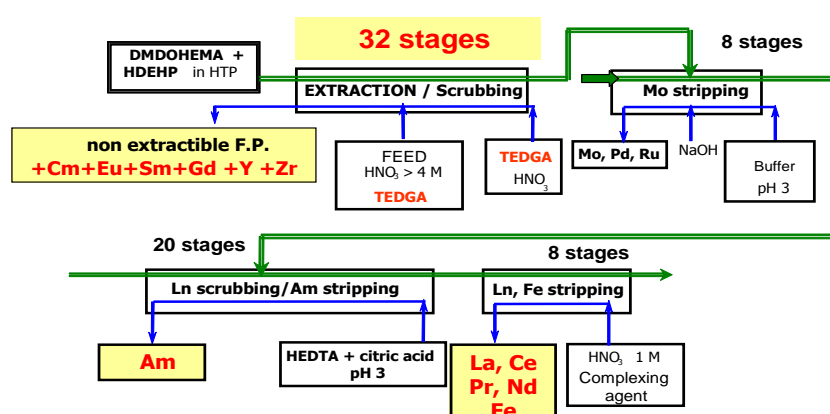


Between 2008 and 2009, numerous experimental data were acquired mainly for the key step (Am+light Lns extraction and scrubbing): distribution coefficients versus $[\text{HNO}_3]$, $[\text{TEDGA}]$, $[\text{cations}]_{\text{org}}$, identification and stoichiometry of complexes both in the organic and aqueous phases, determination of stability constants between Ln and TEDGA. The data were used to implement a phenomenological model in the PAREX process code.

A spiked counter-current test involving mainly the innovative extraction-scrubbing step was performed in December 2009 in the glove boxes of the ATALANTE facility on a surrogate feed spiked with lanthanides, ^{241}Am and ^{244}Cm , to assess the Am/Cm and Am/heavy Lns separation efficiency in comparison with the modelling. Satisfactory results were obtained: more than 97% of americium was recovered, with decontamination factor higher than 1000. The concentrations of the radioisotopes measured during the test were in good agreement with the values calculated using the PAREX code.

Finally, a complete hot test, using a genuine PUREX raffinate was carried out in April 2010, in order to confirm these promising results (see Figure 1.10). This counter-current hot test lasted about 60 hours. It consisted of 68 mixer-settler stages to insure americium only extraction-scrubbing-stripping. Good agreement between experimental and simulated performances was obtained: more than 99% of americium initially in the raffinate was extracted and the decontamination factor of americium versus curium is more than 500.

Figure 1.10. EXAm process flowsheet tested in April 2010 to recover the sole Am(III) from genuine PUREX raffinate



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Chapter 2: Japan

2.1 Evaluation of Cm production in various types of spent fuels

Cm is the main contributor of heat and neutron emission in minor actinides (MA) bearing fuel, as shown in Table 2.1. The present chapter deals with the evaluation of Cm production in various types of spent fuels (SF) [1], which is fundamental data necessary for Cm management. The spent fuels considered in the present evaluation are listed in table 2.2. They are LWR fuels of two different burn-ups (LWR-45G and LWR-60G), LWR MOX fuel (LWR-MOX), FBR fuel with U and Pu [2] (FBR-MOX) and FBR fuel with minor actinide (MA) recycle [3] (FBR-MA). The ORIGEN-2 code [4] with the cross section library of ORILIBJ33 [5] was used for the burn-up calculation.

Table 2.1. Characteristics of main isotopes of transuranium elements

Isotope	Half-life (y)	Neutron emission ($\text{g}^{-1}\text{s}^{-1}$)	Decay heat (W g^{-1})
^{237}Np	2.14×10^6	0.90	0.00002
^{238}Pu	87.7	3.6×10^4	0.56
^{239}Pu	2.41×10^4	96	0.002
^{240}Pu	6.54×10^3	1.3×10^3	0.007
^{241}Pu	14.4	1.23	0.004
^{242}Pu	3.76×10^5	2.0×10^3	0.0001
^{241}Am	432.2	7.0×10^3	0.11
^{243}Am	7.38×10^3	5.4×10^2	0.0071
^{242}Cm	0.446	2.9×10^7	120
^{243}Cm	28.5	1.3×10^5	1.7
^{244}Cm	18.11	1.2×10^7	2.8

* Spontaneous fission and (α , n) reaction

Table 2.3 shows actinide composition in each type of spent fuel (SF) cooled for 5 years after discharge [1]. Composition of MA, Np, Am and Cm, in 1 ton of SF cooled for 5 years is shown in Figure 2.1. Total amounts of MA and Cm are 1.27kg and 0.043kg (ratio of Cm to MA is 3.4%) in LWR-45G, which increase with the burn-up and are 1.88kg and 0.147kg (7.9%) in LWR-60G. These values are much larger in MOX fuel. The amount of Cm in LWR-MOX is 0.63kg, which is about 14 times larger than that in LWR-45G. FBR-MOX contains a smaller amount of Cm than LWR-MOX, but FBR-MA contains the largest amount of Cm, 1.46kg, in 1 ton of spent fuel. Total amounts of MA in FBR-MOX and FBR-MA are 7.2kg and 7.5kg respectively.

