

## RECYCLING SCHEMES OF AMERICIUM TARGETS IN PWR/MOX CORES

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### Abstract

From the orientation studies performed so far, both ways to recycle Am in PWR/MOX cores, homogeneous in MOX or heterogeneous in target pins, appear feasible, provided that enriched  $\text{UO}_2$  is used as support of the MOX fuel. Multiple recycling can then proceed and stabilise Pu and Am quantities.

With respect to the Pu multiple recycling strategy, recycling Am in addition needs 1/3 more  $^{235}\text{U}$ , and creates 3 times more Curium. Thus, although feasible, such a fuel cycle is complicated and brings about a significant cost penalty, not quantified yet.

The advantage of the heterogeneous option is to allow to manage in different ways the Pu in MOX fuel and the Am in target pins. For example, should Am remain combined to Cm after reprocessing, the recycling of a mix of Am+Cm could be deferred to let Cm transform into Pu before irradiation. The Am+Cm targets could also stay longer in the reactor, so as to avoid further reprocessing if possible.

## Introduction

Fast neutron reactors are best suited to burn Plutonium and, if required, Americium. But in case the deployment of these reactors comes to be largely delayed, it is useful to investigate under which conditions Americium could be recycled in PWR cores, provided it can be recovered at fuel reprocessing.

Americium recycling could proceed either in a homogeneous way if a mixed oxide (U, Pu, Am)O<sub>2</sub> is manufactured and irradiated; or in a heterogeneous way if special Am target pins are fabricated and irradiated together with the usual (U, Pu)O<sub>2</sub> MOX fuel.

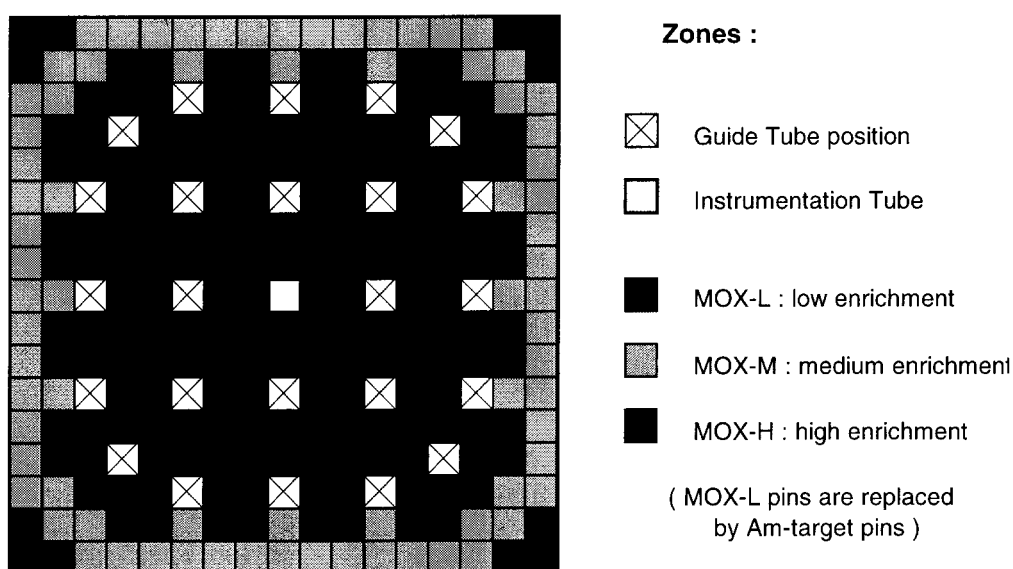
These two recycling modes have been modelled in detailed PWR core physics calculations. Results of homogeneous recycling were reported earlier [1] and are only briefly recalled here. The results of heterogeneous recycling calculations are discussed in details. The problems associated with fuel and target manufacture, will shortly be recalled, too.

