Americium Incineration by Recycling in Target Rods using Coated Particles

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Abstract *– This paper proposes a type of target rod based on the use of coated particles, for an efficient incineration of americium in nuclear reactors. The analysis takes advantage of the experience gained in the past from long duration irradiation without damage of coated particles with plutonium oxide kernels. A conservative theoretical evaluation of the gas pressure inside the coated particles at the end of irradiation allows comparing the well known conditions of the plutonium oxide particles which were successfully irradiated to high burn-up, with a preliminary design of americium oxide particles.*

INTRODUCTION

Amongst the minor actinides that are responsible of the potential radio-toxicity of spent fuel, americium deserves special consideration, in particular for a waste storage time of about 1000 years. Today, heterogeneous recycling in specific target rods to be located inside reactor cores can be considered as technologically feasible at mid-term [1].

Recycling of plutonium and americium in PWR stabilizes progressively the global inventory of actinides resulting from the reactor operations and so reduces the mass of waste (and the associated radio-toxicity) in comparison with the UO_2 once-through option. But, the minor actinide burnt fraction is not larger than 50 %, even in the best conditions, and the reduction of waste radio-toxicity after some 10000 years storage amounts only to a factor 8 in comparison with the UO_2 once-through case [2]. These studies were initially performed in collaboration with EDF/SEPTEN.

As the final aim of the incineration in target rods is to obtain a destruction efficiency of 90 % by fission of the americium atoms, a residence time of the targets during about 10 years in a fast reactor must be considered.

TOWARDS THE CONCEPT OF TARGETS USING COATED PARTICLES

Candidate targets with americium distributed inside an inert matrix were already tested, but only up to a limited burn-up, because considerable swelling occurred, due to the accumulation of helium gas bubbles produced by alpha decay of the created higher isotopes.

For instance, the EFTTRA-T4 experiment [3] performed in the High Flux Reactor at Petten (NL) on pellets with 12 % americium distributed inside an inert matrix ($MgAl_2O_4$), was successful up to a 28 % fission level reached after 358 full power days. Afterwards, a considerable swelling was noticed and attributed to the accumulation of helium gas bubbles produced by the alpha decay of curium created by neutron capture.

It was deduced that a dispersion of spherical particles with optimal size between 50 and 300 microns would minimize damage to the inert matrix; moreover, the target rods should contain a significant amount of porosity or/and void fraction. To cope with helium and fission gases containment, the idea arose of applying to the target rods the coated fuel particles concept, as used in High Temperature Gas Reactors.

A coated particle includes a central fuel kernel surrounded by several layers, first a porous pyrocarbon (PyC) buffer layer and then the pressure vessel generally constituted of three dense coating layers, wherefrom the name of TRISO particle. The porous buffer layer of the coating provides void volume to contain the gases produced during fuel operation. The dilution of the fissile oxide of the kernel into porous carbon plays a similar role.

A calculation model of the internal pressure inside the particles at the end of a very long irradiation period was developed, assuming a complete release of the helium and of the gaseous fission products, as well as the production of CO due to oxygen excess resulting from fission. The helium build-up during the long residence time necessary to reach a very high burn-up in reactor is calculated by a properly benchmarked nuclear code.

The feasibility study of a coated particle option requires a resistance criterion linked to the high pressure level. This criterion was obtained by application of the calculation model to previous experimental tests performed on TRISO coated plutonium particles up to very high burn-up (e.g.~600 000 MWd/tM), which showed an excellent behavior.

THE COATED PLUTONIUM OXIDE PARTICLES

Over thirty years ago, all the steps of the fabrication process for coated plutonium particles were established and the behavior under irradiation was tested and analyzed for several types of particles [4].

In particular, significant tests on such plutonium fuels with diluted kernels were performed during five years in the experimental reactor of the international DRAGON project. The TRISO particles had been produced in a pilot fabrication unit, under co-operation between Belgonucleaire and the Belgian Nuclear Research Center.

A powder agglomeration process had been used to manufacture the kernels. Fig.1 shows a ceramography of a diluted kernel containing about 120 µg Pu.

