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Americium and Curium heterogeneous transmutation in moderated S/A in the Framework of CNE Scenarios studies

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Abstract

This paper presents the transmutation of Americium and Curium in a heterogeneous mode in the framework of the 1991 French Law concerning waste management. Two scenarios with moderated targets are presented : a 100% fast reactor (EFR) scenario multirecycling Pu+Np with targets of Am+Cm placed in core and a mixed PWR (UOX fuel) and fast reactor (50% of EFR) multireycling Pu+Np and containing targets in core and in the blanket region. The design of the target is based on classical fast fuel S/A technology (pins, spacer wires,...) and should reach the goal of 90% fission rate.

1 Introduction

The introduction of moderated sub-assemblies in a fast reactor core is one of the most promising options to transmute Minors Actinides. The idea is to combine the high cross sections existing in the thermal and epithermal energy range to the high neutron flux level found in a fast reactor [1]. Furthermore, this strategy for americium allows in situ curium management by burning part of the curium produced by americium transmutation. Targets are moderated with Zirconium hydride and are optimized to achieve an objective of 90% fission rate within various technical limitations.

Several target positions were studied and for each of them, a choice of calculation scheme was made to be as close as possible to reality.

The optimization will be separated in two phases : first, simplified calculations to establish first results and then heterogeneous complete calculation on specified case using previous specifications. These last complete calculations will correspond to nuclear scenarios in which nuclear waste are managed.

2 Basic physics of moderated targets and the corresponding calculation scheme

The calculation of this kind of target may involve several problems for deterministic codes. We are dealing with a sub-critical moderated medium where the whole neutron energy range is covered. Moreover, the major source of neutrons is made of fast neutrons coming from the core and being thermalized in the target. Therefore, the calculation routes are the following one :

- simplified calculation :
  - preparation of cross sections with the ECCO cell code [2] which allows the original geometry description of the S/A, the use of an external source as neutron feeder to represent the current coming from the core, a fine energy group calculation (1968 energy groups for the major isotopes) for the explicite treatement of resonances and good treatement of low ξ materials, self-shielding calculation and up-scattering calculation
  - use of the previous cross sections condensed in 33 energy groups for the core calculation with a R-Z geometry (cylindrization of the core) using the finite difference type code BISTRO. Two target positions can be calculated : in the center of the core or in the first blanket region.

- use of the previous cross sections condensed in 33 energy groups for entire core calculation made with ERANOS [3]. Targets placed in the blanket region well described by a R-Z cylindrized geometry will be calculated in Transport theory with the finite difference BISTRO module and targets replacing fuel S/A better described with the original hexagonal geometry are calculated using the nodal VARIANT module of ERANOS, with a diffusion approximation.

The simplified calculation route will be used for the optimization and the complete one for the application to scenarios.

3 Optimization scheme

3.1 Philosophy

The strategy to optimize our targets is based on the following consideration : how to get the highest fission rate and highest mass consumption possible without going above technological limits ? A 90% fission rate is at least the minimum rate that we can ask for a once-through strategy. The studied technological properties are the following ones :

- damage dose on cladding and hexagonal tube : this limit is fixed at 200 dpa (NRT),
- temperature in the actinides pins must be lower to the fusion temperature of AmO_2 which is $2173°C$,
- the clad thickness must maintain the mecanical integrity of the pins with regards to gaz production (Helium and gazeous fission products),
- pressure drop should be coherent with the standard fast reactor values.

To understand all occuring problems, three kind of S/A will be here studied :

- the first one called theoretical design will show the highest mass comsumption possible without realistic design consideration (see figure 1),
- the second one based on a standard fast fuel S/A with two kinds of pins (moderator pins and actinide pins) with the same radius, withno specific technological considerations added,
- the third one is similar to the previous one except that pins can have different radii. The figure 2 is an example of such designs. Furthermore, new concerns are added on moderator and fuel cladding, radius of spacer wires, pellet-cladding interactions for the fuel. This kind of design is supposed more realistic.

3.2 Basic design assumptions

3.2.1 Target Contents

The americium pins are made of actinide oxyde mixed with MgO, an inert matrix. Two volumic proportions of $(Am+Cm)O_2$ were used : 13 and 17% (a 15% case was added for the third design).

The actinide isotopic composition is fixed for the optimization procedure (see tab 1).

3.2.2 Clad thickness

For the first and third kind of targets, some assumptions are made concerning clad thickness : if r_{pellet} is the radius of the actinides pellets, the inner radius of the clad is given by :

$$r_{clad}^{in} = r_{pellet} \cdot 1.07$$

to allow the irradiation-induced swelling of the fuel. Furthermore, the external radius is taken to be :

$$r_{clad}^{ext} = r_{pellet} \cdot 1.07 \cdot 1.15$$

These values are used for the neutronic optimization, for the final design, a real clad thickness calculation will be performed.
For the second design the external radius of the pins is fixed to 0.3175 cm.

