MINOR ACTINIDES TRANSMUTATION SCENARIO STUDIES WITH PWRs, FRs AND MODERATED TARGETS

Jean-Paul Grouiller,[1] **Sylvie Pillon,**[1] **Cyrille de Saint Jean,**[1] **Frederic Varaine**[1]
Lydie Leyval,[2] **Guy Vambenepe**[3] and **Bertrand Carlier**[4]

1. CEA/DEN, Cadarache, BP 1, F-13108 St Paul lez Durance, France
2. CEA/DEN, Valrho-Marcoule, B.P. 171, F-30207 Bagnols sur Ceze, France
3. EDF/SEPTEN 12-14 avenue Dutrievoz, F-69628 Villeurbanne Cedex, France
4. FRAMATOME ANP, Tour Framatome, La Défense 6, F-92084 Paris-la-Défense Cedex, France

Abstract

Using current technologies, we have demonstrated in this study that it is theoretically possible to obtain different minor actinide transmutation scenarios with a significant gain on the waste radio-toxicity inventory. The handling of objects with Am+Cm entails the significant increase of penetrating radiation sources (neutron and γ) whatever mixed scenario is envisioned; the PWR and FR scenario involving the recycling of Am + Cm in the form of targets results in the lowest flow.

In the light of these outcomes, the detailed studies has allowed to:

- design a target sub assembly with a high fission rate (90%);
- define a drawing up of reprocessing diagram with the plant head, the minor actinide separation processes (PUREX, DIAMEX and SANEX).

Some technological difficulties appear in manipulating curium, principally in manufacturing where the wet process (“sol-gel”) is not acquired for (Am+Cm).

Objectives

The aim of this study was to assess the performance of all the separation – transmutation scenarios chosen using conventional technologies. Performance is evaluated over two stages:

- transition between the current situation and the steady-state situation;
- the steady-state situation.

This study was a theoretical exercise, carried out by CEA in collaboration with "EDF" (French Electricity Company) and Framatome. It does not predict any underlying industrial strategy, nor the electricity generator's support for any studied scenario, the relevance of which would need to be assessed, following the study, according to industrial realities in an economic context possibly prevailing in the time period under consideration.

Description of chosen scenarios

Up to 2010, the scenarios have a common period, after this date the chosen scenarios vary and are compared with the "open cycle" scenario.

In steady state, scenarios are based upon three main reactor fleet families:

- A pure PWR reactor type fleet loaded with MIX fuel (Pu in an enriched U support):
  - Reference: Pu only multi-recycling (PWR(MIX)Pu);
  - Variation 1: homogeneous Pu, Np, Am and Cm multi-recycling (PWR(MIX)Pu+MA).
- A pure isogenerator FR type reactor fleet:
  - Reference: Pu only multi-recycling (FR-Pu);
  - Variation 1: Pu, Np, Am and Cm homogeneous multi-recycling (FR-Pu+MA).
- A pure PWR type reactor fleet loaded with UOX fuel and FR type sub-generator reactors:
  - Reference: multi-recycling of Pu only (PWR+FR-Pu);
  - Variation 1: Pu, Np homogeneous multi-recycling and once through cycling of (Am + Cm) targets (PWR+FR-Pu+MA).

Scenario joint suppositions

Fleet evolution up to the year 2010 was simulated using the COSI code [1] starting from the situation existing in 1998 with recycling of Pu in the form of MOX in PWR type reactors.

The fleet's electrical power is 60 GWe producing 400 TWhe, reload average burn-up are of the order of 60 GWd/t for UOX and MIX fuels and approximately 140 GWd/t for FNR fuels. Prior to reprocessing a minimum cooling time of 5 years is required; fuel ageing time is 2 years.

