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Minor actinides transmutation scenario studies with PWRs, FRs and moderated targets

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Abstract

In this study it is demonstrated that it is theoretically possible to obtain different minor actinide transmutation scenarios with a significant gain on the waste radio-toxicity inventory using current technologies. The handling of materials containing Am + Cm entails a significant increase of penetrating radiation sources (neutron and γ) whatever mixed reactor scenario is envisioned: The PWR and fast reactor scenario involving the recycling of Am + Cm in the form of targets results in the lowest mass 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 reprocessing scheme for the plant head and 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 well suited for (Am + Cm).

1. Introduction

The aim of this study is the assessment of the performance of several choices in separation–transmutation that make use of conventional technologies. The 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 (French Atomic Energy Commission) 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 this study, according to industrial realities in an economic context possibly prevailing in the time period under consideration.

2. Description of chosen scenarios

Up to 2010, the scenarios have on a common period, after this date the chosen scenarios vary and are compared with the 'open cycle' on scenario. In steady state,

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scenarios are based upon three main reactor fleet families:

Scenario 1: A PWR reactor type fleet loaded with MIX fuel (Pu in an enriched U support).

- Reference: Only Pu multi-recycling (PWR-(MIX)Pu).
- Variation 1: Pu, Np, Am and Cm multi-recycling in a fuel assembly (PWR(MIX)Pu + MA).

Scenario 2: A pure isogenerator fast reactor (FR) type reactor fleet.

- Reference: Only Pu multi-recycling (FR-Pu).
- Variation 1: Pu, Np, Am and Cm multi-recycling in a fuel assembly (FR-Pu + MA).

Scenario 3: A 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 multi-recycling in a fuel assembly and once through cycling of (Am + Cm) targets (PWR + FR-Pu + MA).

3. General assumptions

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 annually, 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 with loss rates of 0.1%. Two loss rates are considered for minor actinides: 1% and 0.1%. Uranium from reprocessing is stored.

4. Transition scenarios

Starting from the fleet situation in 2010, the various selected options were studied for each scenario. 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 irradiated fuels permits a transition strategy to be implemented with various options introduced (that is to say the reactors existing in 2010 are progressively replaced or modified at the rhythm imposed by Pu availability; thus the MIX fuel loaded in a 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 the nuclear material inventory evolution to be calculated (mass and isotopes), in the installations, reactors and storage in facilities, and in waste packages. Fig. 1 shows the evolution of the Pu inventory in the fleet.

Fig. 2 shows the evolution of the Am + Cm inventory in the fleet for the different recycling scenarios. The inventory includes the masses contained in the incineration cycle as well as the accumulation of masses

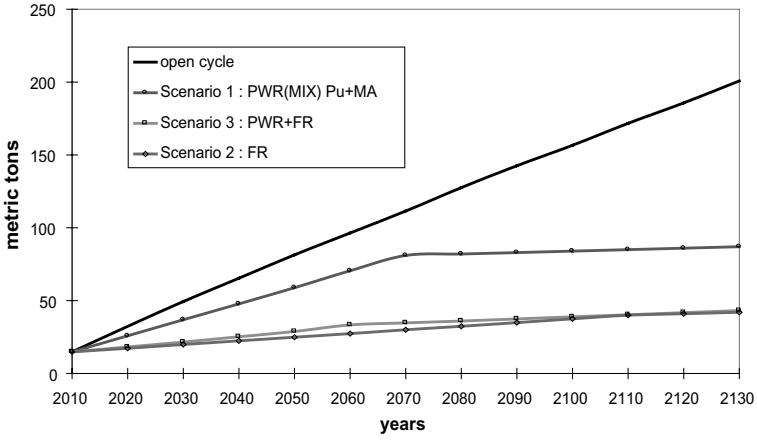


Fig. 1. Evolution of the Pu inventory for the different scenarios.

