

CONSTRAINTS OF THE FABRICATION OF AMERICIUM OXIDE TARGETS

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Abstract

If a special fabrication chain is devoted to prepare AmO₂ target pins in a MOX fuel fabrication plant, the dose rates will be considerably increased with respect to the present MOX (UO₂, PuO₂) fuel pin fabrication practice.

This memo shows results of orientation calculations, as recently performed at BN, with the close follow-up by EDF.

The accent is stressed on the validation of the calculational route via the evaluation of measurements available.

1. INTRODUCTION

After plutonium, americium is the second most important transuranium radio-isotope for what concerns the long-term waste radiotoxicity. With its half-life of 433 years, americium 241 contributes to a large part of the activity and toxicity of waste after 1000 years, while its decay raises the content in Np237. Americium 243 is less important as its half-life is 7380 years.

Ways to reduce the amount of americium by transmutation via reactor irradiation, either in pressurised water reactors (PWR) or in fast reactors, are thus under investigation.

At BELGONUCLEAIRE, the merits of homogeneously spreading americium oxide, supposed recovered at reprocessing, into the mixed oxide (MOX) fuel, have first been identified. Core physics calculations were first run to define the range of Am mixing fractions and Pu enrichments which would be required for such fuels in PWRs. Then, the radioprotection implications of manufacturing these special fuels have been determined from an extrapolation of the fuel fabrication conditions in present MOX fuel plants like that of BELGONUCLEAIRE at Dessel (1).

The results of these investigations were published at the preceding OECD meeting in Cadarache (2) and later on in open conference (3).

The heterogeneous irradiation of Am in PWR is being considered in turn. Target fuel pins would contain AmO₂ with an inert material, like (Al₂ Mg O₄), transparent to neutrons. These pins could be inserted either in the bundle of MOX fuel assemblies, or in dedicated assemblies containing only target pins. A paper has presented solutions which seem acceptable from the point of view of reactor designers and operators (4) : they would allow to reduce the americium content by irradiation in a way which does not deteriorate reactor performances and safety.

What would be the burden of heterogeneous Am recycling on the MOX fuel fabrication plants ? This is the subject of current studies ; first results are presented in this paper. The accent is stressed on the validation of the results obtained.

ASSUMPTIONS ON OXIDE POWDERS AND TARGET PINS

The first assumption is that the MOX fuel fabrication plant would have a chain dedicated to AmO₂ fuel, in parallel to the usual PuO₂-UO₂ chain.

The front-end and back-end could stay in common, i.e. :

- it is imagined to receive AmO₂ powders from the reprocessors in the same packing and in similar storage vaults than for PuO₂-UO₂ at present,
- bundles of target fuel pins could be directed to assembly mounting halls, shipped in containers suited to on-site transportation.

In practice, for the sake of comparative calculations, the storage vault currently used has been supposed to contain pure AmO₂ powder instead of pure PuO₂ powder.

The incoming Am contains the following isotopic fractions : Am 241/242/243 = 62.4 % / 0.3 % / 37.3 %.

They are typical of americium fully recovered (with a 99 % yield) from the PWR fuel discharged after a burnup of 44 Gwd/t, 5 years after discharge. The isotopic Pu composition retained for comparison is :

Pu 238 / 239 / 240 / 241 / 242 / Am 241 = 2.4 / 53.4 / 23.6 / 12.3 / 7.1 / 1.2 %

The target pin is supposed to contain 20 % AmO₂ diluted with Al₂O₃. This relatively low AmO₂ content has been found optimal in view of a large reduction factor by irradiation, of 90 % for Am241 and 50 % for Am242 (4). The MOX pin retained as a reference for comparison contains 7.8 % Pu, i.e. an enrichment equivalent to 3.7 % U235 in PWRs, which becomes typical of MOX fuel nowadays.

The AmO₂ dedicated assembly to be constituted would consist, as the standard 900-MWe PWR assembly, of 264 active pins and 25 guide-tubes, arranged according to a 17 x 17 square lattice ; the active height is 3.66 m.

COMPUTER CODES USED

Three-dimensional dose-rate calculations have been performed using the codes QAD-CGGP (6) for gamma-rays and MCNP-4A (7) for neutrons. The radioactive sources have been established using ORIGEN-2 (8), taking into account irradiation history and the hypotheses given above.

PRELIMINARY COMPARATIVE RESULTS

- Storage of powders

Figure 1 shows a sketch of the storage vaults ; each vault can comprise a maximum of 5 cans containing each 3 kg oxide powders. The dose rates are calculated at 30 cm distance from the shielded doors.

Table I gives dose rates calculated for the presence of PuO₂ or AmO₂ powders, respectively, and their ratios.

