TRANSMUTATION OF AMERICIUM AND CURIUM: REVIEW OF SOLUTIONS AND IMPACTS

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Abstract

Several papers have presented studies on scenarios based on Plutonium and minor actinides recycling. Recycling Neptunium allows a reduction of long-term radiotoxicity. Recycling Americium allows a reduction of radiotoxicity of more than a decade between 100 years and 10 000 years. The gain in radiotoxicity is limited due to the Curium produced in the reactors. The Curium transmutation enables greatly the radiotoxicity reduction but presents large consequences on the fuel cycle facilities. The study compares the following solutions: once-through cycle of Americium (and Curium) in dedicated assemblies in the fast reactor, dedicated fast reactor for Americium (and Curium) and the dilution of the actinides in PWR.

Introduction

Several papers [1,2] were presented on scenarios studies of actinides transmutation. This one is focused on the Americium and Curium management with various options, from a once-through transmutation to a multiple recycling scheme.

The transmutation of minor actinides (MA) is coupled on a proper use of Plutonium (Pu). The Plutonium is an energetic material and its recycling in a FBR (Fast Breeder Reactor) with the Uranium permits the use of almost all the initial Uranium and increases the energetic gain by several decades. Today, the studies for an efficient transmutation of minor actinides are linked on different ways of utilising Plutonium [in LWR, in FBuR (Fast Burner Reactor), in FBR]. Based on scenarios of Plutonium recycling, the transmutation of minor actinides is studied and different possibilities are compared.

Recycling Neptunium allows a reduction of long-term radiotoxicity (1 Million of year) and is feasible in a MOX fuel in a homogeneous mode. Several studies have already evaluated the Neptunium transmutation and will not be recalled here.

Recycling Americium allows a reduction of radiotoxicity of more than a decade between 100 years to 10 000 years, but its transmutation produces Plutonium and Curium. Thus the radiotoxicity reduction is limited to a factor 10 or less due to the Curium production in the reactors. To increase the reduction factor up to 100, it is needed to recycle the Curium.

The Curium transmutation enhances greatly the radiotoxicity reduction but presents large consequences on the fuel cycle facilities (fabrication and reprocessing plants, transport, …). In order to evaluate and compare scenarios, an evaluation of:

- The radiotoxicity in the storage.
- The mass flux per element (separated or not).
- The consequences on the cycle physic parameters (activities, neutron emission, …).

is presented in the paper for the following solutions (among others):

- Once-through of Am (and Cm) in dedicated assemblies in FR.
- Dedicated FR (CAPRA) for Am (and Cm).
- Dilution of the actinides in PWR.

Americium production within Plutonium recycling scenarios

The decrease of minor actinides inventory is based on Plutonium recycling scenarios. Indeed, Plutonium is the major element for radiotoxicity and mass in the storage. Table 1 describes the minor actinides production in nuclear park with Plutonium recycling in PWR (Pressurise water reactors) or FR (Fast reactors).

Table 1. **Mass flow to the storage kg/TWhe for a nuclear park (400 TWhe, 60 GWe)**
(Loss: 0.1% for Pu)

	100% PWR UOX (open cycle)	100% FR	32% FR + PWR UOX	18% FR + PWR UOX & MOX	100% FR MOX with enriched U
Pu	28.1	0.16	0.09	0.08	0.05
Np	1.7	0.50	1.2	1.3	1.5
Am	1.3	3.2	4.3	4.6	4.5
Cm	0.26	0.34	0.75	1.0	2.25

The consequences are calculated on the radiotoxicity. The gain on this parameter with Plutonium recycling compared to an open cycle is between a factor 3 and 6 for the time scale (100/10 000 years). The radiotoxicity is then dominated by the Americium, and after, the Curium, the Plutonium losses and the Neptunium for the same time scale. The Curium masses produced are significant, 10% to 20% of the amercium masses for a Plutonium recycling and up to 50% for the Plutonium recycling in a MOX with enriched Uranium support. This last concept, allowing the feasibility, has lower performances compared to a 100% FR park (50% for the Americium and a factor 7 for the Curium).

The once-through option for Americium and Curium recycling

The once-through recycling with a high fission rate (more than 90 % of atoms) is a very attractive solution because:

- The production of Curium due to Americium recycling is managed inside the concept.
- The reprocessing of the target can be simplified, compared to multiple reprocessing (just clean the material, if needed).

The gain on the radiotoxicity varius the fission rate in the target, compared to the open cycle are given in Table 2 for the nuclear park and given in the 4th column of Table 1 (PWR UOX§MOX and FR).

Table 2. **Reduction of radiotoxicity compared to open cycle, using targets in dedicated assemblies (moderated) in FR**

Factor of the radiotoxicity reduction/open cycle	100 years	1 000 years	10 000 years
Open cycle	1	1	1
Pu recycling	3	2,5	4.5
recycling Pu + targets Am			
Maximum (Fission rate 100%)	20	23	15
Fission rate = 90%	12	17	10
Fission rate = 95%	16	20	13
Fission rate = 98%	17	21	14
Pu recycling + targets Am + Cm			
Maximum (Fission rate 100%)	490	400	390
Fission rate = 90%	40	45	30
Fission rate = 95%	72	81	55

For an Americium recycling (without Curium recycling), the maximum gain reachable at 10 000 years after irradiation is clearly limited by the Curium provided by the nuclear park (in the PWR UOX § MOX and in FR). The value at this time is the minimum gain for the time scale between 100 years and 1 million of years. This is why we choose it as a key parameter to compare the transmutation impact in this study.