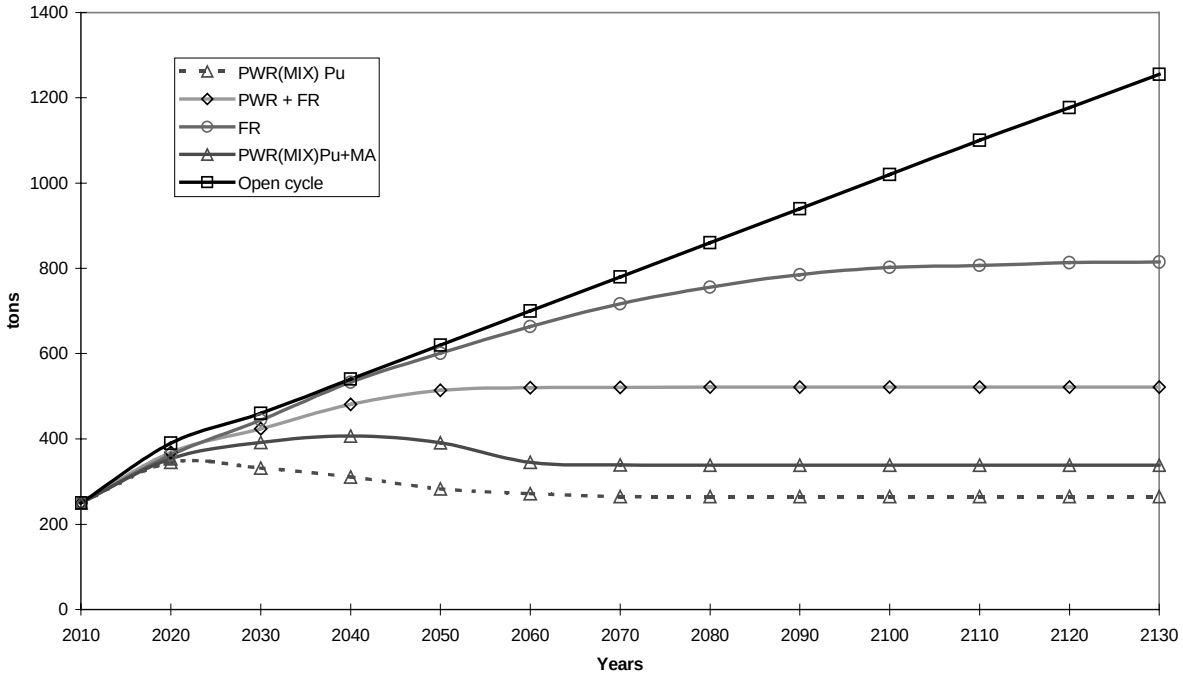
In a moderated FR spectrum, (Am + Cm) target irradiation limits in a moderated spectrum lead to a maximum fission rate of 90%.

U and Pu have reprocessing loss rates of 0.1%. Two loss rates are considered for minor actinides: 1% and 0.1%. Uranium from reprocessing is stored.

Transition scenarios

Starting from the fleet situation in 2010, the various selected options were studied for each scenario. Using the COSI code makes it possible to take into account the fleet's status in 2010 with both the irradiated fuels (UOX and MOX) stored in pools, the cycle functions (enrichment, manufacturing, reprocessing), the various types of reactors and the associated fuels. Pu contained in the irradiated fuels allows a transition strategy to be implemented with various options introduced (that is to say that the reactors existing in 2010 are progressively replaced or modified at the rhythm imposed by Pu availability; thus the MIX fuel loaded PWR fleet is reached after 20 years, a fleet composed of 45% PWR (UOX) and 55% FR is reached after 60 years and a pure FR fleet after 130 years. This detailed simulation of fleet evolution allows nuclear material inventory evolution to be calculated (mass and isotopes), in the installations, in the reactors, storage in facilities, and in waste packages. The Figure 1, hereunder, shows fleet Pu inventory evolution.