## Basic assumptions and methods of calculation

A nuclear network made of 900-MWe PWRs is taken as a basis, with a quarter-core MOX fuel management and an average discharge burn-up of 44 GWd/t. To allow an equilibrium to be reached between transUranium production and consumption, the Plutonium and Americium quantities recovered from reprocessing, 5 years after PWR core discharge, are refabricated over 2 years and then recycled in the same type of reactor. This recycling has been simulated explicitly, using a macrocell comprising a MOX assembly and its neighbouring UO<sub>2</sub> assemblies, a number of successive times large enough to verify that equilibrium is effectively reached.

Figure 1 shows the sketch of the MOX assembly, subdivided into three concentric zones of different Plutonium enrichments, so as to flatten the power and burn-up distribution in the radial direction.

Figure 1. Sketch of MOX Assembly



The MOX fuel is designed to be equivalent in energy to  $\text{UO}_2$  fuel enriched to 3.7 % in  $^{235}\text{U}$ . At discharge all assemblies, loaded with  $\text{UO}_2$  or with MOX, will have been irradiated 4 calendar years at a load factor of 75 %.

Core physics calculations were run using a recent version of the WIMS code package [2] and a JEF 2.2 based library with 172 neutron energy groups. This library is well adapted to high-burn-up MOX fuel irradiation: as an example, the giant low-energy resonance of  $^{242}\text{Pu}$  at 2.7 eV is covered by 8 groups. Also, the burn-up chains explicitly include 3 Americium isotopes and 4 Curium isotopes.

### **Homogeneous recycling of Plutonium and Americium**

In the case of homogeneous recycling [1], the MOX fuel pins contain, either ( $\text{UO}_2\text{PuO}_2$ ) as usual, so as to provide a reference, or ( $\text{UO}_2\text{PuO}_2\text{AmO}_2$ ) in the variant MOX-X. First of all, calculations were done using depleted  $\text{UO}_2$ , containing only 0.25%  $^{235}\text{U}$ , as MOX fuel support.

For the MOX-X recycling case, the required Pu+Am enrichments grow significantly with the successive recycling steps. In the reference MOX (Pu) case, these are calculated to be 7.4, 9.1 and 10.4%, while in the MOX-X case they would reach 8.7, 12.2 and 15.1 %, respectively. This leads to serious difficulties to keep the MOX loading compatible to the usual design and safety objectives. It is generally admitted that a Pu enrichment of 12 % is about the upper limit to keep non-positive coolant void reactivities. This void effect has been calculated for the beginning-of-life situations with fresh fuel which are most penalising. Only the MOX-X1 step indeed retains a non-positive void effect; the subsequent steps MOX-X2 and MOX-X3 would have positive void values, what is not presently acceptable.

Two remedies were proposed in [3] to keep Pu enrichments in an acceptable range: enhanced moderation, or use of enriched Uranium as MOX support.

The enhanced moderation consists of increasing the pin spacing (or reducing the pin thickness) so that the MOX assembly gets a larger water moderation. With the standard PWR lattice the volume ratio of water to oxide is 1.9; one could increase it to 3 or more (but the core volume is also increased). To identify the trends, calculations have been run for a few recycling steps in the same 900-MWe PWR conditions as above.

Thanks to the enhanced moderation, the necessary Pu enrichments are substantially lowered. When recycling Pu only, they become, respectively 4.75% (MOX1) and 6.3% (MOX2), instead of 7.4 and 9.1%. When recycling Pu+Am, they become 5.25% (MOX-X1) and 8.0% (MOX-X2).

This fuel concept can very effectively burn  $^{239}\text{Pu}$ , but it also produces much  $^{242}\text{Pu}$ , source of  $^{243}\text{Am}$ . While this helps to stabilise total Pu quantities at a lower level, the Am quantities are not efficiently decreased, and the Cm quantities are growing.

The enhanced moderation, very promising in the first steps, is however not a better solution with repeated recycling as the enrichments again become prohibitive and the void effect positive. Such conclusions meet those found in [4]: at equilibrium the Pu enrichment reaches about 18%, which is by far excessive.