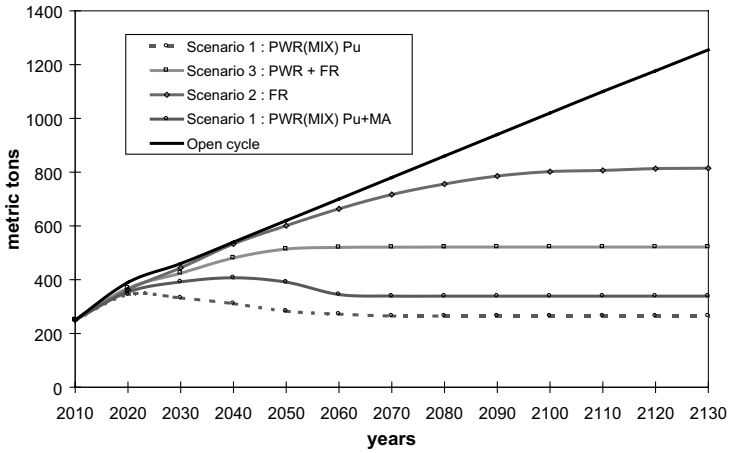


Fig. 2. Evolution of the Am + Cm inventory for the recycling scenarios.

remaining in the final waste. In any case, minor actinide recycling results in a fleet inventory less than in the open cycle. The pure MIX loaded PWR fleet provides a minor actinide inventory that is greater by a factor of 2 than the other two fleets with FRs.

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%.

Table 1 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 (manufacturing, reactor, storage, reprocessing).

Scenario 1 using MIX fuel leads to the lowest Pu inventory in the fleet and the highest minor actinide inventory (the major part being curium). The large curium flux associated with this high inventory (3.6 tons per year) has to be handled during multi-recycling operations, which implies a significant increase in protective measures against penetrating radiation at all of these operations.

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[a]	5[a]

[a] Without Am and Cm accumulated in irradiated targets.

The scenario 3 with locally moderated targets leads to a Pu inventory greater than 50% with respect to scenario 1, and the lowest minor actinide inventory. A minimal curium flux (0.2 ton per year) is present and has to be handled during once through cycling operations.

5. Scenarios at steady state

Figs. 3–5 give the steady-state stabilisation level of the nuclear park for the various Pu and minor actinides recycling scenarios. Evolution, over time, of the radiotoxic inventory through ingestion (ICPR-72 coefficients) of ultimate waste (Pu, Np, Am Cm) produced, every year, by the various fleets, is given in Fig. 6 with a theoretical 0.1% actinide loss during reprocessing.

To assess the efficiency of an actinide incineration option, the radiotoxic inventory is analysed in the 500 and 100 000 years range, i.e. the 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 radio-toxicity reduction by a factor ranging from 3 to 10, according to the cooling time, and it is noted that FRs are performing twice as well 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. Table 2 gives the times where the waste radiotoxicity inventory is the same of natural uranium one.

The heterogeneous once through cycling of Am + Cm (which does not require used target reprocessing) in FRs already allows a reduction factor of 60, which 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.

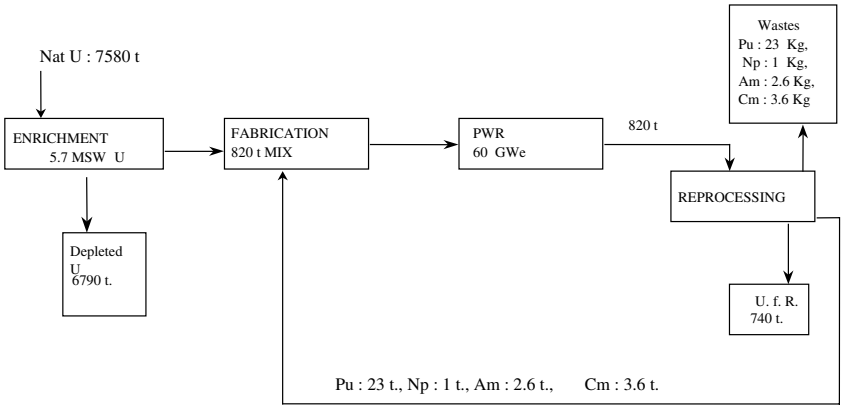


Fig. 3. Scenario 1.