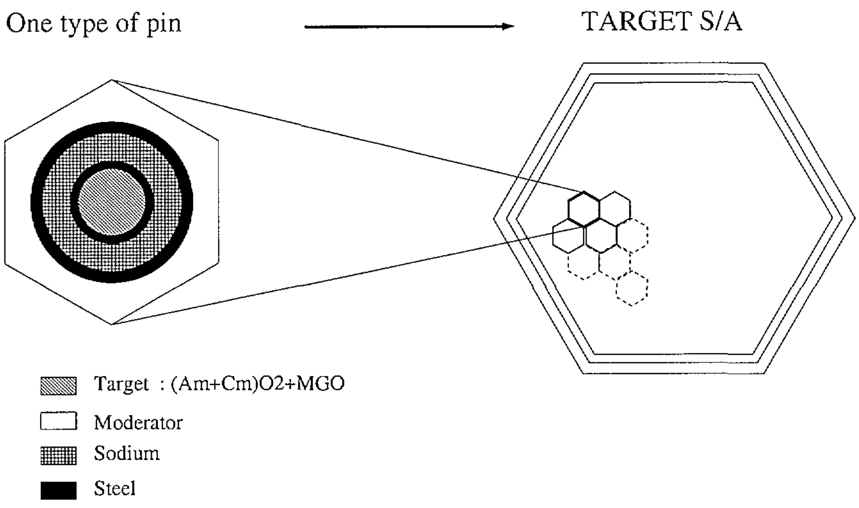

Figure 1: theoretical design.

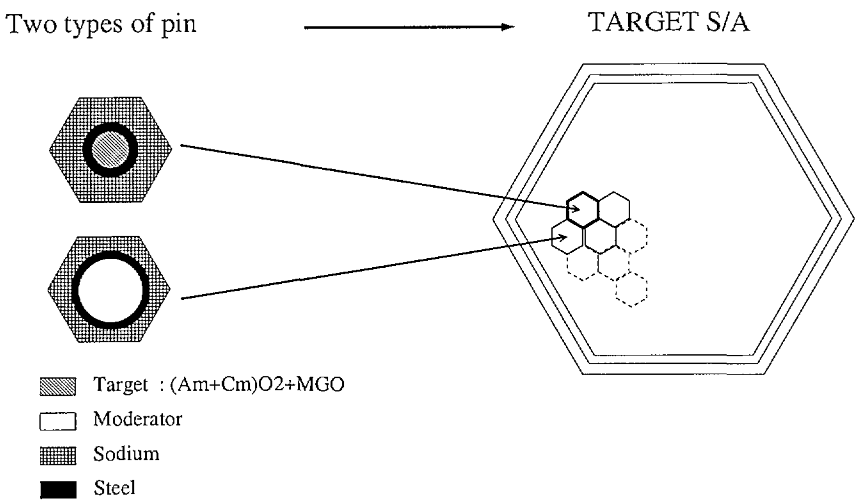

Figure 2: Realistic design.

Am241	71.5
Am242f	0
Am242m	1.6
Am243	19.7
Cm242	$4 \cdot 10^{-3}$
Cm243	0.2
Cm244	6.3
Cm245	0.7
Cm246	$4 \cdot 10^{-2}$
Cm247	10^{-5}
Cm248	0

Table 1: Weight percentage used for optimization procedure

3.2.3 Spacer wires

For the first design no spacer wires where modelled. The spacer wires of the second type of target are fixed to a diameter of 0.145 cm. For the last realistic design, a rule was used to obtain the radius of the spacer wires : it should be at least equal to $0.14 \cdot r^{ext}_{clad}$.

3.3 Optimization ?

For a fixed design of the target, we want to optimize some kind of parameters to achieve the best possible transmutation with regards to technical bounds. For the theoretical concept, the radius of the americium pellet is the chosen parameter. For the second target we choose as a parameter the number of americium pins with respect to the number of moderator pins. The total number of pins must be equal to 469. The last concept is slightly different. As we have two kind of pins with different radius, we must choose a number of pins that will allow the mechanical support of all pins with spacer wires. As a result, we must have a factor 2 between the two different pins. We choose to fix at 312 the number of Actinides pins, and to 157 the moderator pins. The free parameters are then the actinide pellet radius and also the moderator pellet radius within the limitation due to the spacer wire.

3.4 Results : mass consumptions for targets in core

3.4.1 Results for the theoretical design

We can see in figure 3 that the highest possible mass consumption for a target S/A placed in core is around **137 g/TWeh** in the case of a volumic proportion of actinides oxyde of 17% and around **122 g/TWeh** in the 13% case. The mass consumption on these figures are set to zero when one of the two goals ($\tau_F = 90\%$ and $\sum dpa_{max}(NRT) < 200$) is not achieved. The last possible solution is indicated by the circle marker.