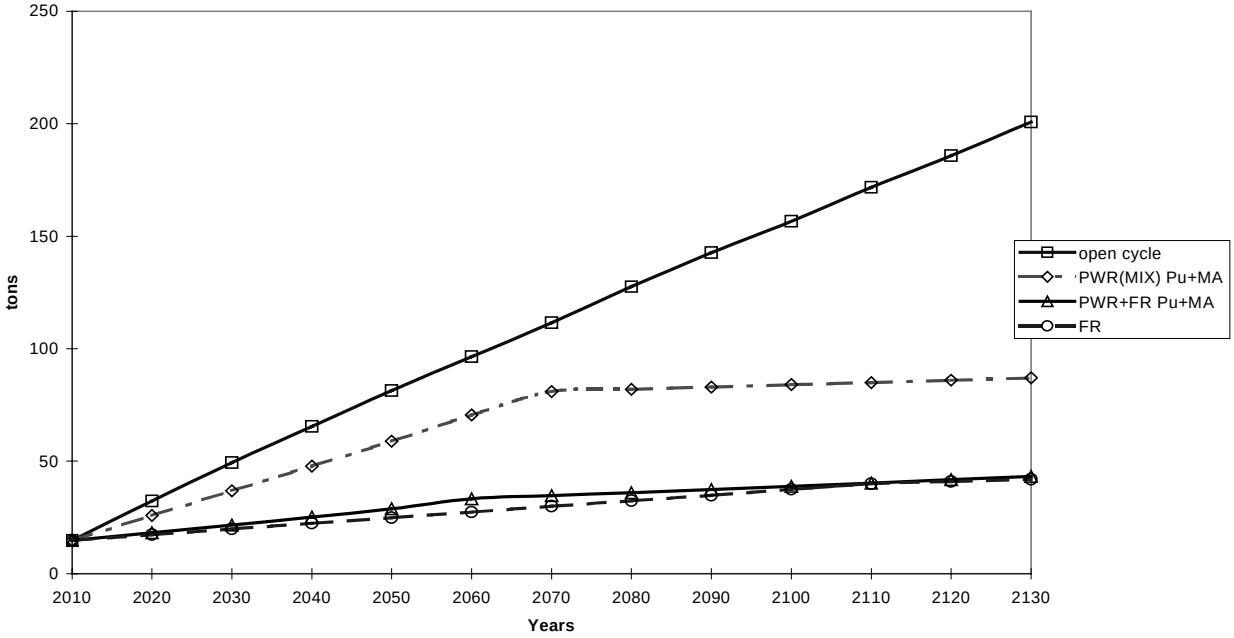
Figure 1. **Pu inventory**



The Figure 2, hereunder, shows Am+Cm inventory evolution in the fleet for the latter recycling scenarios. The inventory includes the masses contained in the incineration cycle as well as the accumulation of masses remaining in the final waste. In any case, minor actinide recycling results in a fleet inventory less than the one which is in the open cycle. The pure MIX loaded PWR fleet provides a minor actinide inventory that is greater than the one of the other two fleets with FRs, by a factor of 2.

The 10% of Am + Cm remaining in irradiated targets that are not reprocessed entails a 100 kg/year increase in Am + Cm for the PWR+FR fleet at steady state (fleet inventory + wastes). This growth caused by waste accumulation is 60 kg per year for MIX loaded in PWRs and 30 kg per year for the FR fleet with a minor actinide reprocessing loss rate of 1%.

Figure 2. **(Am + Cm) inventory**



The following table gives the steady state stabilisation level of nuclear matter inventories for the various scenarios. These inventories (tons) include the weight of every element present in the cycle installations (Manufacture, Reactor, Storage, Reprocessing).