Another remedy was thus tried, which is to replace in the MOX fuel the support Uranium, usually depleted, by Uranium enriched in  $^{235}\text{U}$ . So the Pu enrichment of the MOX fuel can be kept

equal to a constant, acceptable value, for example 8% in the average.  $\text{UO}_2$  with an increasing enrichment would thus be added to  $\text{PuO}_2$  as multiple recycling proceeds, so as to compensate for the progressive increase of the Plutonium quantities to be recycled and their isotopic degradation. In such a way, an equilibrium can indeed be reached where the discharged Plutonium remains identical to the loaded one, in quantity and quality. The same approach can be applied as well to the combined recycling of Pu+Am, recovered at each step.

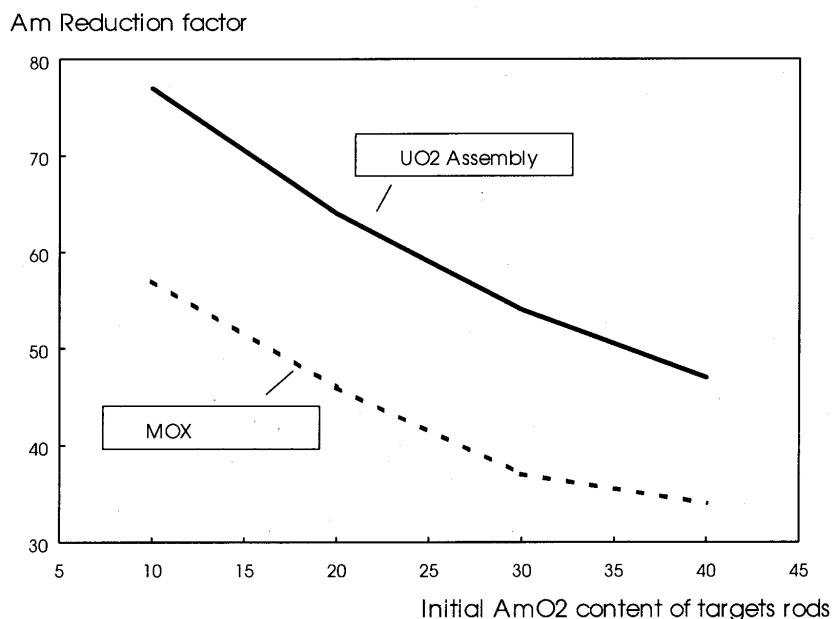
## Heterogeneous recycling of Americium: detailed results

### *A practical heterogeneous concept*

In the heterogeneous concept, Americium oxide is inserted in a matrix in the form of special target rods. The assumption is to mix  $\text{AmO}_2$  and  $\text{Al}_2\text{O}_3$  or  $\text{UO}_2$ ; other support materials are currently envisaged, and  $\text{Am}_2\text{O}_3$  could be envisaged as well, but as far as core physics is concerned, the present studies retain a sufficiently good orientation value.

Former core studies have shown that low fractions of 10 to 20%  $\text{AmO}_2$  in volume would be optimal to transmute Americium (see Figure 2), but here this parameter is allowed to vary in order to accommodate the growing Am quantities.

Figure 2. **Heterogeneous recycling of Americium in PWR** (mass reduction after 44 GWD/t)



Reference [5] has shown the merits of two promising concepts, in which the Am target pins are placed, either in peripheral core assemblies, or in the corner pin positions of each MOX assembly. These two concepts are equally possible solutions with acceptable core performances and Pu+Am incineration properties. However, the MOX corner pin positions appear to fuel manufacturers and to core designers most practical. The neutron flux is also twice as high as in the peripheral assemblies. Detailed calculations have been performed for the Am targets in MOX corner positions, and their results are displayed below.

On Figure 1 with the sketch of the MOX assembly, the 12 corner pins, usually containing Pu with a lower enrichment, are now replaced by Am target pins. This is a good buffer zone between the  $UO_2$  and MOX fuels so that the Am transmutation is relatively high, thanks to the thermalized spectrum of the  $UO_2$  zone, see Figure 2. The Pu enrichments of the 2 remaining medium- and high-content MOX zones need to be re-adapted. It is assumed that the Am target pins stay in the core as long as the MOX pins, i.e. 4 years, before being reprocessed. When Pu and Am are recycled after reprocessing of spent fuel (including the Am targets), the raise in Am quantities is coped with a raising fraction of Am in the target pins for the successive MOX-X recycling steps.

