IMPACT OF PLUTONIUM AND AMERICIUM RECYCLING IN PWR ON MOX FUEL FABRICATION

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Abstract

In a study with European partners for the Commission of the European Community, BELGONUCLEAIRE examines the implications of recovering americium at fuel reprocessing and recycling both plutonium and americium as MOX fuel in light water reactors.

The effect on the neutron physics of a 900-MWe PWR, operated by quarter-core reloadings up to a burnup of 45 GWd/t, with a quarter of the assemblies fuelled with MOX, is checked with respect to the reference strategy of recycling Pu only.

At the first recycling step with Pu and Am, both supposed recovered at reprocessing, a 10 % Pu content is already needed to guarantee the burnup : with Pu alone, this was 8 % ; the 10 % content level was necessary at the second recycling only. The raise in Pu content due to the addition of Am will severely limit the number of recycling steps allowable.

In the refabrication plant, the mixing of Pu with Am, as an extrapolation of the present MOX fabrication practice, will need some addition of shielding to the blending glove box ; this extra-shielding, while costly, seems nevertheless feasible and acceptable.

INTRODUCTION

In the frame of a joint study for the Commission of the European Community (CEC, DGXII), relative to strategic and technical aspects of nuclear waste transmutation [1], BELGONUCLEAIRE examines the implications of recycling plutonium and americium in the form of MOX fuel in light water reactors. First, the effects on reactor physics of such a strategy with multiple recycling steps are calculated, then the problems of MOX fuel refabrication are identified with reference to the present operation conditions of the MOX fuel fabrication plant of BELGONUCLEAIRE in Dessel, Belgium [2].

The hypotheses which have been assumed for a multiple recycling of homogeneous mixed-oxide fuels containing americium in addition to Pu will be given ; the multiple recycling of Pu alone is taken as a reference case in this study.

The physics aspects are first dealt with ; these aspects are deserving further verification in a close cooperation with ECN, Petten [3].

The paper is then centred on the MOX fuel fabrication aspects related to radiation dose protection in the MOX fuel plant. This part of the study offers some similarity with a preceding paper published in cooperation with EDF, which had considered that Pu and minor actinides could be recycled in fast reactors [4]. A major difference is that, while the former study had considered also the recycling of neptunium and curium, the present one is concentrated on the effect of adding americium only to plutonium, in a more realistic or shorter term approach, provided that chemistry research to recover americium proves successful.

HYPOTHESES FOR RECYCLING IN LWR

The reactor assumed for recycling is a 900-MWe PWR, characterized by a quarter-core fuel management with an average discharge burnup of 45 GWd/t. The MOX fuel is designed to be equivalent in energy to UO_2 fuel enriched to 3.7 % U235. At discharge all assemblies, loaded with UO_2 or with MOX, have been irradiated 4 calendar years at a load factor of 75 % in the average.

The lead times in the fuel cycle are four years for core irradiation, three years for cooling up to reprocessing, and two years for refabrication and transport before core reloading.

The multiple recycling strategy in the reference Pu core is as follows :

- in the first stage, MOX 1, 141 kg of MOX fuel are burnt in parallel to 859 kg of UO_2 fuel ; these 141 kg correspond to the mass of Pu which can be recovered from 1 tonne of UO_2 spent fuel from the initial PWR operation ; these 141 kg are a result from the calculations done, and they correspond well to the frequently quoted ratios of one MOX assembly for 7 initial UO_2 assemblies ;

- it is assumed that at the following reprocessing operation, the Pu quantities out of this one MOX assembly and of six UO_2 assemblies are mixed together : insofar, the Pu composition available for recycling is not the Pu from first MOX generation, but a blend ;

- on these premises, our calculations indicate that in the second stage, MOX 2, 167 kg of MOX fuel can be burnt together with 833 kg of UO_2 fuel : there is now one MOX assembly, with degraded Pu, to be blended with 5 spent UO_2 assemblies, and so on.

This is a simplified simulation of what could occur in reality in a whole reactor park, without considering the influence of lag times. A detailed representation of lag times, reactor availabilities and fuel cycle plant peculiarities has been made in the parallel CEA study [5].

SOME RESULTS OF PWR PHYSICS CALCULATIONS (Pu CORE)

From the core physics calculations, some salient results only will be presented, so as to make clear the approach followed. These results have been obtained using the WIMS code package with the data library WIMS-86, with cross-section improvements for the Am and Cm isotopes.

These results are to be considered as preliminary, because a second series of calculations is underway, in cooperation with ECN, Petten [3], using the most recent JEF2 cross-section data.