For gamma-rays, the rise from PuO₂ to AmO₂ is by a factor 185 ; for neutrons this is by a factor 3.

To reduce the dose rates to the same levels needs the addition of 7 cm lead thickness to the shielded door.

TABLE I
Powder Storage Vaults
Dose rates at 30 cm distance from the door (mSv/h)

	Gamma dose	Neutron dose	Total
PuO ₂	0.015	0.019	0.034
AmO ₂	2.77	0.060	2.83
Ratio	185	3	82

- Handling and transportation of pins

A transfer of 200 pins is considered towards the assembly mounting hall, inside of a steel canister of a square section by 21 x 21 cm and a 5 mm thickness.

The IAEA rules for transportation of radioactive materials limit the dose rates to 2 mSv/h on surface and 0.1 mSv/h at 2 m distance. These limits have only an indicative value, as the considered transfer remains inside of the site, and is not a public road transportation.

Table II gives dose rates calculated at 1 m distance, for 200 pins either corresponding to the reference MOX conditions, or to AmO₂ target pins.

For gamma-rays, the rise from MOX to AmO₂ is by a factor 2780 ; for neutrons it is about 7. To reduce the dose rates to the same levels needs typically the addition of about 4 cm lead and 4 cm resin to the shielding, and to transport only half of the pins, i.e. 100, at the same time.

TABLE II
Transfer Canister for Pin Bundles
Dose rates at 1 m distance (mSv/h)

	Gamma dose	Neutron dose	Total
MOX pins	0.097	0.135	0.232
AmO ₂ target pins	270	0.94	271
ratio	2.780	7	1170

VALIDATION OF THE COMPARISONS

Much work has been focused on the validation of the results, especially because the extrapolation to be done from the present MOX fuel fabrication experience is huge.

Some aspects of this validation are detailed hereafter.

1. Dose from storage of MOX fuel

The dose rate has been measured at BELGONUCLEAIRE Dessel in front of the door of a storage vault containing 5 PuO₂ powder cans (double boxes), and compared to calculations.

The measurement was made at 63 mm distance from the external face of the door, which is made of sandwich layers of stainless steel, lead, boron containing glass, polythene and cadmium, see Figure 1.

Each of the 5 cans contained about 3 kg of PuO₂, with the following isotope composition :

Pu238 / 239 / 240 / 241 / 242 / Am241 = 1.32 / 57.38 / 24.15 / 6.11 / 5.11 / 5.93 %.

This measurement was selected for calibration of the methods, as it refers to an aged plutonium with as much as nearly 6 % Am241 / Pu.

The comparison between calculation and measurement is given in Table III below.

TABLE III
Calibration of Methods
1. Storage of aged PuO₂

Dose Rates (μSv/h)	Calculated	Measured
from gamma-rays	22.5	23*
from neutrons	33	29

* 23 : is the average of 5 measurements ; the extreme values were 23 ± 7 ; background was discounted.

The differences between calculation and measurement are quite acceptable.

Similar measurements were also made with the door open ; the differences (C-E)/E were about 20 % for gamma-rays, what is also acceptable. For neutrons, the calculations overestimate the dose as above.

Note that with this type of Pu, Am241 contributed to about 50 % of the gamma dose.

2. Dose from a transport container

The case of the transport of 2 fresh MOX assemblies into a specific container with 2 positions has been considered. Results of measurements are available for 3 m-long MOX assemblies of 14 x 14 pins, with an average Pu enrichment of 5.3 %

A double steel wall, 4 cm-thick in total, comprises a 4-cm thick resin layer.

Table IV gives the comparison of calculated and measured dose rates on the surface of the container.

TABLE IV
Calibration of Methods
2. Transport container (MOX)

Dose Rates ($\mu\text{Sv/h}$)	Calculated	Measured
from gamma-rays	28.8	32
from neutrons	241	208

The comparison is satisfactory. The neutron calculation, by Monte-Carlo method with a statistical uncertainty of 3 %, overestimates the measurement by 15 % ; fluctuations of the resin composition could explain this deviation.

3. Dose from a SUPERFACT pin

The irradiation experiment SUPERFACT has been conducted in the fast reactor PHENIX, up to a burnup of 5 %. The capsule contained in particular special pins loaded with americium, which had been fabricated at TUI Karlsruhe, see (5).

Pin SF No.14 contained mixed oxide, the metal being 20 % Am241, 20 % Np237 and 60 % depleted uranium. The radial pin dimensions were typical of PHENIX : 5.42 mm pellet o.d., 6.55 mm cladding o.d.

For this fresh pin, dose rates have been measured at TUI. The measurement retained for this calibration is at a distance of 1 m from the pin, without any additional shielding. According to TUI (5), their calculation reproduces well the measurement.