3.4.2 Results for the second design

We can see in figure 4 that the highest possible mass consumption for a target S/A placed in core is around **112 g/TWeh** in the case of a volumic proportion of actinides oxyde of 17% and around **96 g/TWeh** in the 13% case. This is less important than the theoretical design because we do not have enough space for increasing at the same time the actinide and moderator volumes.

3.4.3 Results for the third design

We can see in figure 5 that the highest possible mass consumption for a target S/A placed in core is around **104 g/TWeh** in the case of a volumic proportion of actinides oxyde of 17%, around **98 g/TWeh** in the 15% case and around **92 g/TWeh** in the 13% case. This is less important than the theoretical design and a little bit under second target values. Several points are given in this figure corresponding to different moderator pellet radius. The benefit of this target will be explained in chapter 3.6.3.

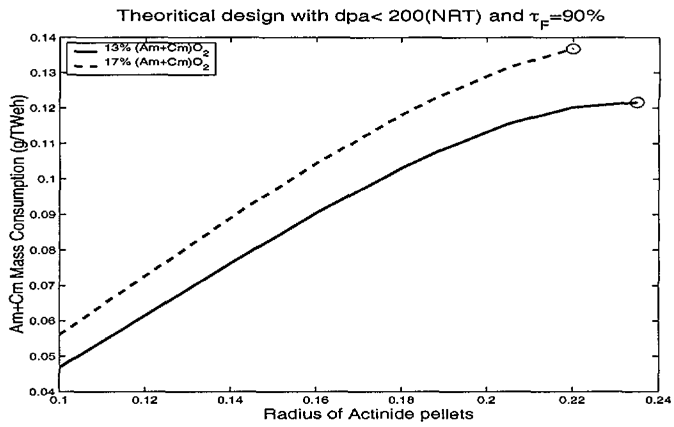

Figure 3: theoretical design : Am+Cm mass consumption.

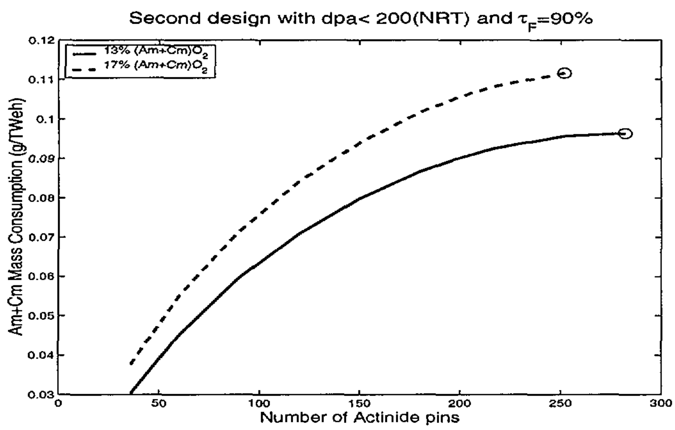

Figure 4: Second design : Am+Cm mass consumption.

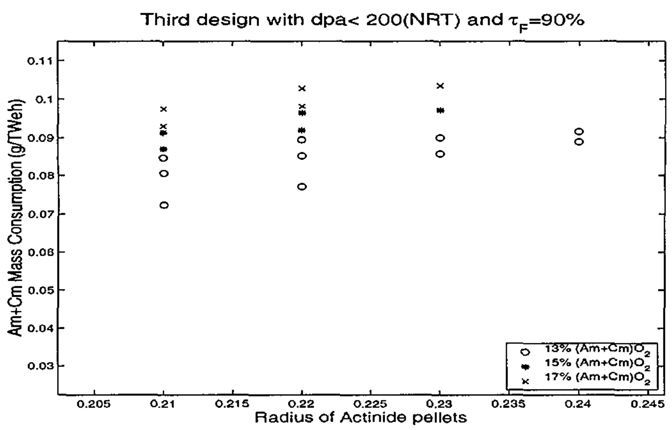

Figure 5: Third design : Am+Cm mass consumption.

Table 2 shows for the three designs (17% case) the best achieved mass consumption for targets placed in the blanket region.

Target	1	2	3
Mass (g/TWeh/Target)	30	26	25

Table 2: Best mass consumption for the three designs when targets are placed in the blanket.

3.6 How to choose the design ?

We have seen that in terms of mass consumption, the two most realistic designs show results lower by a factor 30% with respect to the theoretical design. To optimize the target we should also take a look to the remaining technological limitations.