Three types of results are given below, concerning, respectively, the Pu enrichments needed, the quantities of Pu reprocessed and blended, and the isotopic composition of Pu available for refabrication. In fact, only the latter ones are directly used in the second part of the study. The latest time allowed for refabrication, 2 years after reprocessing, is assumed : this corresponds to a large build-up of Am^{241} in Pu.

TABLE I

MULTIPLE RECYCLING OF Pu IN LWR

MAIN PHYSICS RESULTS

Successive MOX Recycling Steps	(UO_2)	MOX 1	MOX 2	MOX 3
(Pu+Am) content in MOX fuel (%)	-	7.8	10.4	11.8
Quantities of Pu recovered for 1 tonne of heavy metal at latest refabrication time (kg)	11.0	17.4	22.2	25.9
Isotopic composition of Pu+Am for the next refabrication (%):				
Pu 238	2.3	3.0	3.6	
Pu 239	53.2	47.0	44.0	
Pu 240	24.7	28.2	30.2	
Pu 241	11.8	12.4	12.4	
Pu 242	6.8	7.9	8.5	
Am 241(*)	1.3	1.4	1.4	

(*) from the decay of Pu241 during refabrication time.

It can be seen that the required Pu enrichments grow significantly with the successive recycling steps. They lead to increasing difficulties to keep the MOX loadings compatible to the usual design and safety objectives, so that the third step, MOX 3, probably represents in practice an upper limit for this multiple recycling scheme.

COMBINED Pu+Am RECYCLING : CORE PHYSICS RESULTS

In this case, it is assumed that not only Pu, but also Am can be recovered at spent fuel reprocessing ; recycling in the form of MOX fuel (Pu + Am) is tempted in the same PWR under the same conditions. A recovery yield of 99.5 % for Pu and 98 % for Am has been assumed ; for such orientation studies, such high recovery yield values are representative enough.

The reasoning developed above for recycling Pu alone, is applied again but for the sum of Pu and Am, with unchanged burnup targets. The major results of core physics calculations follow in Table II.

TABLE II

MULTIPLE RECYCLING OF Pu + Am in LWR

MAIN PHYSICS RESULTS

Successive MOX Recycling Steps	(UO_2)	MOX 1	MOX 2	MOX 3
(Pu+Am) content in MOX fuel (%)				
Pu	-	9.8	13.6	16.6
Am	-	0.5	1.1	1.6
Quantities of Pu and Am (kg) for 1 tonne of heavy metal at latest refabrication time				
Pu	10.9	17.9	23.9	29.2
Am	0.6	1.4	2.3	3.0
Isotopic composition of Pu and of Am for the next refabrication (%)				
Pu : 238	2.3	3.5	4.7	
239	53.8	48.2	45.7	
240	25.1	28.4	30.2	
241	11.9	12.2	11.5	
242	6.8	7.7	8.0	
Am : 241	70.6	64.6	63.7	
242m	0.1	0.6	1.0	
243	29.3	34.8	35.3	

With respect to the enrichments needed to recycle Pu only, the enrichments required in this Pu+Am case are still higher. It is doubtful that the second step, MOX 2, will still be acceptable : an additional study, centred on the safety aspects, would be needed to resolve this question of limitation. The first step only, MOX 1 (PuAm) will further be considered.

CRITICAL STAGES IN THE MOX FABRICATION PLANT

In the MOX fuel fabrication plant of BELGONUCLEAIRE at Dessel [2], the front-end stages of the manufacturing process are the most critical ones in terms of radiation protection, since they correspond to handling operations on pure oxide powders.

Currently, PuO_2 powders are received from the reprocessors and first stored ; then they are introduced in a glove box to be milled and blended together with UO_2 powders to produce the so-called primary blend. These dose intensive operations are followed by secondary blending, pressing and sintering, before the sintered pellets are put into fuel pin claddings : dilution first, and canning afterwards, lower the dose rates.

The case of the powder storage had particularly been considered in the previous similar study [4]. Here, this is the case of the primary blending which is retained as the most typical. A sketch of the blending device, schematized for the sake of the calculation is given on Fig. 1.

Neutron and gamma dose rates at a distance of 30 cm from the external glove box wall have been calculated for a given configuration of blending devices (the UO_2 silo is not represented), glove box and shielding, so that the "reference" successive thicknesses surrounding the powders are, in cylindrical geometry :

a) 2 mm steel of the silo bottle ;

b) 100 mm of polythene acting as neutron shield, between two thin layers of 0.5 mm steel ;

c) 10 mm plexiglass, glove box wall ;

d) 36 mm of "KIOWAGLASS", a composite material including 30 % of lead.