Such an increase in MA amount in spent fuel brings about the increase of heat emission from actinides. Figure 2.2 shows heat emission from actinides and fission products in 1 ton of spent fuel, and its decay. Heat emission from actinide in MOX fuel (LWR-MOX, FBR-MOX and FBR-MA) is more than that from fission products when the fuel is cooled more than 5 years.

Figure 2.3 shows heat emission from each isotope of actinides in 1 ton of spent fuel, and its decay. Main contributors of heat emission are ^{244}Cm , ^{241}Am and ^{238}Pu in all the cooling time examined and ^{242}Cm in shorter cooling time.

Table 2.2. Characteristics of main isotopes of transuranium elements

Symbol	Reactor and fuel	Burn-up	Initial composition
LWR-45G	LWR UO ₂ fuel	45 GWd/t (38MW/t×1184.2d)	4.5% U-235
LWR-60G	LWR UO ₂ fuel	60 GWd/t (38MW/t×1578.9d)	4.5% U-235
LWR-MOX	LWR MOX fuel	45 GWd/t (38MW/t×1184.2d)	6.0% Pu + NU
FBR-MOX	FBR in OECD/NEA benchmark exercise	78.8 GWd/t (72MW/t×1095d)	17.0% Pu + DU(0.3%)
FBR-MA	FBR with MA recycle Core fuel	147.1 GWd/t (44.1MW/t×3335d)	19.8% Pu + DU(0.3%) 0.92% MA
	Blanket fuel	21 GWd/t	DU

NU:natural uranium, DU:depleted uranium, MA: ^{237}Np , ^{241}Am , ^{243}Am , ^{244}Cm

Table 2.3. Actinide composition in spent fuel cooled for 5 years after discharge, as weight (g) per 1ton of initial heavy metal

	LWR-45G	LWR-60G	LWR-MOX	FBR-MOX	FBR-MA*
²³² U	2.17E-03	3.98E-03	9.36E-04	2.59E-03	1.52E-02
²³³ U	2.32E-03	2.66E-03	1.53E-03	5.92E-03	1.15E-02
²³⁴ U	1.39E+01	2.48E+01	8.86E+01	2.24E+02	1.83E+02
²³⁵ U	1.13E+04	6.18E+03	3.00E+03	1.18E+03	8.67E+02
²³⁶ U	5.70E+03	6.16E+03	7.21E+02	3.31E+02	4.39E+02
²³⁷ U	4.27E-05	5.13E-05	1.88E-04	2.66E-04	1.63E-04
²³⁸ U	9.24E+05	9.11E+05	9.06E+05	7.45E+05	7.36E+05
²³⁹ U	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
²³⁶ Np	7.96E-04	1.16E-03	5.72E-04	2.45E-03	1.18E-02
²³⁷ Np	6.59E+02	8.93E+02	2.12E+02	2.83E+02	5.70E+02
²³⁸ Np	1.54E-07	2.07E-07	3.63E-06	2.90E-05	3.24E-05
²³⁹ Np	1.19E-04	2.71E-04	1.10E-03	1.14E-03	1.23E-03
²³⁶ Pu	7.31E-04	1.26E-03	3.07E-04	8.57E-04	2.65E-03
²³⁸ Pu	2.57E+02	4.80E+02	1.62E+03	3.60E+03	2.03E+03
²³⁹ Pu	6.62E+03	6.65E+03	1.46E+04	9.62E+04	8.42E+04
²⁴⁰ Pu	2.60E+03	3.20E+03	1.28E+04	4.57E+04	4.72E+04
²⁴¹ Pu	1.38E+03	1.66E+03	6.06E+03	8.59E+03	5.28E+03
²⁴² Pu	6.57E+02	1.17E+03	5.12E+03	1.18E+04	5.54E+03
²⁴³ Pu	1.40E-13	1.28E-12	3.43E-12	9.50E-13	1.20E-10
²⁴⁴ Pu	4.54E-02	1.44E-01	5.18E-01	1.78E-01	4.53E-01
²⁴¹ Am	4.30E+02	5.21E+02	2.37E+03	5.19E+03	3.89E+03
^{242m} Am	8.19E-01	1.11E+00	1.94E+01	1.54E+02	1.73E+02
²⁴² Am	9.80E-06	1.32E-05	2.32E-04	1.85E-03	2.06E-03
²⁴³ Am	1.38E+02	3.15E+02	1.28E+03	1.32E+03	1.44E+03
^{244m} Am	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
²⁴⁴ Am	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
²⁴² Cm	1.06E-02	1.72E-02	1.42E-01	4.79E-01	4.75E-01
²⁴³ Cm	4.13E-01	9.20E-01	8.87E+00	1.69E+01	1.28E+01
²⁴⁴ Cm	3.94E+01	1.31E+02	5.39E+02	2.61E+02	1.14E+03
²⁴⁵ Cm	3.30E+00	1.37E+01	7.37E+01	2.01E+01	2.60E+02
²⁴⁶ Cm	3.07E-01	2.00E+00	5.20E+00	8.73E-01	4.30E+01
²⁴⁷ Cm	3.94E-03	3.60E-02	9.63E-02	2.67E-02	3.36E+00
²⁴⁸ Cm	2.81E-04	3.89E-03	6.40E-03	8.39E-04	3.10E-01
Total An	9.54E+05	9.38E+05	9.54E+05	9.20E+05	8.90E+05