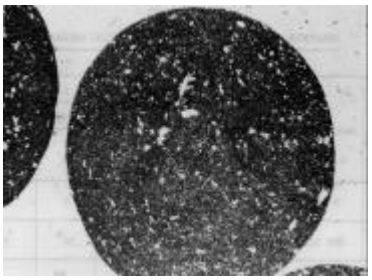

Fig. 1 Diluted Pu oxide kernel (x200)

The R & D program demonstrated that such kernels can also be produced by a sol-gel process.

The fuel irradiated in the DRAGON reactor successfully achieved burn-ups as high as roughly 600 000 MWd/tM. Fig 2 shows an example of ceramographic examinations of those particles containing about 15 µg Pu each, confirming the absence of damage after irradiation.

FRESH

BURNT

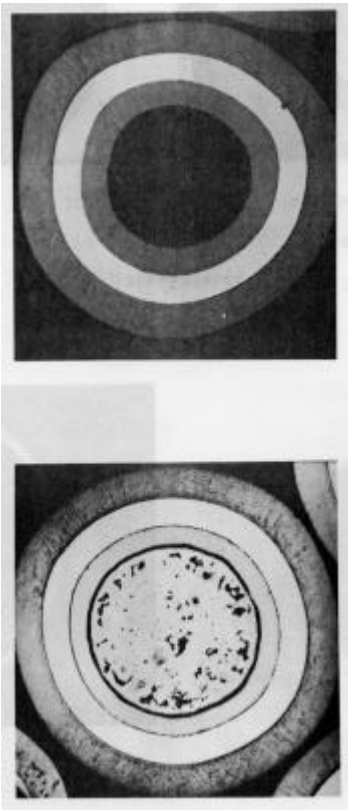

Fig.2 Fresh and highly irradiated coated Pu particles (600 000 MWd/tM)

Other tests were conducted in the early 70's in the German test reactor FRJ2 to compare coated plutonium particles with different carbon dilution levels in the kernel down to zero. For the pure Pu oxide kernels, the missing void volume was compensated by increasing the buffer layer thickness.

The Pu content per particle was several times higher than in the DRAGON fuel and the kernels were somewhat bigger, but the behavior up to a final burn-up of 180 000 MWd/tM was also successful. Fig.3 is a cross section of one of

these irradiated particles (enlarged 80 times) showing some fragmentation of the kernel without damage for the coating.

Fig.3 Particle irradiated in FRJ2 reactor (~120 µg Pu/particle, 180 GWd/tM)

The tests demonstrated the superiority of the diluted kernel over the pure oxide kernel concept.

Table 1 compares typical values of the main parameters for both examples of diluted kernel Pu particles. The pressure evaluation is based on the gas production (obtained by means of the ORIGEN2 code and validated by a successful comparison with the gaseous inventories in EFTTRA-T4 test) and on the ideal gas law.

TABLE I. Main Features of Coated Pu Particles

Tested in reactor :	Dragon	FRJ2
Pu mass in kernel (µg)	15	120
Burn-up (GWd/tM)	600	180
Initial diluted kernel diam. (µm)	300	550
Initial porosity in buffer (%)	50	50
Coated particle diameter (µm)	640	890
Average temperature (°C)	1200	1300
Evaluated inner pressure (MPa)	37	10

Table 1 shows that the coated Pu particles withstood internal pressures of several tens of bars without failure, up to 37 MPa according to the conservative calculation assumptions.

FEASIBILITY OF AMERICIUM TARGETS

One important contributor to the gaseous source inside the particle is the helium atoms accumulated up to the end of the irradiation. For the same consumption rate of actinides in a fast reactor spectrum, the americium oxide particles would contain about 2000 times more He than the Pu particles, essentially due to the build-up of curium isotopes.

The pressure level in the particles at the end of the irradiation is used here as the criterion to select the features of Am particles proposed for the target rods, in comparison with the Pu fuel particles successfully irradiated in the past.

One assumes that the affinity of the solid fission products for oxygen is the same for both plutonium oxide and americium oxide cases. The oxygen excess released at the fission and not captured by the fission products first converts the under-stoechiometric fraction of the fabricated Pu oxide (i.e. Pu_2O_3) into PuO_2. Then, the residual oxygen excess reacts with the diluting carbon and/or the porous PyC layer creating an additional partial pressure of CO. The fabrication conditions of americium oxide can be adapted in order to favor the formation of Am_2O_3 and to reduce the amount of CO created during the irradiation.