But again, when using depleted Uranium support in MOX, the Pu enrichments would soon exceed the 12% enrichment limit. The depleted U is, therefore, replaced at first step by natural U, and further on by enriched U, so as to keep the Pu enrichment fixed at 8%. The same type of progressively enriched  $UO_2$  support is used in the Am target pins.

### Multiple recycling results

This (Pu+Am) multiple recycling strategy has been explicitly modelled by calculations, using a macrocell representing the MOX assembly with its  $UO_2$  environment. After 15 recycles, the equilibrium is nearly reached for Pu and Am quantities : the unloaded amounts are equal to the loaded ones in the PWR system. The isotopic vectors, too, stay stable from any discharge to the next one.

Figure 3 illustrates the approach to equilibrium in the case where Pu alone is recycled, and Figure 4 when Pu and Am are simultaneously recycled. Tables I and II also show below, for the two cases, Pu only or Pu and Am, how are varying the fraction of MOX assemblies, the  $^{235}U$  enrichment of the  $UO_2$  support (it stays at 3.7% in the  $UO_2$  assemblies), and the volume fraction of Am in the targets.

Figure 3. **Multiple recycling of Pu in MOX fuel with enriched  $^{235}U$  support**  
Increase of a) U enrichment b) MOX fraction in core loading

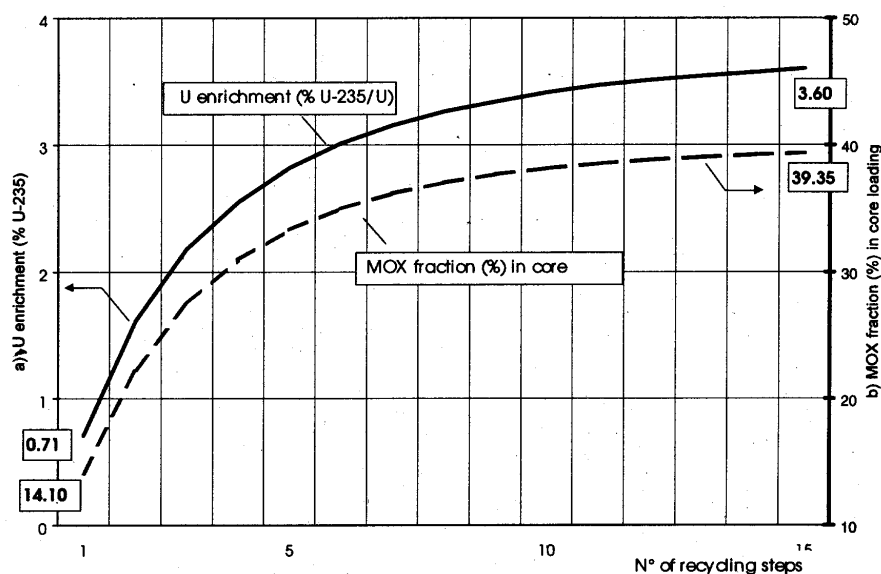


Figure 4. Multiple recycling of Pu and Am in targets with enriched  $^{235}\text{U}$  support  
 Increase of a) U enrichment b) MOX fraction in core loading c) AmO<sub>2</sub> fraction in targets