* Composition of FBR-MA was calculated as a mixture of core fuel and axial blanket with a ratio of 69 to 31.

Figure 2.1. Weight of minor actinides, Np, Am and Cm, in 1 ton of spent fuel cooled for 5 years

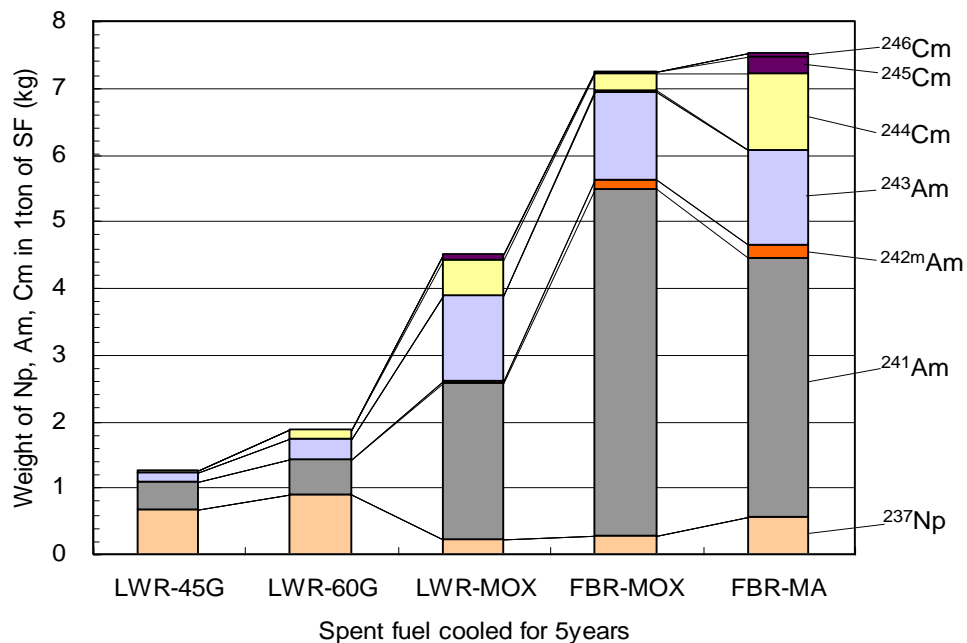


Figure 2.2. Heat emission from actinides and fission products in 1 ton of spent fuel, and its decay