Note that b) and d) are additional shields which are not present in the standard fabrication line.

For the primary blending operation, a maximum of 60 kg oxide can be handled, of which 30 % can be PuO_2 (containing a small amount of Am241). All calculations have therefore been run for this maximum quantity of 18 kg PuO_2, kept constant, but with a variable composition as given in Table I (Pu case) or Table II (Pu+Am case).

In the Pu+Am case, it is assumed that AmO_2 is added to these 18 kg of PuO_2.

The total AmO_2 content is for example 0.97 kg for the MOX 1 (PuAm) case. In the MOX 1 (Pu) case, it was 0.22 kg.

DOSE RATE CALCULATION RESULTS

The gamma source at fabrication time is calculated by simulating the successive steps (irradiation, cooling, reprocessing, and time shift up to refabrication), using the code ORIGEN 2 [6] with ad-hoc libraries. The losses at reprocessing are assumed to be 0.5 % for Pu and 2 % for Am.

The dose rates due to gamma rays are obtained using the standard gamma shielding computer code QAD-CG [7] for the 18 energy group spectrum coming from ORIGEN. Build up factors are fitted for the glove box materials.

For the neutron doses, the one dimensional transport programme ANISN-ORNL [8] has been employed with the EURLIB 15/5 cross-section library (15 neutron group and 5 secondary gamma ray ones).

Gamma and neutron fluxes are converted into equivalent dose rates using standard conversion factors.

The computing processes have been validated via OECD benchmarks for spent fuel transport and also by comparison with measurements on MOX sources.

Table III gives the dose rates at 30 cm from the glove box calculated as emanating from the reference PuO_2 powder (MOX 1) and from the variant powders corresponding either to a second Pu recycling step MOX 2 or to a first recycling of Pu and Am.

TABLE III

Impact on MOX Fuel Fabrication
at 30 cm from glove box of primary blending

Case	Dose rates (μSv/h)		
	Gamma	Neutrons	Total
MOX 1	9.7	7.3	17
MOX 2	9.7	9.1	18.8
PuAm	67.4	8.0	75.4
PuAm + 20 mm additional steel	17.5	5.8	23.3

Dose rates are expressed in μSv/h. In the reference MOX 1 case, the total dose rate of 17 μSv/h can be split into 7.2 (primary gamma-rays) + 7.3 (neutrons) + 2.5 (secondary gamma-rays). A value of 20 μSv/h (= 2 mrem/h) is taken as a guiding value for these comparisons, although it is not a real limit in the plant. Indeed, the staff will not stay longer than needed near the glove box of primary blending, according to the ALARA principle.

The results show that the conditions for the MOX 2 fuel fabrication are very similar to those of the MOX 1 fuel. The main neutron source via (α, n) reactions from Pu238, increased indeed marginally only. This is a favourable result of blending the Pu from MOX fuel with the Pu from UO_2 fuel.

On the other hand, the addition of americium at reprocessing, assuming a 2 years delay between PWR fuel reprocessing and refabrication, induces a strong increase of the gamma dose, so that the total dose becomes 4,5 times larger than for the reference MOX case. This would require an addition of steel shielding ; a layer of 25 mm steel is needed to reduce the total dose to a level similar to the reference case. (Table III also gives the calculation results for an addition of 20 mm).

DISCUSSION AND CONCLUSIONS

On the viewpoint of core physics, the effect of recovering americium in addition to plutonium at reprocessing, and of introducing it in the MOX fuel, is that at the first recycling step already a 10 % (Pu+Am) enrichment is needed. In the usual Pu recycling case, this is 7.8 % ; the 10 % enrichment level is needed at the second recycling step only.

Recycling Am in addition to Pu in the MOX fuel limits the number of recycling steps to one instead of two or three. Actually, more refined core calculations would be needed to identify clearly the limits for a safe core operation.

It is assumed that reprocessing takes place for spent MOX assemblies diluted into the bulk of spent UO_2 assemblies. In this way, the quality of the recovered plutonium, while progressively degraded, does not deteriorate too quickly.

This dilution explains why the dose rates at refabrication do not increase markedly with the recycling steps when Pu only is recycled : from the first to the second step (MOX 1 to MOX 2), the dose rates at the considered critical stage of fabrication increase slightly only.