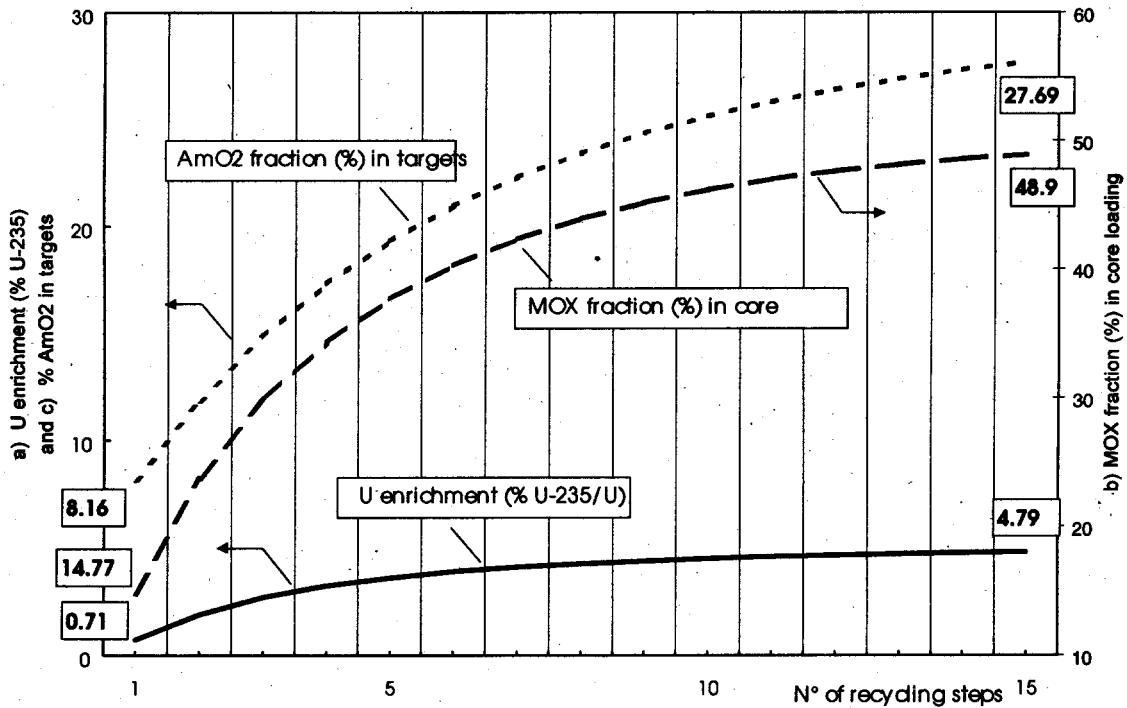


Table 1. Multiple Pu recycling using enriched UO<sub>2</sub> in MOX

Recycling steps	UO <sub>2</sub> enrichment in $^{235}\text{U}$ (%)	MOX fraction in core (%)
1	0.71	14.1
2	1.6	22.3
3	2.2	27.5
5	2.8	33.3
10	3.4	38.1
<b>15 (equil.)</b>	<b>3.6</b>	<b>39.4</b>

Table 2. Pu+Am recycling using enriched UO<sub>2</sub> in MOX and target pins

Recycling steps	UO <sub>2</sub> enrichment in $^{235}\text{U}$ (%)	MOX fraction in core (%)	Volume fraction of AmO <sub>2</sub> in targets (%)
1	0.71	14.8	8.2
2	1.9	23.7	11.9
3	2.7	29.8	14.9
5	3.6	37.7	19.3
10	4.5	46.2	25.1
<b>15 (equil.)</b>	<b>4.8</b>	<b>48.9</b>	<b>27.7</b>

The equilibrium between production and consumption of Plutonium and Americium needs:

- that about 50% of all assemblies be fuelled with MOX (vs 40% with Pu only).
- that the UO<sub>2</sub> support of the MOX assemblies be enriched to 4.8% in <sup>235</sup>U (versus 3.6% with Pu only).

The isotopic vectors at equilibrium are:

$$\begin{array}{l}
 {}^{238/239/240/241/242}\text{Pu} = 4.4/36.8/25.7/11.9/21.2 \% \text{ (recycling Pu only),} \\
 \text{or} \quad 11.1/34.9/23.3/10.9/19.8 \% \text{ (recycling Pu+Am),} \\
 \text{and} \quad {}^{241/242/243}\text{Am} = 56.9/0.3/42.8 \% \text{ (Pu only) or } 43.6/0.5/55.9 \text{ (Pu+Am).}
 \end{array}$$

Table 3 gives for the UO<sub>2</sub> once-through cycle, and for the Pu and Pu+Am equilibrium cycle, the quantities of heavy material to be loaded or discharged per year at equilibrium for a yearly electricity production of 1 TWhe.