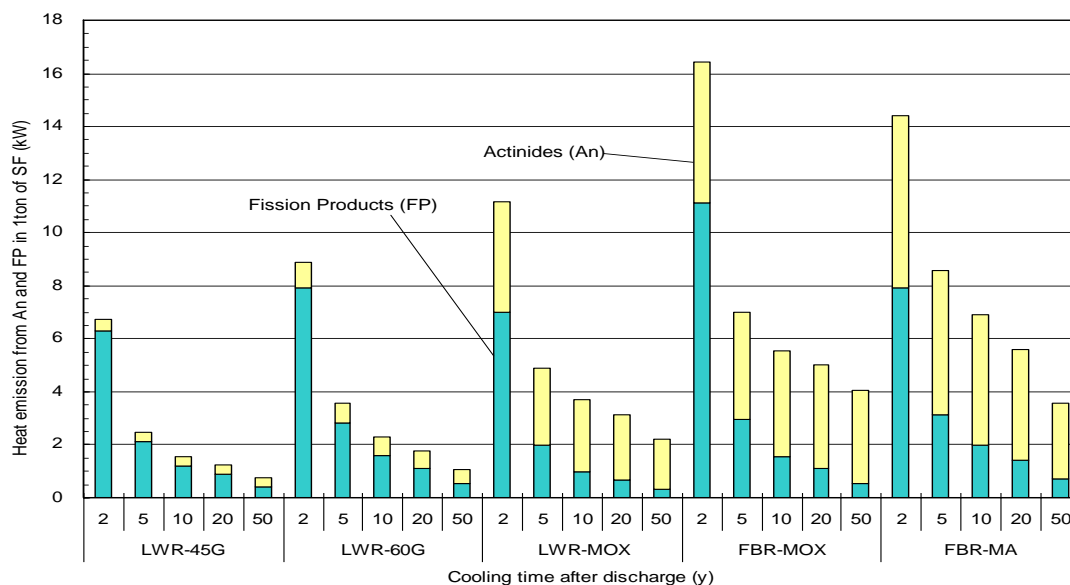
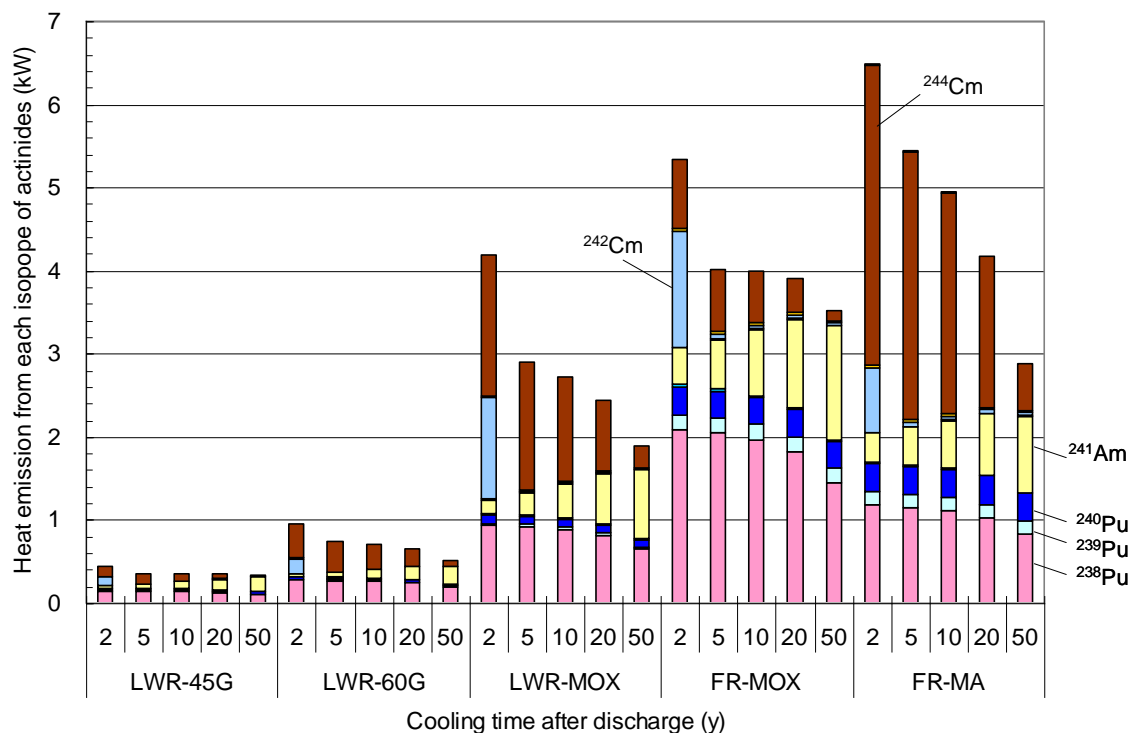


Figure 2.3. Heat emission from actinides in 1 ton of spent fuel, and its decay

2.2 Study on Cm separation

A recent study on Cm separation from Am in Japan has been carried out in the framework of the research and development of reprocessing based on ion exchange method by using tertiary pyridine resin [6],[7]. The used resin was synthesised by polymerisation of 4-vinylpyridine (80wt %) and m/p-divinylbenzene (20wt%) on the surface of high porous silica beads [8],[9]. By using the silica-supported resin, the pressure loss in the chromatography operation is considerably suppressed, and ions can easily diffuse into the resin because of the high porous structure. The resin was confirmed to have rather good stability against gamma-irradiation in nitric acid media and much better in hydrochloric acid media [10]. Tertiary pyridine resin has two functions; one is a function of weakly basic anion exchanger, and the other is a function of the soft donor ligand [7], which can be used for the separation of trivalent actinides (Am and Cm) from lanthanides [11] and the mutual separation between Am and Cm [6],[7].