On the other hand, the operating temperature of the Am target rods will be quite lower than that one of HTR Pu fuel because the average power rating in targets will be much lower than in the tested coated Pu oxide particles.

Some recommendations were deduced from the tests and studies on coated Pu particles. They are used here as “design” criteria:

- Kernel diameter between 300 and 500 µm,
- 30 % initial porosity in the kernel,
- TRISO coating layer thickness > 85 µm,
- Maximum inner pressure = 37 MPa.

The feasibility study focuses on americium target rods for a fast reactor similar to Superphenix (SPX), i.e. one meter active length and a fuel rod diameter of 7.14 mm (in core) or 13 mm (in reflector). The mass of americium per target rod depends on those dimensions and on the maximum amount of Am per coated particle taking account of the “design” criteria.

Table II presents the effects of the variation of the following parameters:
TD = target diameter (mm),
MT = Am mass per target (g),
MK = Am mass per particle kernel (µg),
BU = tentative burn-up (GWd/tM),
KD = initial diluted kernel diameter (µm),
PL = initial 50% porous layer thickness (µm),
TL = initial TRISO layers thickness (µm),
PD = coated particle diameter (µm),
OT = average operating temperature (°C),
on the evaluated inner gas pressure at the end of irradiation = GP, for 6 different parameter combinations. The italic bold values correspond to the modified parameters in each case, in comparison with the reference case. The variation of mass and geometrical parameters illustrates the sensitivity of the internal pressure to these variables.

TABLE II. Coated Am Particle Types

Case / **Parameter**	1 (Ref.)	2	3	4	5	6
TD (mm)	13	***7.14***	13	13	13	13
MT (g)	25	***7***	***15***	***15***	***15***	***15***
MK (µg)	30	***30***	***20***	***20***	***20***	***20***
BU (GWd/tM)	880	880	880	880	***600***	880
KD (µm)	354	354	310	310	310	310
PL (µm)	73	73	***38***	***38***	***38***	***83***
TL (µm)	87	***95***	***157***	***157***	***157***	***112***
PD (µm)	674	690	700	700	700	700
OT (°C)	727	727	727	***927***	727	727
GP (MPa)	**37**	**37**	**59**	**71**	**23**	**27**

The “basic” set of parameters (Reference case 1) corresponds approximately to targets to be put at the periphery of a fast reactor core similar to SPX. This case provides at end-of-irradiation a gas pressure level similar to the plutonium particles tested to very high burn-up. The same pressure value is obtained in case 2 dealing with a target of smaller diameter containing a particle type very similar to case 1, with the same volumetric loading of particles in the target.

Case 3 shows the influence of a relative reduction of the porous coating layer thickness and case 4 includes the additional effect of an increase in the operating temperature. Both effects introduce a significant increase of the final pressure inside the coated particles. It could theoretically be contained by a larger thickness of the TRISO layers, but the available experimental background does not support this assumption.

If the burn-up were reduced by 30 % (case 5), keeping the “basic” operating temperature of the Am targets, the pressure would be 37 % below the defined limit. Similar pressure reductions can be obtained for the target final high burn-up if the thickness of the porous layer is increased to the detriment of the TRISO layer (case 6).

A compromise between the Am amount to be reasonably introduced in targets, the target dimensions, the mass of Am per particle and the necessary coating size parameters seems feasible in order to limit the final gas pressure in the particles to a value demonstrated in the past experience. One must keep in mind that this pressure is an upper limit being evaluated by a conservative model.

Additionally, gaseous products from a possible failure of some particles would be partially retained in the matrix embedding the coated particles. Moreover, in a fast reactor, the target would be inserted in a cladding and the gases escaping the target matrix would collect in the gas plenum of the rod. Provided the gas plenum is adequately designed, it would only induce a small increase of the pressure in the target rod to a level admissible for reactor operation and final disposal of the rod..

CONCLUSION

This feasibility study of americium target rods using coated particles points out the need of a diluted kernel with a limited americium content and a strong TRISO coating.

Acceptable designs of particles can be evaluated as a function of their americium content by reference to plutonium particles having reached a high burn-up level without coating damage in the past.

On these bases, about 90 % fission in the americium particles can be obtained, if adequate parameters are fixed for the structure and the americium content of the particles. But this design approach needs confirmation from additional research and development as well as from demonstration tests.

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