In contrast, changing to a combined Pu + Am recycling strategy multiplies the dose rates by a factor 4.5. An additional steel shielding thickness (25 mm) is needed to bring the dose rates back to the same level.

This extra-shield hinders the fabrication and increases somewhat the cost of operations. Nevertheless, it seems feasible as an extrapolation of standard MOX fabrication conditions.

The justification of such extra-costs is to be found in the attainable reduction of radiotoxicity of the nuclear waste for long-term storage times (over 300 years) [9].

Penalties on refabrication are similar, whether Pu and Am have been recycled once in PWR, like assumed here, or recycled several times in fast reactors, like assumed in [4].

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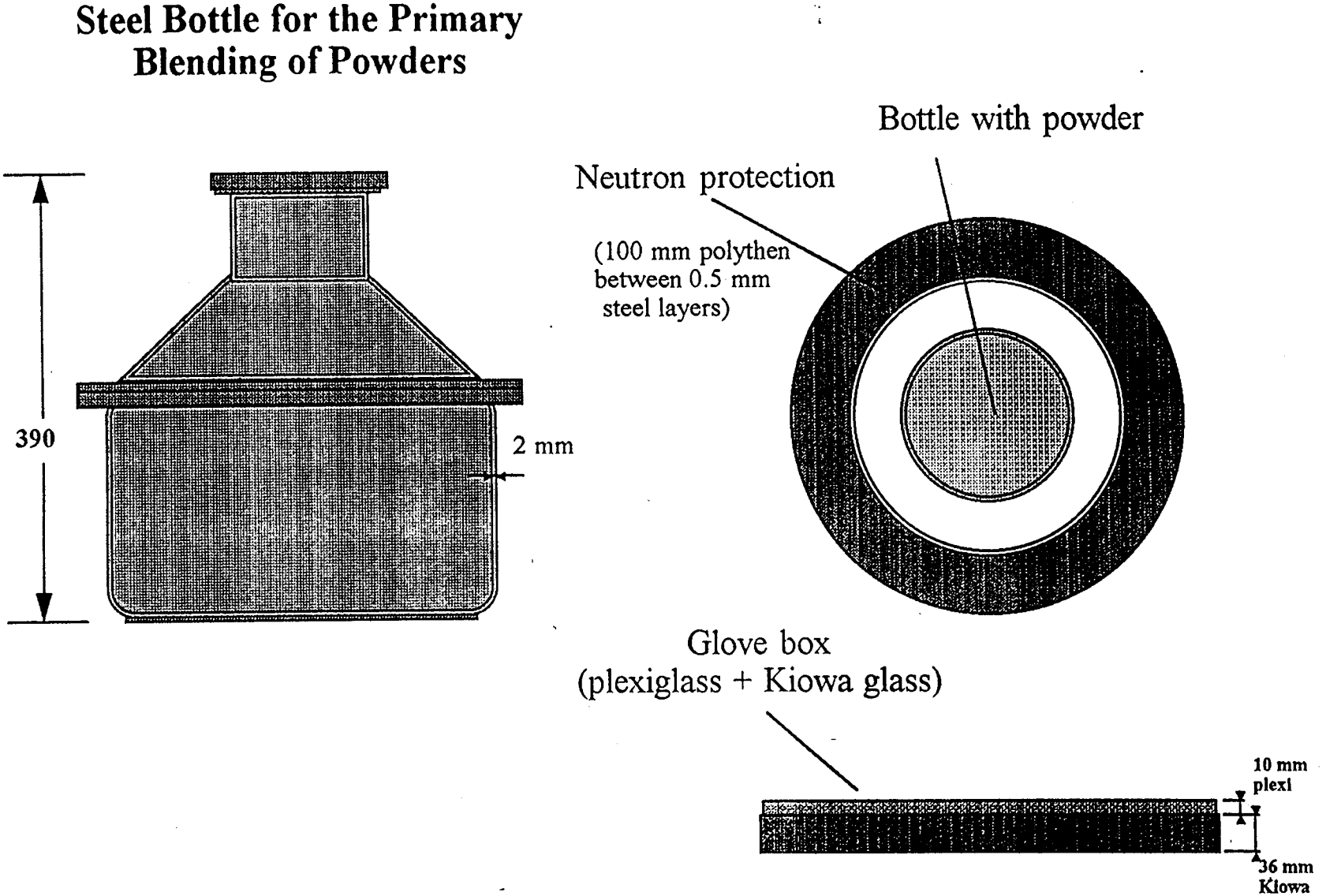

Fig. 1. Geometrical model for dose rate calculations with primary blending bottle