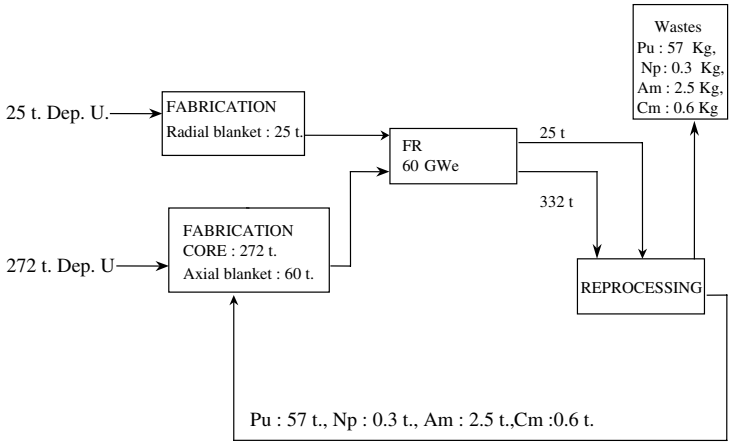


Fig. 4. Scenario 2.

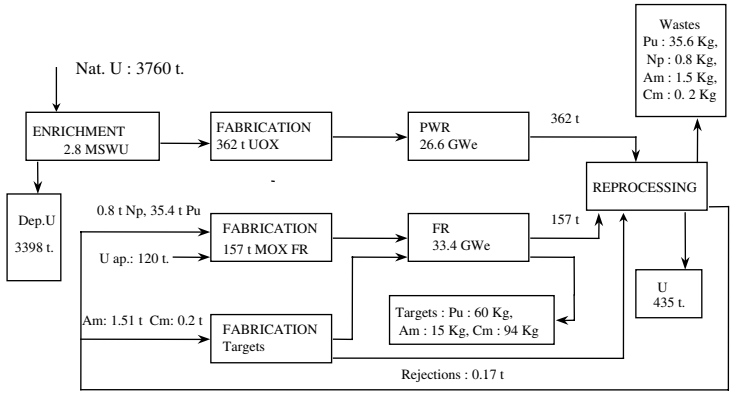


Fig. 5. Scenario 3.

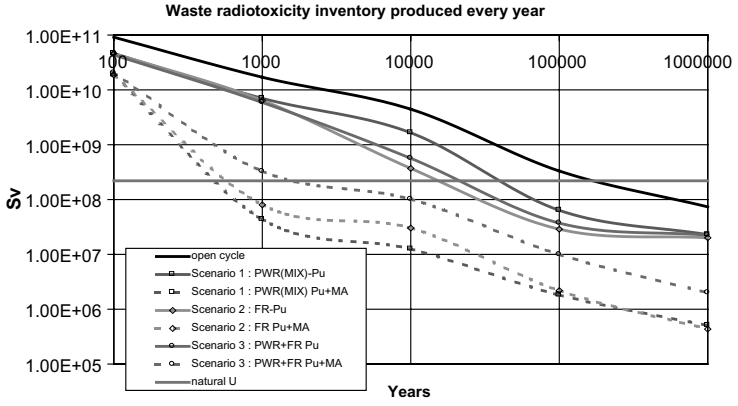


Fig. 6. Waste radio-toxicity inventory.

Table 2
Radio-toxicity inventory

Scenarios	Time (years) rad (Scénario)–rad (Unat)
Open cycle	200 000
Scenario 1: Pu recycling in PWRs (MIX)	40 000
Scenario 2: Pu recycling in FRs	20 000
Scenario 3: Recycling in FRs, homogeneous mode for Pu + Np, once through cycling for Am + Cm	2000
(Pu + Np + Am + Cm) recycling in FRs (Scenario 2) or PWRs(MIX) (Scenario 1)	500

Changing 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.

6. 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 necessary, however, to conduct studies in order to assess:

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

Table 3 shows physical characteristics of materials to be manufactured at the fabrication and reprocessing

Table 3
Physical scale

	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	1.5E10
Target fabrication 1.6 t/year (Am + Cm)	1.4	0.2	2.3	6.3E09
MOX FR fabrication330 t/year	2.5	0.6	0.9	2.2E09
MOX fabrication 12% Pu			1.7	7.3E07
MIX reprocessing 820 t/year	2.6	3.6	6.5	1.5E11
MOX FR reprocessing330 t/year	2.5	0.6	1.4	2.8E09
Mixed reprocessing				
360 t UOX	1.4	0.2	1.82 UOX	6.6 E8 UOX
160 t MOX	1.4	0.2	0.85 MOX	5.0E8 MOX

plants. These results are the basic data of technical feasibility studies. In these studies, we have assumed the californium produced by Cm transmutation, stays in the waste with fission products.