**Table 3. Mass balances for three recycling strategies**  
(kg/yr corresponding to 1 TWhe)

Strategy	Once-through UO <sub>2</sub>	MOX Recycling Pu only	MOX Recycling Pu+Am
<sup>235</sup> U	460	429	480
<b>Pu</b>	130	363	432
<b>Am</b>	6.3	29.4	72.0
<b>Cm</b>	0.5	5.8	16.4

In Table III, the quantities in bold characters go to waste.

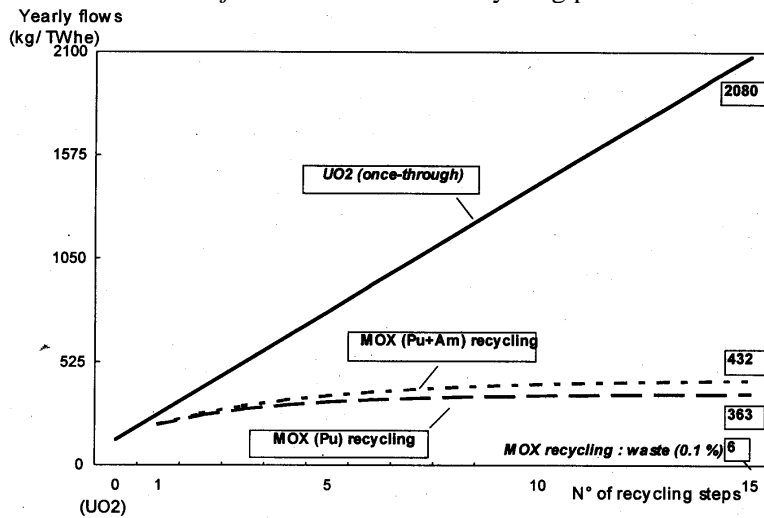
One can see the penalties incurred by recycling Am in addition to Pu: all mass flow rates have increased. In particular, the Cm masses are 3 times higher.

On the other hand, calculations have also checked that in these multiple recycling schemes, even when half of the assemblies are to be MOX ones, the power distributions can remain conform to the criteria throughout irradiation.

### ***Waste radiotoxicity reductions***

The success of these multiple recycling strategies in PWRs can easily be observed for what concerns the quantities of Pu and Am which are, either recycled and stabilised, or rejected as waste. As an example, Figure 5 shows that with the MOX (Pu) recycling strategy already, the success is significant with respect to the reference UO<sub>2</sub> once-through case. The MOX (Pu+Am) recycling strategy offers about the same type of success.

Figure 5. Accumulation of Pu quantities rejected as waste  
*rejected as waste as recycling proceeds*

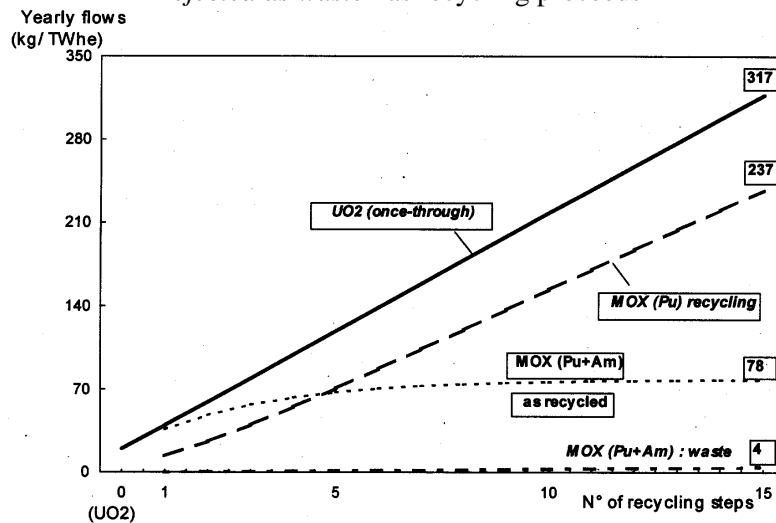


It is necessary to convert the quantities of the various Pu, Am and Cm isotopes into toxicities, to check what is effectively the reduction in terms of waste toxicities. One has of course to specify for what range of waste storage times one wants to reduce the toxicity.