Table 1. **Steady state stabilisation level**

Scenarios	Pu	Np	Am	Cm
PWR(MIX)Pu	260			
PWR(MIX)Pu+MA	340	13	34	47
FR-Pu+MA	810	4	32	8
PWR+FR-Pu+MA	510	11	18*	5*

* without Am and Cm accumulated in irradiated targets

The PWR V1 scenario using MIX fuel leads to the lowest Pu inventory in the fleet and the highest minor actinide inventory (the major part being curium). A large curium flux is associated with this high inventory (3.6 tons per year) to be handled during multi-recycling operations, which implies a significant increase in protective measures against penetrating radiations at the various of these operations.

The PWR – FR V1 scenario with locally moderated targets leads to Pu inventory upper than 50% with respect to PWR V1 and the lowest minor actinide inventory. A minimal curium flux (0.2 ton per year) is associated to be handled during once through cycling operations.

Scenarios at steady state

The following diagrams give the steady state stabilisation level of nuclear park for the various Pu and Minor Actinides recycling scenarios.

Diagram 1

PWR(MIX) – 60 Gwe – 400 Twhe/Year
Burn up: 60 000 MWd/t
(reprocessing losses : 0.1%)

Pu + Np + Am + Cm recycling in PWR(MIX) **Annual balance**

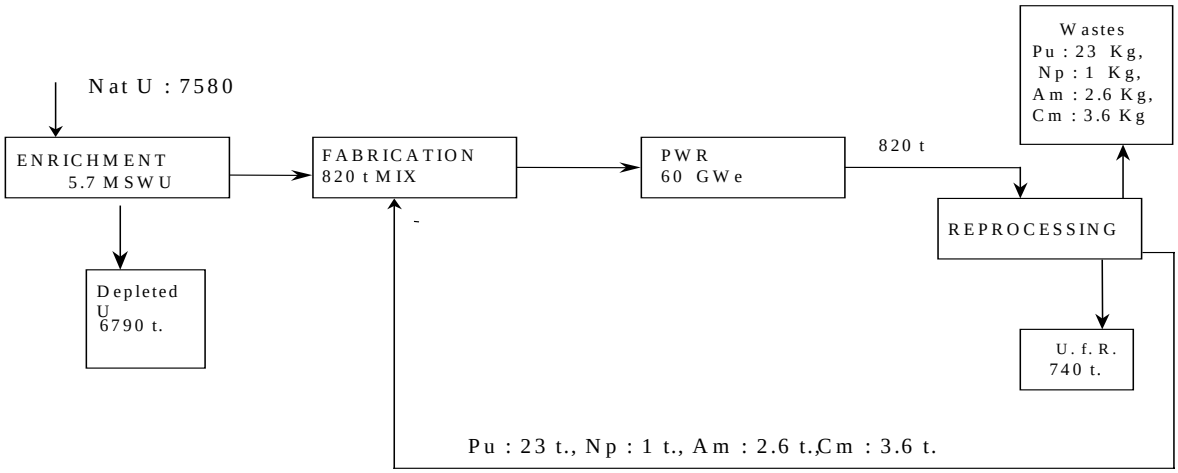


Diagram 2

FR(MOX) – 60 Gwe – 400 TWHe/year

Burn up FR: 140 000 MWd/t
(reprocessing losses : 0.1%)