The separation of Cm from Am is achieved by chromatography in nitric acid and methanol medium. Alcohol promotes the interaction between the actinide cations and pyridine, due to its dehydration effect [12], [13]. Many methods appeared in former studies using alcohol for the mutual separation of actinides, which were developed for the purpose of actinide analysis [14]. After experiments with Am and Cm tracer, the separation method was tested with a small amount of real dissolved fuel of the test fast reactor "JOYO" [6]. Feed solution to the mutual separation of Cm and Am was prepared by dissolving the dried residue with the 2:3 (in volume) mixture of 8M nitric acid and methanol. The residue was obtained by heating the Am-Cm fraction to dryness. The fraction was separated by ion exchange from other actinides and fission products in the former step. Eluent composition was the same as above. Figure 2.5 shows the results of

the chromatographic separation test [6]. Cm was almost perfectly separated from Am. The decontamination factor (DF) against Am in the Cm fraction was 8.7×10^4 and DF against Cm in the Am fraction was 2.2×10^3 . This separation method would be very attractive for the mutual separation of Am and Cm if the use of lower alcohol could be accepted in industrial plant.

Figure 2.4. Chemical structure of tertiary pyridine resin

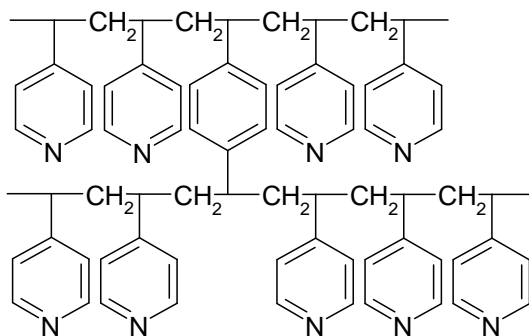
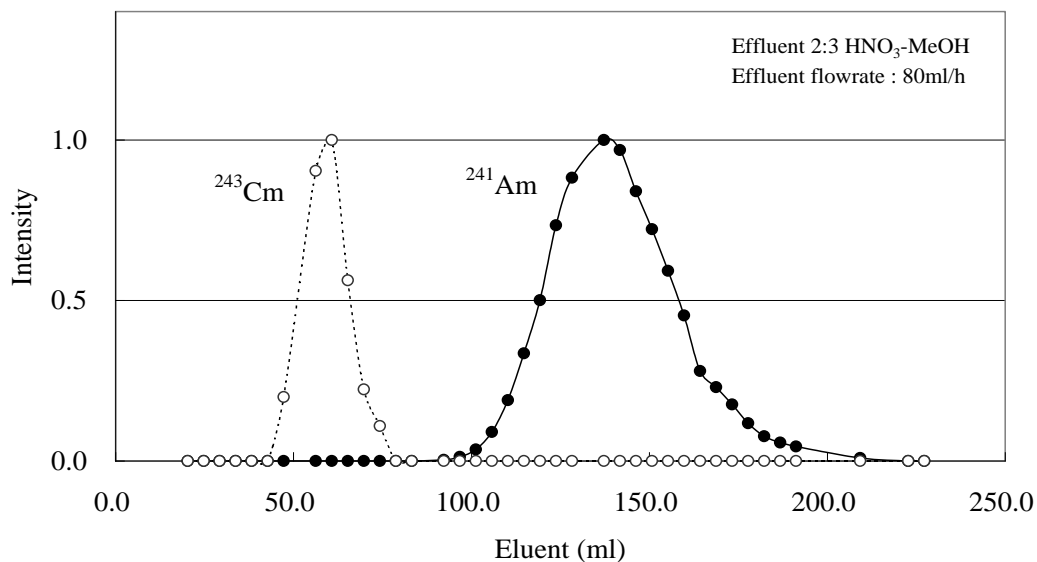


Figure 2.5. Chromatogram of Am and Cm separation by ion exchange using tertiary pyridine-type resin



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Chapter 3: USA

Neither americium nor curium is separated from fission product waste in used fuels that are currently reprocessed. However, both americium and curium have radioisotopes (^{241}Am , ^{242}Cm , and ^{244}Cm) that generate significant amounts of decay heat. Also, ^{241}Am decays to ^{237}Np (half-life = 2.1 million years), which is the major long-lived contributor to radiotoxicity in stored wastes. Therefore, advanced fuel cycles need to recycle both ^{237}Np and ^{241}Am .

The need to separate curium from americium is lessened in longer-aged fuel because the decay of ^{242}Cm (half-life = 162 days) and ^{244}Cm (half-life = 18.1 years) is significant. Moreover, the need to separate curium from americium is also lessened in aged fuels because the amount of ^{241}Am (and thus the total americium) increases with time as a result of the in-growth from decay of ^{241}Pu (half-life = 14.3 years). The effects are illustrated by the data shown in Table 3.1.