Table V below compares two sets of calculation results, the one from TUI (5), and the other one with the BELGONUCLEAIRE methods, described above.

Both calculated routes have been applied in parallel to a standard PHENIX pin, of same radial geometry, containing mixed oxide $\text{UO}_2\text{-PuO}_2$ with a Pu enrichment of 25 % and a Pu isotopic composition of :

Pu238 / 239 / 240 / 241 / 242 / Am241 : 1.3 / 60.4 / 23.4 / 9.9 / 4.5 / 0.5 %

TABLE V
 Calibration of Methods
 3. SUPERFACT pin with Am

Dose Rates (mSv/h)	Calculated at TUI	Recalculated
SUPERFACT pin 14 :		
- from gamma-rays :	0.25 ¹⁾	0.33
- from neutrons :	- ²⁾	0.00057
Standard PHENIX pin :		
- from gamma-rays :	0.0065	0.0068
- from neutrons :	0.00018	0.00028

¹⁾ measured : 0.26

²⁾ no measurement quoted

For the SF14 pin, Table V shows an overestimate of about 30 % for the gamma-ray dose. While our calculations are showed to be conservative, the difference could be associated with the gamma source spectrum found with ORIGEN2 (TUI used a source from the KFK improved version KORIGEN).

For neutrons, the values for the PHENIX pin shows a difference by about +50 % ; the dose due to neutrons is anyhow much smaller than the gamma dose.

From the PHENIX pin to the SF14 pin, the gamma dose increases by a factor 40 to 50, and the neutron dose by a factor 2.

These cross-checks may be considered satisfactory for the sake of orientation calculations.

PROVISIONAL CONCLUSIONS

The fabrication of AmO₂ target pins in a MOX fabrication plant indicate that, with respect to the dose rates experienced in present MOX (UO₂ PuO₂) fuel fabrication, the dose rates emanating from AmO₂ powders, from target AmO₂ pins and from bundles of such pins, would be significantly higher if no re-inforced shielding was applied. The raise of the dose rates would reach factors like :

- 80 in front of the shielded doors of the oxide powder storage vaults
- 50 from a single Am target pin compared to a standard fast reactor pin geometry,
- 2800 from a canister transferring 200 pins to the mounting hall.

The increase in shielding thickness necessary to lower dose rates to the levels currently observed in MOX fabrication, has been determined by calculation.

It appears that the fabrication of AmO₂ target pins should preferably be done in shielded hot cells.

As the americium mass recoverable from PWR irradiated fuel (after 5 years) is about 6 % of the plutonium mass, and considering the assumption made here (MOX pins with 7.8 % Pu, target pins with 20 % AmO₂), the production of Am target pins would amount to about 1/50 of the MOX pins.

The transportation of dedicated AmO₂ assemblies obviously needs a re-inforced shielding on the containers. The practicability of such transportation is being studied in further details.

A large effort has been spent to check the validity of the calculations. To that aim, 3 important steps of the fabrication chain have been covered by experimental validation :

- the storage of incoming powders for which measurements are obtained from BN/Dessel for relatively aged PuO₂ batches,
- the transport container, for which measurements are obtained for current MOX assemblies,
- and the constitution of single pins containing 20 % AmO₂, for which TUI Karlsruhe had measured and calculated dose rates.

The comparisons made bring confidence in the validity of the predictions.

REFERENCES

- [1] D. HAAS, A. VANDERGHEYNST & J. van VLIET (BELGONUCLEAIRE)
R. LORENZELLI & J.L. NIGON (COGEMA)
Mixed-Oxide Fuel Fabrication Technology and Experience at the BELGONUCLEAIRE and CFCa
Plants and Further Developments for the MELOX Plant
Nuclear Technology, Vol. 106 (April 1994)
- [2] T. MALDAGUE, S. PILATE, A.F. RENARD
Impact of Plutonium and Americium Recycling in PWR on MOX Fuel Fabrication
Proc. of the 3rd International Information Exchange Meeting on Actinide and Fission Product
Partitioning and Transmutation, Cadarache, France, December 12-14, 1994.
OECD/NEA
- [3] A. RENARD, S. PILATE, T. MALDAGUE, A. LA FUENTE, G. EVRARD
Implications of Plutonium and Americium Recycling on MOX Fuel Fabrication
GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems,
Versailles, France (September 1995).
- [4] T. MALDAGUE, S. PILATE, A. RENARD (BELGONUCLEAIRE)
A. HARISLUR, H. MOUNEY, M. ROME (ELECTRICITE DE FRANCE)
Core Physics Aspects and Possible Loadings for Actinide Recycling in Light Water Reactors
GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems,
Versailles, France (September 1995).
- [5] G. NICOLAOU & al.
Experimental Nuclear Data in Relation to Irradiation Experiments of Minor Actinide Targets in Fast
Reactors
GLOBAL'95, International Conference on Evaluation of Emerging Nuclear Fuel Cycle Systems,
Versailles, France (September 1995).
- [6] QAD-CGGP, a Combinatorial Geometry Version of QAD-P5A, a Point Kernel Code System
for Neutron and Gamma-Ray Shielding Calculations using the GP Buildup Factor
Contributed by JAERI and ORNL - CCC-493 (1987)
- [7] J. BRIESMEISTER (Editor)
MCNP, a General Monte Carlo Code for Neutron and Photon Transport
LA-7396-M (September 1986, revised April 1991)
- [8] A.G. CROFF
A User's Manual for the ORIGEN2 Computer Code
ORNL/TM 7175 (July 1980)