Pu + Np + Am + Cm recycling in FR(MOX) **Annual balance**

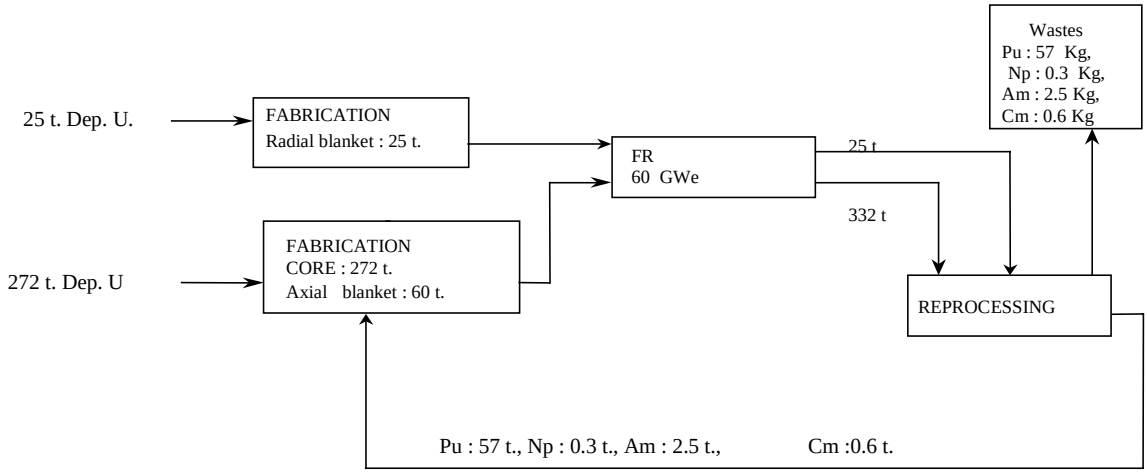
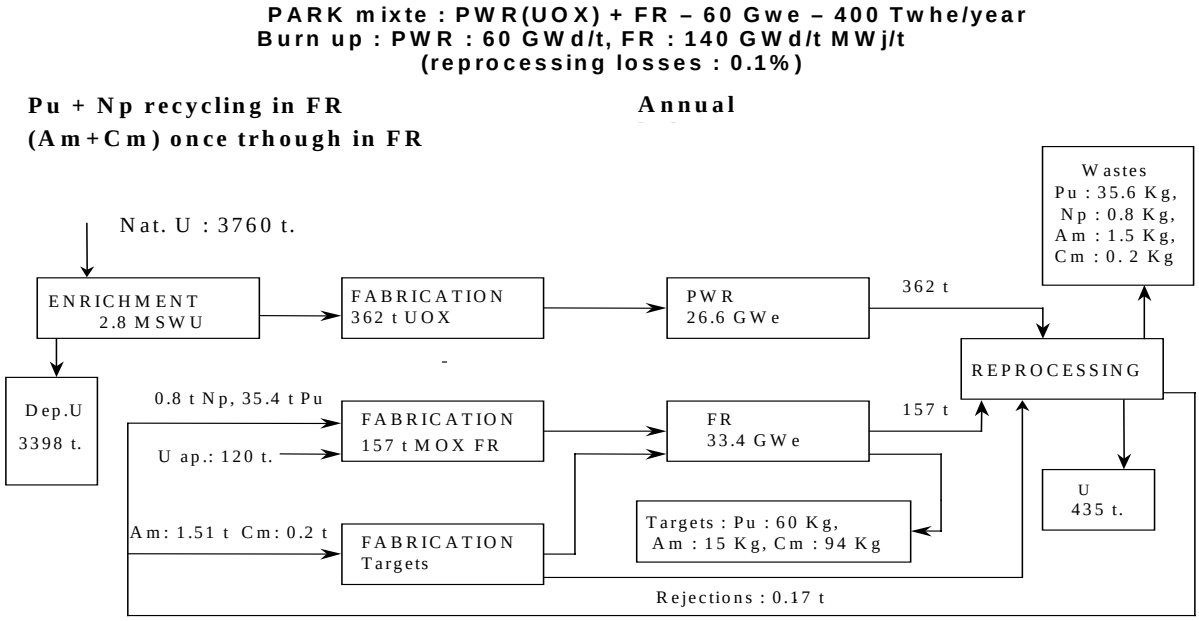
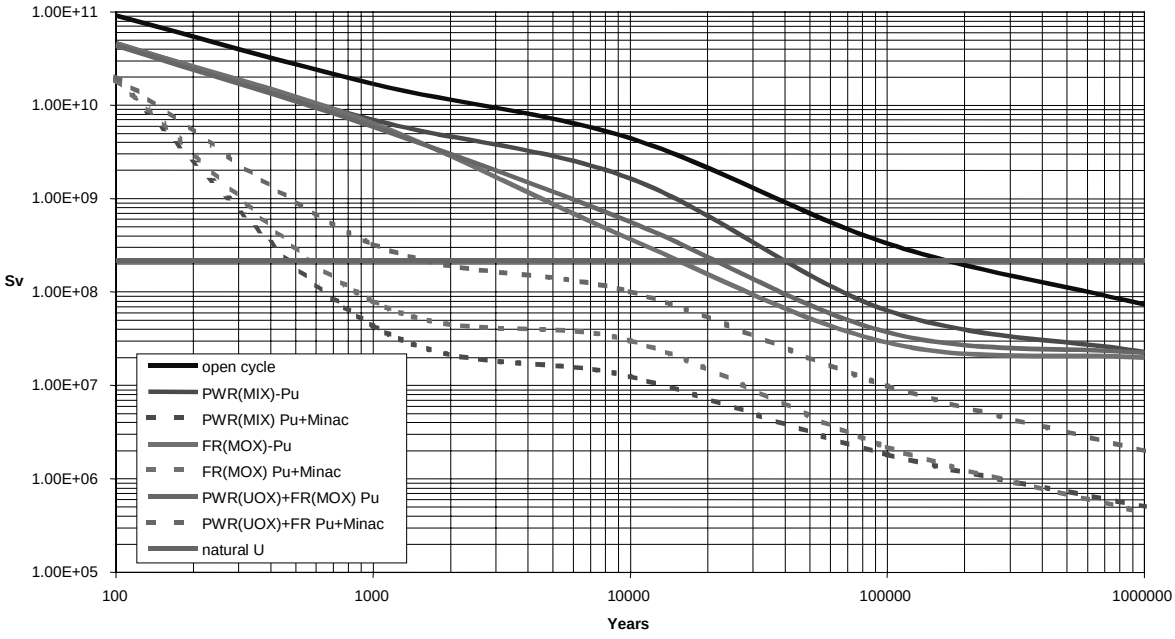