Two typical storage times appear important: 1 000 and 10 000 years. For 1 000 years, the major sources of toxicity are known to come from the following isotopes:  $^{241}\text{Pu} + ^{241}\text{Am}$ , then  $^{240}\text{Pu}$ ,  $^{239}\text{Pu}$  and  $^{243}\text{Am}$ . In the once-through case, these isotopes contribute as follows to the waste toxicity:  $^{241}\text{Pu} + ^{241}\text{Am}$ : 65%;  $^{240}\text{Pu}$ : 20%;  $^{239}\text{Pu}$ : 13%;  $^{243}\text{Am}$ : 2%.

Figure 6 illustrates the relative effect of the MOX recycling strategies on the 1 000 year-toxicity by displaying the accumulation of the  $^{241}\text{Pu} + ^{241}\text{Am}$  quantities in the once-through case, and as recycling proceeds, in the two MOX (Pu, or Pu+Am) strategies. One can see that there is a remarkable success for the latter one, as could be easily predicted.

Figure 6. Accumulation of  $^{241}\text{Pu} + ^{241}\text{Am}$  quantities  
*rejected as waste as recycling proceeds*





The comparison after 10 000 years is less conclusive. Indeed, not only  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{243}\text{Am}$  are playing an important role in the toxicity, but also the raise in Cm quantities, as  $^{244}\text{Cm}$  rapidly decays into  $^{240}\text{Pu}$  and  $^{245}\text{Cm}$  has a half-life of 8 500 years, comparable to that of  $^{240}\text{Pu}$  (6 550 years).

Table 4 enables a judgement to be made on the compared quantities of  $^{239}\text{Pu} + ^{240}\text{Pu} + ^{243}\text{Am} + ^{244}\text{Cm} + ^{245}\text{Cm}$  rejected as waste as an indicator of the waste toxicity after 10 000 years.

**Table 4. Indications on waste toxicity after a storage time of 10 000 years  
[quantities of  $^{239+240}\text{Pu} + ^{243}\text{Am} + ^{244+245}\text{Cm}$  (kg) rejected as waste after 15 recycling steps]**

UO <sub>2</sub> once-through	MOX (Pu) multiple recycling	MOX (Pu+Am) multiple recycling
1 701	168	209

For such a storage time, the MOXX (Pu+Am) recycling strategy is obviously less good than the simpler MOX (Pu) one. The reason for this paradox is given by the larger accumulation of Curium quantities.

In conclusion, the success of the MOXX (Pu+Am) recycling can be qualified, with respect to the MOX (Pu) one, as follows :

- Largely favourable for waste storage times around 1000 years.
- Not quite as good for waste storage times around 10000 years.

### **Fuel fabrication aspects**

The homogeneous mixing of Am oxide into the usual (UO<sub>2</sub>PuO<sub>2</sub>) fuel was examined, with respect to the present fuel fabrication conditions at the BELGONUCLEAIRE MOX fuel plant [7]. Front-end operations, like the powder blending in the milling glove box, are most dose-intensive [8]. It was found that the combined recycling of Pu+Am corresponded to a substantial increase of the doses, by a factor 4.5. This can be coped with extra shielding layers, what complicates the fabrication and somewhat increases the cost of operations, but still seems a feasible extension of the standard MOX fabrication process.

In comparison, the fabrication of Am oxide target pins in a MOX fabrication plant in view either to feed the corners of MOX assemblies or to constitute dedicated assemblies, increases the doses by large factors, like typically 80 in front of the shielded doors of the oxide powder storage vaults, or even 2 800 from a canister transferring 200 pins to the mounting hall. This means that the fabrication of Am oxide targets should be done in shielded hot cells. More details can be found in [8].