Table 3.1. Fraction of curium in americium-curium expressed as percent at 45 GWd/MT burn-up mass and decay heat-variation with decay time

	Mass (g/ton)			Decay heat (W/ton)		
	1 y	5 y	30 y	1 y	5 y	30 y
^{241}Am	135	407	1.272	15.3	46.4	145
^{243}Am	105	105	105	0.67	0.67	0.67
Total Am	240	512	1.377	16.0	47.1	146
^{242}Cm	3.84	0.11	0.0026	467	1.3	0.32
^{244}Cm	35.3	30.3	11.6	100	85.6	32.9
^{245}Cm	2.2	2.2	2.2	0.01	0.01	0.01
Total Cm	41.9	33.0	14.2	568	87.6	33.6
%Cm in Am-Cm	14.9	6.1	1.02	97.3	65.0	18.7

Previous studies [1] have shown that both curium and americium have radioactive emissions (neutrons and beta-gamma photons, respectively) that require shielded facilities for separation processes and recycle target fabrication. This finding is illustrated by the data shown in Table 3.2.

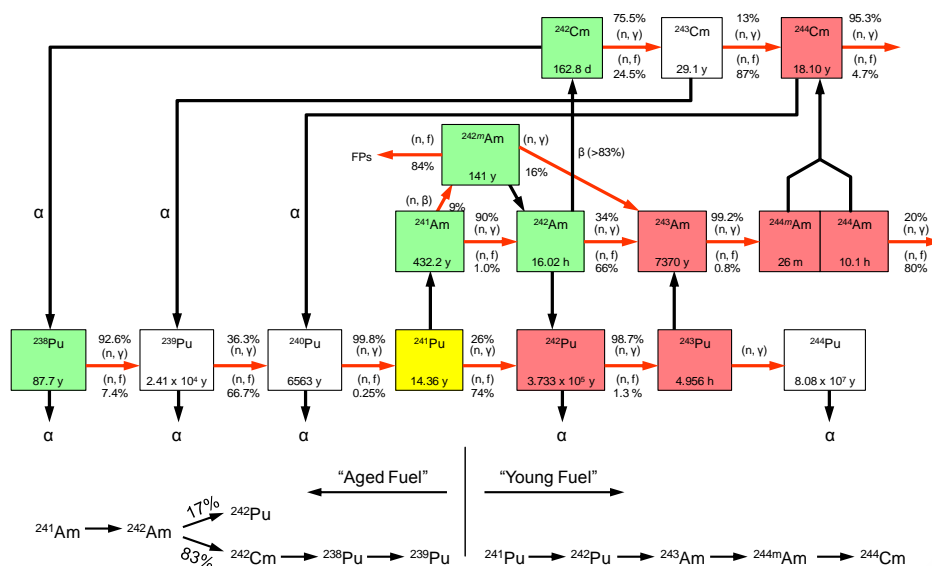
Table 3.2. Comparison of contributions^a of the transuranic elements

	Decay heat ^b	Spontaneous fission neutrons ^b	Photons ^b
Pu	45%	0.6%	18%
Np	0%	0.0%	0%
Am	11%	0.02%	70%
Cm	44%	99.4%	12%
%Cm in Am-Cm	80	~100	14.6
^a Based on calculations by J. Stillman, "Proliferation Resistance Metrics for Separated Streams of Reactor Grade Transuranics," <i>Trans. Am. Nucl. Soc.</i> , 92, 282–283 (June 2005). ^b Used fuel that has been irradiated for 55 GWd/MT and decayed for 5 years.			

Even though the type of shielding required is different for americium (gamma shielding) and curium (neutron shielding), both types of shielding can be provided together in hot cell construction. Even if curium is separated from americium, subsequent treatment of each separated element will require shielded facilities. Therefore, separation of curium from americium does not lessen the need for shielded facilities for recycle target fabrication (americium) or decay storage (curium).

The benefits of reprocessing used fuels after a significantly long storage period (30 to 70 years) have been determined and described in recent systems studies [2]. One of the major benefits is the alteration of the transuranium (TRU) actinide transmutation pathway to produce lighter plutonium nuclides (²³⁸Pu, ²³⁹Pu) rather than heavier element nuclides (²⁴²Pu, ²⁴³Am, ²⁴⁴Cm, ²⁵²Cf, etc.), as illustrated by the pertinent portion of the chart of the nuclides, shown in Figure 3.1.