Diagram 3



Evolution, over time, of the radiotoxic inventory through ingestion (CIPR 72 coefficients) of ultimate waste (Pu, Np, Am Cm) produced, every year, by the various fleets, is given in the Figure 3 hereunder with a theoretical 0.1% loss of actinides during reprocessing.

Figure 3. **Waste radiotoxicity inventory produced every year**



To assess the efficiency of an actinide incineration option, the radio-toxicity inventory is analysed between 500 and 100 000 years; period where the gain is obtained from Pu, Am and Cm recycling. The gain obtained with Np recycling appears after 500 000 years.

With respect to the open cycle, Pu recycling allows a reduction factor ranging from 3 to 10, according to the cooling time, and it is noted that FRs are approximately twice as performant as MIX loaded PWRs.

Homogeneous minor actinide multi-recycling, with a loss rate of 0.1%, allows a reduction factor ranging between 200 and 400 with a pure MIX PWR or pure FR fleet. The table, hereunder, gives the times where the waste radio-toxicity inventory is the same of natural uranium one.

Scenarios	**Time (years)** **Rad (scénario) ~ Rad (Unat)**
Open cycle	200 000
Pu recycling in PWRs (MIX)	40 000
Pu recycling in FRs	20 000
Recycling in FRs, homogeneous mode for Pu+Np, once through cycling for Am+Cm	2 000
(Pu+Np+Am+Cm) recycling in FRs or PWRs(MIX)	500

The heterogeneous once through cycling of Am + Cm (which does not require used target reprocessing) in FRs already allows a reduction factor of 60; this is approximately two times lower due to 10% of actinides remaining in the irradiated targets. The Am only once through cycling in FRs allows a reduction factor of 30.