The handling of Am + Cm containing materials entails a significant increase in penetrating radiation sources (neutron and γ) whatever the scenario envisioned; scenario 1 involving the recycling of Am + Cm in the form of target results in the lowest flow.

In the light of these results, the detailed studies are underway (with 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.

7. Design of moderated target sub-assembly

The studies (neutronics and thermal) led to a selection of an assembly with target rods of Am + Cm in an inert support and moderation rods [2] to obtain a very high fission rate (90%). The theoretical target concept (Fig. 7) has the highest mass consumption with an acceptable maximum linear power. This target sub-assembly (S/A) respects all technological limitations: clad thickness, pressure drop, temperature in pins, mechanical stability.

The study of manufacturing process for (Am + Cm) pellet (90% Am and 10% Cm) shows some difficulties [3] (see Fig. 8):

- The radiation emissions (γ(Am) and neutrons (Cm)) will necessitate a large radiological protection.
- The thermal emission (α(Cm)) will require continuous cooling and the avoidance of interim storage.
- Wet processes will be necessary but it has only been achieved in laboratory for U and Pu. We think it will be possible to extrapolate for Am only but not without difficulty for (Am + Cm).
- In any case the manufactory process will use hot cells equipped with similar technology as used for vitrification.

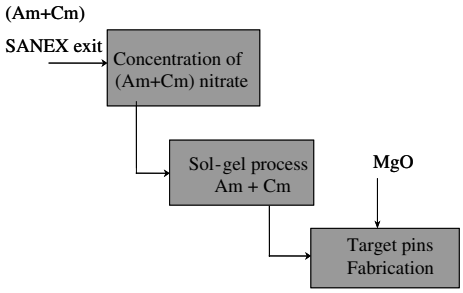


Fig. 8. Definition of a manufacturing process for (Am + Cm) pellet.

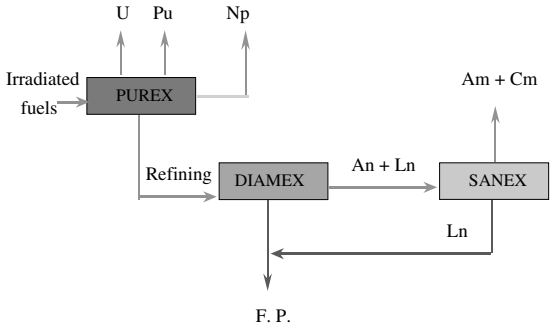
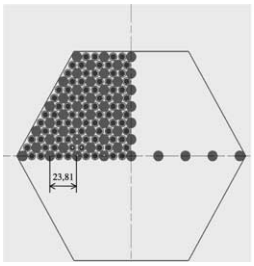


Fig. 9. Definition of reprocessing process.

The study on reprocessing processes has been based on the PUREX process with some extensions to take into account the specific aspects of scenario (see Fig. 9):

- 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) [4].
- Assuming minor actinides separation processes have been entirely defined (these are currently under development in the R&D Atalante facility), the results do not show potential difficulties for these scenarios.



Volume percent $(Am+Cm)O_2$	20.81
Target pin diameter O	5.23 mm
Moderating pin diameter O	9.2 mm
Moderator	YH_2
Max temperature	2050 °C
Inert matrix	MgO
(Am+Cm) transmutation balance	110 g/Twhe/S.A.

Fig. 7. (Am + Cm) Target S/A concept.

8. 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 improvement in 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 target results in the lowest flow.

In the light of these outcomes, it has been possible to:

- Design a target S/A with a high fission rate (90%).
- Define plan for the reprocessing for the plant head and the 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 easily suited 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 found.

References

[1] J.P. Grouiller, B. Sicard, G. Flamenbaum, M. Mus, J. Martin, O. Comellini, J.G. Devezeaux, in: Proceedings of Fast Reactors and Related Fuel Cycles, October 1991, KYOTO/Japan.
[2] C. de Saint Jean, J. Tommasi, F. Varaine, N. Schmidt, D. Plancq, in: Proceedings of GLOBAL 2001, Paris, France, 9–13 September 2001 (on cd-rom).
[3] S. Pillon, S. Grandjean, F. Drain, N. Schmidt, J.M. Escleine, A. Ravenet, B. Valentin, C. de Saint Jean, J. Nucl. Mater. in press.
[4] C. Madic, CR Phys. 3 (2002) 797.