Figure 3.1. Transmutation benefits of older fuel—alters transmutation pathway to produce lighter plutonium nuclides rather than heavy curium nuclides

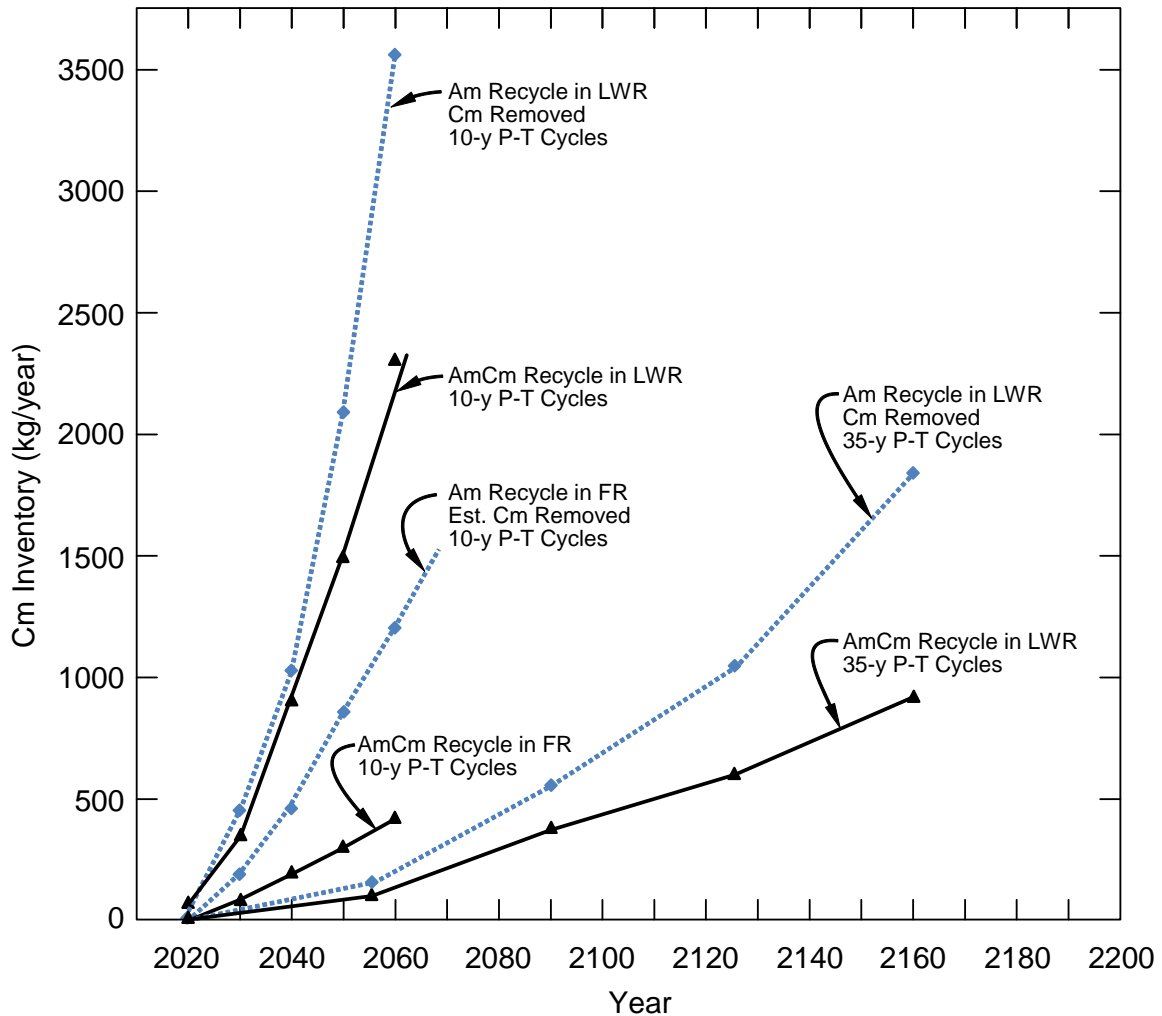


4

In this chart, “aged fuel” is defined as two or more half-lives of ^{241}Pu (>30 years). Recycling lesser-aged fuels (“young fuel”) produces larger amounts of the heavy element TRU nuclides (^{242}Pu , ^{243}Am , ^{244}Cm , ^{252}Cf , etc). In the United States, an inventory of aged fuel is accumulating and the reprocessing of such fuel (recycling the “oldest-fuel-first”) may be taken advantage of.