Improving minor actinide reprocessing losses from 0.1% to 1% has a limited influence, since at the best it allows waste radio-toxicity to be lowered by a factor of 4. When the minor actinide loss rate is 0.1%, Pu losses (0.1%) in waste have a significant contribution.

Detailed studies

Using current technologies, we have demonstrated in this study that it is theoretically possible to obtain different minor actinide transmutation scenarios with a significant gain on the waste radio-toxicity inventory.

It is however necessary to conduct studies in order to assess:

- the technical feasibility of various industrial operations in the fuel cycle;
- the environmental impact.

This handling of objects with Am+Cm entails the significant increase of penetrating radiation sources (neutron and γ) whatever the scenario envisioned; the (PWR-FR V1) scenario involving the recycling of Am + Cm in the form of targets results in the lowest flow.

In the light of these outcomes, the detailed studies underway (with the EDF, Framatome and Cogema) focus on this scenario as a priority and the results will be extrapolated with two other minor actinide homogeneous recycling scenarios.

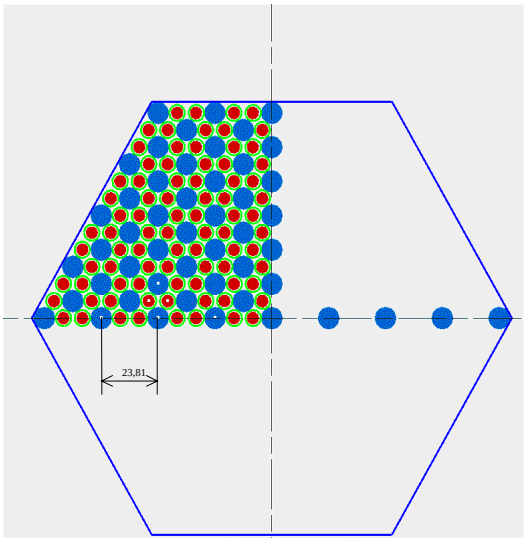
The table hereunder shows the physical scale of interest of objects to be manufactured at the fabrication and reprocessing plants. These results are the basic data of technical feasibility studies. In these studies, we have supposed the californium produced by Cm transmutation, stays in the waste with fission products.

	Flux Am (t/year)	**Flux Cm** (t/year)	**Decay heat** (kW/assembly)	**Neutron sources** (n/s/assembly)
MIX fabrication 820 t/year	2.3	3.6	4.9	1.5 E10
Target fabrication 1.6 t/year (Am+Cm)	1.4	0.2	2.3	6.3 E09
MOX FR fabrication 330 t/year	2.5	0.6	0.9	2.2 E09
MOX fabrication 12% Pu			1.7	7.3 E07
MIX reprocessing 820 t/year	2.6	3.6	6.5	1.5 E11
MOX FR reprocessing 330 t/year	2.5	0.6	1.4	2.8 E09
Mixed reprocessing 360 t UOX 160 t MOX	1.4	0.2	1.82 UOX 0.85 MOX	6.6 E8 UOX 5.0 E8 MOX

Design of moderated target S/A

The studies (neutronics and thermal) led to a selection of an assembly with target rods of Am+Cm on an inert support and a moderation rods [2] to obtain a very high fission rate (90%). The theoretical target concept (Figure 4) has got the highest mass consumption with an acceptable maximum linear power. This target S/A respects all the technological limitations: clad thickness, pressure drop, temperature in pins, mechanical sustaining.

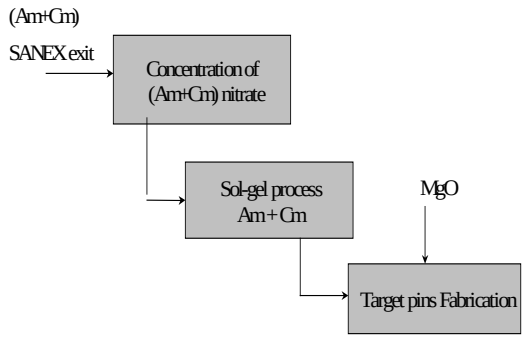
Figure 4. **(Am+Cm) Target S/A concept**



Volume percent (Am+Cm)O	20.81
Target pin diameter	5.23 mm
Moderating pin diameter	9.2 mm
Moderator	YH_2
Max temperature	2 050°C
Inert matrix	MgO
(Am+Cm) transmutation	110 g/Twhe/S.A

Figure 4. **(Am+Cm) Target S/A concept** (contd).