### **Discussion and conclusions; further work**

#### ***Summary of the main observations***

Chemistry experts are presently requested to develop industrial ways to produce pure Am oxide from reprocessing. When this endeavour will materialise, the recycling of pure Am targets as considered above will be made possible, and the studies presented above will bear full significance.

From our orientation studies, both homogeneous and heterogeneous recycling schemes appear feasible. Multiple recycling of Pu+Am in PWRs is however possible only if enriched  $\text{UO}_2$  is used as support of the MOX fuel. The fabrication of target pins should be hosted in hot cells rather than in glove boxes, so that fabrication costs are likely to be higher; the cost increase cannot be quantified yet.

The efficiency of stabilising total Am quantities in the whole cycle, is comparable in both cases. Target pins placed at MOX corners or in dedicated assemblies are both viable. The solution of MOX corners, presented in details here, is most effective, as the neutron flux is higher and the spectrum softer, what favours Am transmutation.

It is shown that recycling Am in addition to Pu leads to significant penalties: one third more  $^{235}\text{U}$  is needed, the MOX fraction is increased from 40 to 50% in the PWRs, and there is a large raise of the Cm amounts. This problem of Cm deserves further studies.

While the multiple recycling of Pu+Am is effective to reduce the waste toxicity for storage times of about 1 000 years by a large factor (the MOXX (Pu+Am) strategy is 3 times better than the usual MOX strategy), there is no improvement but rather a slight deterioration for waste toxicities after 10 000 years. This is due to the enhanced production of Cm, by a factor 3, from Am irradiation.

### ***Modified schemes for Am recycling***

An alternate scheme which deserves to be investigated implies to keep the Am target pins 2 or 3 times longer in the reactor, so as to be entitled to reject these targets without further reprocessing. Obviously a certain part of the overcosts can be avoided in such a way. The balance is to be established, to check whether this prolonged irradiation can reveal globally beneficial. Parametric studies have already been launched, which indicate the potential interest of such a scheme, but full scale recycling calculations are still needed to confirm the advantages.

On the other hand, Am is not yet separated nowadays in an industrial scale from Cm and lanthanides at fuel reprocessing. These orientation studies have thus been complemented in order to know what would be the effect of accompanying Cm and lanthanides.

For a relatively small residual ratio of lanthanides on Am+Cm (e.g. 5 to 20%), these could be accepted as they do not penalise significantly core properties neither transmutation. The presence of Cm itself has a favourable influence on the power release of the targets, and therefore on their transmutation properties, which overcompensates the effect of lanthanides.

But the huge emission of alpha, gamma and neutron doses by Cm isotopes is known to be so severely hampering the handling of such powders that so far only gram quantities of Cm were treated, with the appropriate shielding. Hot cells are mandatory for any fabrication.

The fabrication of Am+Cm targets should thus be differed as long as realistically possible, e.g. by some 20 to 40 years, to let most of the Cm transform into Pu and ease this fabrication (anyway in automated cells of the fuel plant). The irradiation of these Am+Cm targets could then last longer than for the usual MOX pins, so as to be able to throw directly the spent targets to waste, avoiding any subsequent reprocessing.

Explicit recycling calculations are needed to verify how feasible are in such schemes the handling of powders before and during target fabrication, and the behaviour of the targets under irradiation ; finally, the residual masses of Am and Cm transferred to the waste are to be assessed.

One advantage of the heterogeneous option is obvious: it makes possible to manage in two different ways the Pu in MOX pins, and the targets. For example, the recycling of Pu could be done with the shortest cooling times possible (3 + 2 years) so as to burn quickly the highly fissile  $^{241}\text{Pu}$ , while the recycling of a mix of Am+Cm could be deferred to let Cm transform into Pu before irradiation. Also, the Am+Cm targets could be allowed to stay longer than the MOX pins, so as to avoid if possible further reprocessing.

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