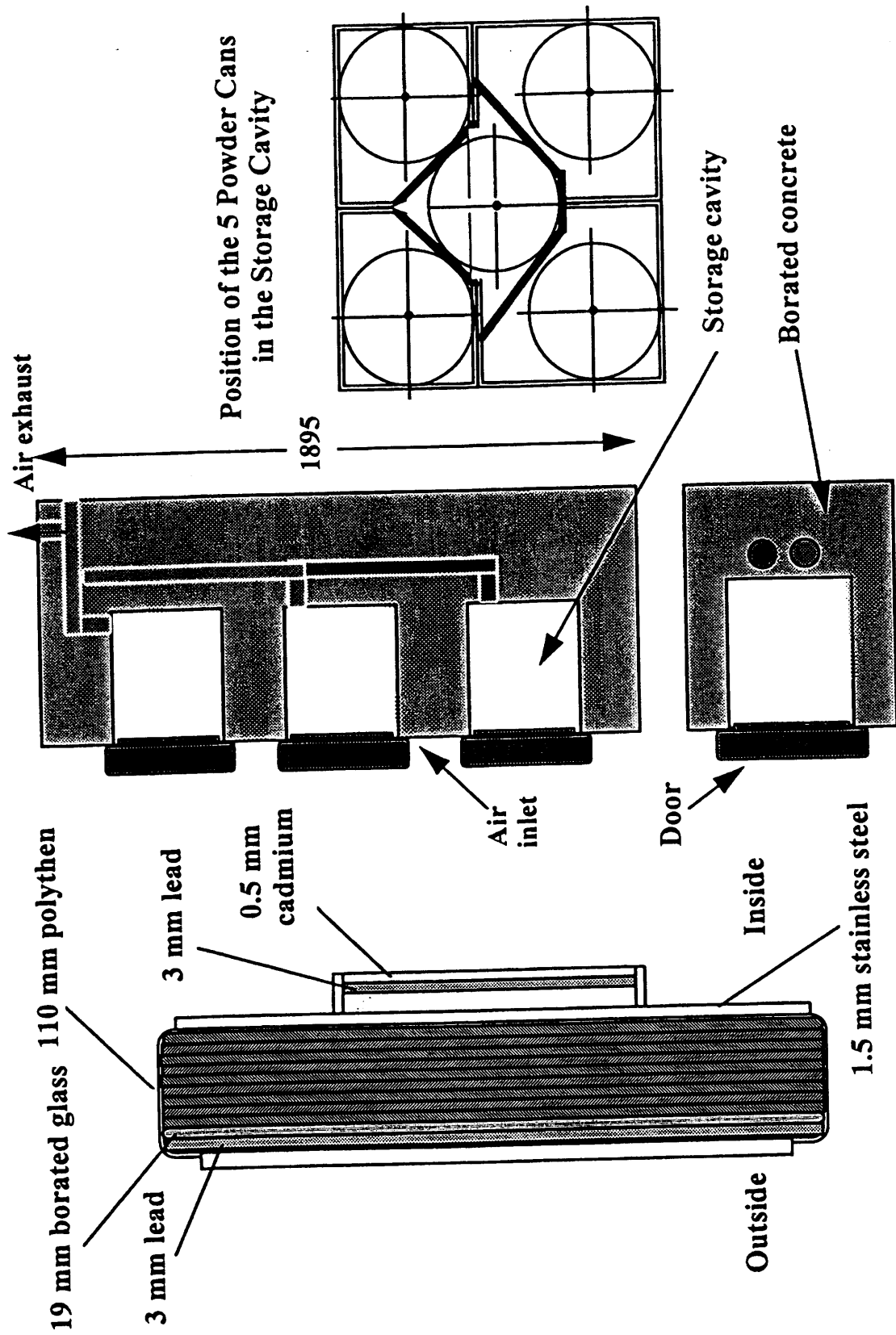


Fig. 1. Plutonium Storage Rack

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