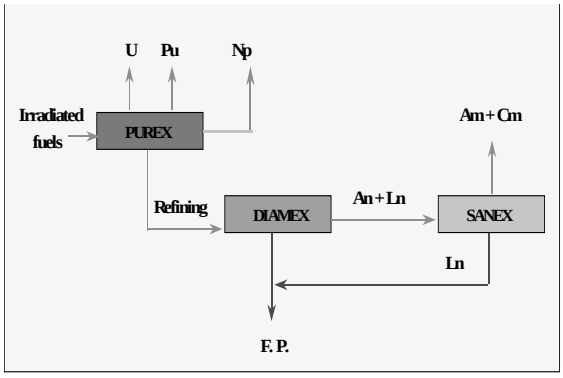
Definition of a manufacturing process for (Am+Cm) pellet



The study [3] of manufacturing process for (Am+Cm) pellet (90% Am and 10% Cm) shows some difficulties:

- The radiation emissions (γ (Am) and neutrons (Cm)) will dictate a large radiological protection.
- The thermal emission (α (Cm)) will impose a continuous cooling, avoid interim storage.
- Wet process will be necessary but it has been used in laboratory for U and Pu. We think it will be possible to extrapolate for Am only but not acquired for (Am+Cm).
- In any case the manufactory process will use hotshops with vitrification technology like R7 and T7 in La Hague.

Definition of reprocessing process



The study on reprocessing process has been made from the PUREX process with some extensions to take into account the specific aspects of scenario:

- Two separate channels to dissolve UOX fuel from PWRs and MOX fuel from FRs.
- PUREX process to separate U, Pu+Np.
- DIAMEX and SANEX processes to separate (Am+Cm).

Supposing minor actinides separation processes have been entirely defined (these are currently under development in the R&D Atalante facility), the results don't show difficulties to put in operation the minor actinides separation for our scenario.

Conclusion

Using current technologies, we have demonstrated in this study that it is theoretically possible to obtain different minor actinide transmutation scenarios with a significant gain on the waste radio-toxicity inventory. The handling of objects with Am+Cm entails a significant increase of penetrating radiation sources (neutron and γ) whatever mixed scenario is envisioned; the PWR and FR scenario involving the recycling of Am + Cm in the form of targets results in the lowest flow.

In the light of these outcomes, the detailed studies has allowed to:

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Some technological difficulties appear in manipulating curium, principally in manufacturing where the wet process ("sol-gel") is not acquired for (Am+Cm).

The conclusions for minor actinides transmutation are:

- Np: solution in homogeneous multi-recycling mode but this solution leads to a significant gain on the waste radio-toxicity inventory only after 500 000 years;
- Am: solution in heterogeneous once through cycling mode in FRs to limit the annual material flux and the Cm production; the reduction factor on the waste radio-toxicity inventory is 30;
- Cm: large impact on radiation and thermal aspects in the cycle; a solution remains to be find.

REFERENCES

[1] J.P. Grouiller *et al.* (1991), *COSI, A Simulation Software for a Pool of Reactors and Fuel Cycle Plants*,"Fast reactors and related fuel cycles", October, KYOTO/Japan.

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[3] S. Pillon *et al.*, *Impact of the Curium Management on the Fabrication of Ma-bearing Targets at an Industrial Scale in the Frame of a Mixed PWR and FR P&T Scenario*, 7th IEM-P&T.