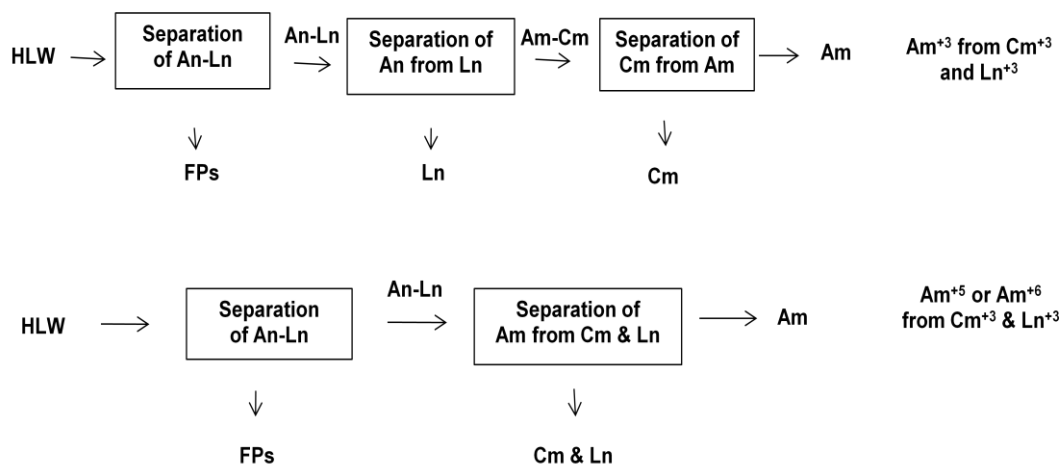
Recent systems analysis studies have compared the effects of recycling both unseparated americium-curium or only separated americium during five successive partitioning-transmutation (P-T) cycles using both thermal spectrum pressurised-water reactors and fast spectrum burner reactors. The data for the curium inventory at the beginning of each transmutation cycle are shown in Figure 3.2 for a P-T series with 5-year decay periods between cycles (10-year P-T cycles) and for a P-T series with 30 years decay periods between cycles (35 years P-T cycles). All three sets of data show that the accumulated curium inventory is greater when the curium is removed and stored at the end of each P-T cycle than when the americium and curium are recycled together. When curium is stored for decay, the decay daughter is ^{240}Pu (half-life = 6570 years), which is another long-lived TRU actinide. When curium is irradiated, however, most of the destruction is by fissioning of ^{245}Cm , ^{247}Cm , and ^{251}Cf . The implications are that it is not beneficial to separate curium from americium. In addition, when such separation does not occur, there is no need for managed storage of the curium.

Figure 3.2. Comparison of curium inventory during multiple partitioning-transmutation cycles when curium is recycled and when curium is removed and stored at the end of each cycle



Most of the reprocessing separative steps being developed for advanced fuel cycles (Figure 3.3) require separate processes for recovery of the combined trivalent actinides (americium and curium) and lanthanide fission products, followed by a complex process for partitioning americium-curium from the lanthanides.

Figure 3.3. Separations processes for americium



If curium is to be removed from the americium, another complex process will be required. However, if a simplified industrial-scale separation process for americium and curium can be developed by means of oxidising americium to the pentavalent or hexavalent state, then it may be more reasonable to consider removal and managed storage of the curium.

3.1 Summary

In summary, most of the factors affecting a decision to recycle curium in the United States appear to favour recycling without separation from americium, while reprocessing the used fuels after a decay storage period of 30 to 70 years. Fuels in this age range are already accumulating in the United States. Further research and development for the advanced fuel cycle may provide an incentive to separate and store the curium.

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List of contributors

Chair

Pascal Baron (CEA, France)

Scientific Secretary

Yong-Joon Choi (OECD/NEA)

Executive Summary

Emory D. Collins (ORNL, USA)

Chapter 1. France

Pascal Baron (CEA, France)

Lionel Boucher (CEA, France)

Christine Coquelet (CEA, France)

Christine Rostaing (CEA, France)

Chapter 2. Japan

Yasuji Morita (JAEA, Japan)

Chapter 3. United State of America

Emory D. Collins (ORNL, USA)

Members of the expert group

Canada

HYLAND, Bronwyn AECL

Czech Republic

UHLIR, Jan NRI

France

BARON, Pascal (Chair) CEA

WARIN, Dominique CEA

Italy

DE ANGELIS, Giorgio ENEA

LUCE, Alfredo ENEA

Japan

INOUE, Tadashi CRIEPI

MORITA, Yasuji JAEA

MINATO, Kazuo JAEA

Korea

LEE, Han Soo KAERI

Russian Federation

IGNATIEV, Victor V. KIAE

KORMILITSYN, Mikhail V. RIAR

Spain

CARAVACA, Concepcion CIEMAT

United Kingdom

LEWIN, Robert G. NNL

TAYLOR, Robin J. NNL

United States of America

COLLINS, Emory D.

ORNL

LAIDLER, James J.

ANL

International Organisations

CHOI, Yong-Joon (Secretary)

NEA

GLATZ, Jean-Paul

EC