In order to have an equilibrium between the production of Americium in the park and the transmutation in the dedicated assemblies disposed in FR with a fission rate of 90%, we need slightly more than 30% of FR in the nuclear park.

In this scenario, the gain of radiotoxicity is about 10 compared to an open cycle (see table 2). This gain is small.

More the FR fraction is large, more the radiotoxicity reduction at 10 000 years will be close to the maximum value (100% for the fission rate) due to the smaller build-up of Americium and the larger efficiency in the targets (larger moderation with high flux). Anyway, the gain on the radiotoxicity is limited to a factor 10 to 20 compared to an open cycle. To go further, it is needed to have a Curium and Americium recycling with a fission rate in the targets of about 95% or more. In this case, it is difficult to obtain an equilibrium between transmutation and production in the park if we do not have a majority of FR.

Multiple recycling of Americium and Curium

An other way than transmutation in a once-through mode is multiple recycling in a homogeneous way in FR in dedicated reactors [3]. The radiotoxicity reductions are linked to the losses during the reprocessing.

The gains in radiotoxicity in this case are given in the Table 3.

Table 3. **Reduction of radiotoxicity compared to open cycle**

Factor of the radiotoxicity reduction open cycle	100 years	1 000 years	10 000 years
Open cycle	1	1	1
Pu recycling	3	3	4.5
Pu + Am			
Maximum	20	23	15
Homogeneous recycling Pu, Am (losses *=1%)	7	10	6
Homogeneous recycling Pu, Am (losses *=0.1%)	8	10	6.5
Pu + Am + Cm			
Maximum	490	400	390
Homogeneous recycling Pu, AM (losses***** =1%)	90	100	120
Homogeneous recycling Pu, AM (losses***** =0.1%)	270	335	310
(if losses (Pu, Am, Cm) = 0.01 %)	1000 <	1000 <	1000 <

* **losses:** loss fraction of A.M – For Pu: 0.1 % in all cases.

The transmutation of Americium and Curium are more favourable in a multiple recycling than in a once-through cycle, the multiple recycling being closer to the maximum values (total transmutation in fission products). The gains are sensitive to the losses rates.

Once through recycling - multiple recycling

The previous results show the interest of a Curium and Americium recycling. The consequences on Curium recycling on the cycle facilities must be evaluated.

For the fabrication

In a once-through scenario, added 10 to 20% of Curium in a fresh fuel to the Americium leads to an increase of power, γ doses and a large gap in the neutron emission. Table 4 gives the main effects when Curium is added to Americium in a target.

Table 4. **Impact of the recycling of Americium and Curium for the fabrication of a target (mass ratio ≅ 20 % of M.A. on inert support)**

	100 % Am	90 % Am + 10 % Cm	80 % Am + 20 % Cm
Power	1	× 2.3	× 3.6
G doses at 1 m	1	× 1.5	× 2
Neutrons source	1	× 120	× 240

In a multiple recycling scenario, impacts are lower. The multiple recycling of Americium and Curium presents lower impacts than for the Curium recycling compared to Americium recycling in a heterogeneous disposition.

Table 5. **Impact of the recycling of Americium and Curium at fabrication**

	Heterogeneous recycling (Target ≅20%)		Homogeneous recycling FR (2.5%)	
	Am	Am+Cm	Am	Am+Cm
Power	× 14	× 32	× 1.7	× 3.9
Dose ϒ	× 1 700	× 2 500	× 76	× 115
Neutron source	× 7	× 840	× 1.4	× 170

Ref: Standard fuel EFR CD 9/91.

The dilution allows to reduce the impact.

For reprocessing

The impact on reprocessing is limited, even for the neutron source, as is shown in Table 6.

Table 6. **Impact of the recycling of Americium and Curium**

	Heterogeneous recycling (Target ≅20%)		Homogeneous recycling FR (2.5%)	
	Am (FR) 90%	Am+Cm (FR) 90%	Am	Am+Cm
Power	× 32	–	× 3	× 3.3
Dose ϒ	÷3.5	–	× 1	× 1
Neutron source	–	–	× 3.5	× 4

Ref: Standard fuel EFR CD 9/91.

The tables show a maximum increase of factor 5 at reprocessing step for the option of homogeneous recycling, and larger for heterogeneous recycling.

Once-through or multiple recycling?

The option of Americium and Curium multiple recycling is still open. This solution allows larger radiotoxicity gains and the homogeneous mode is more efficient than once-through. We can optimise once-through and multiple recycling by a two step once-though, limiting the reprocessing mass flux.

This analysis shows the interest of reprocessing with a high resistance to the neutron emission and to the activity in order to reach a gain on radiotoxicity larger than 100.

Conclusions

- The Americium recycling alone allows small gains on radiotoxicity at 10 000 years.
- To increase this gain, the Curium recycling is needed with the Americium.
- The need of a reprocessing with strong resistance is underlined by these studies.
- This is the same for the fabrication route and fuel stability.
- The main options with a large gain in radiotoxicity and a first feasibility assessment in the reactor are:
  - once-through recycling in dedicated assemblies in FR, in a large number of FR.
  - multiple recycling: in PWR, in all the reactors; in FR, with the Plutonium and in a large number of FR.

REFERENCES

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