3.6.1 Helium and gazeous fission products

The clad thickness is chosen to withstand the pressure due to gazeous fission products and Helium produced during the irradiation. The criteria to ensure clad integrity are the following : the membran primary stress, the same as before plus bending stress and the maximum equivalent stress on the cladding. The methodology used is close to the well-known design rules for irradiated structures [4]. We can see in the figure 6 the volume of gaz produced over irradiation time in such moderated S/A for each pins. As the EFR vessels height are rather important (1.30 meter) we do not face problems to dimension the clad thickness for all designs.

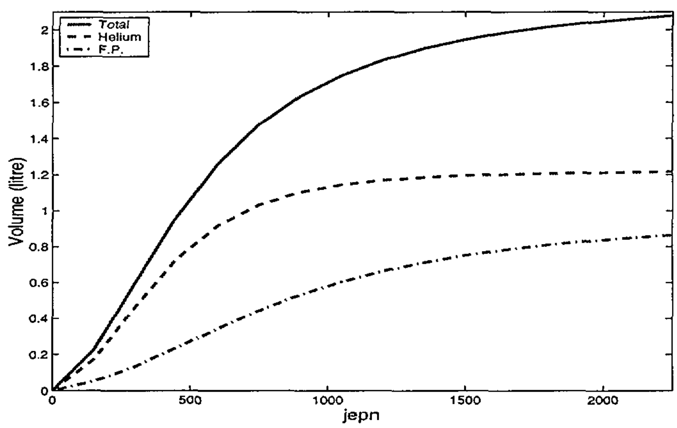

Figure 6: An example of gaz production in one actinide pin.

3.6.2 Pressure drop

We clearly see in the table 3 showing the mean calculation of the pressure drop for the three kind of targets, that the first theoretical design is not realistic : the pressure drop is too high because the coolant flow area is too limited in this concept and also because each sodium cooling channel is separated from the others. For both other designs, the geometry allows a good mixing and flow rate of sodium.

Target	1	2	3
ΔP(bar)	$\sim 10 - 100$	~ 1	~ 1

Table 3: Pressure drop of the targets.

The last but most important technological limit is the temperature in the pin that must be lower than the fusion temperature of AmO_2 (2173 ^{O}C). The key parameter is the maximum linear power in the target. As the first design contains 469 pins (the actinides are diluted) we have for this concept very low power in the pins : always below 315 W/cm Concerning the second design, on the contrary, we have in the best cases around 280 pins, so for the same order of volumic power we will have far more important linear power : the values are always above 471 W/cm for the 17% case and 363 for the 13% case. The last design was originally introduced to solve this problem by increasing the number of actinides pins. As a result the moderator pin radius were increased to achieve a good transmutation. The highest studied linear power is 367 W/cm for the highest mass consumption of 104 g/TWeh (17% case).

The temperature calculation is done with a 2D model using a finite element code. Furthermore, several effect were modelized such that :

- the degradation of the fuel conductivity : we took half of the initial value as starting value,
- the maximum fuel volume swelling was assumed to be 20%,
- the interaction between the fuel pin and the cladding as well as the effect of gaz on this joint conductivity were modelled .

This methodology was formely used to design the ECRIX experiment [5], even if in this case we were more restrictive on limitation values and material behaviour. For each target design results on temperature for several target are listed in table 4. For the third design we gave also the value corresponding to the 15% volumic case optimized for mass consumption.

Target	1	2		3	
W/cm	315	471	363	367	344
g/TWeh	137	112	96	104	98
$T_{pin}(^{O}C)$	<2000	$\gg$2200	~2200	~2200	<2173

Table 4: Center Temperature in actinides pins.

3.6.4 Conclusions

The best realistic design that can achieve the desired transmutation goals and taking into account the technological limitations (with a well-known technology) is the third design. For the core target we choose the 15% volumic case.

For the blanket positions, we do not have this linear power problem (the power is divided by a factor 2-3). We can choose any of the two last kind of designs.

4 Complete heterogeneous calculation

The results of the previous section are important to find a volumic optimization of the target S/A and to compare several target designs. Nevertheless, there is one difficulty remaining to use the previous results. For targets placed in core, as they were obtained with only one target in the center, we must keep in mind that slight differences may occur when going to more targets. We should try to stabilize the power everywhere in the core to be as close as possible to the center problem.

4.1 Studied scenarios and corresponding number of targets

We look at two major scenarios dealing with waste management :

- a 100% fast reactor (EFR) scenario multirecycling Pu+Np. The Am+Cm are placed in dedicated targets in the core,
- a mixed PWR (UOX fuel) and fast reactor (EFR) multireycling Pu+Np and containing targets in the core and blanket regions. The proportion of EFR is around 50% of the whole reactor fleet.

Actinides	100%EFR	UOX+EFR
Am	3.56	3.44
Cm	0.28	0.47
Am+Cm	3.84	3.91
kg/TWeh normalised to EFR		
Am+Cm	3.84	7.02

Table 5: Minor actinides productions in the two studied scenarios.

To obtain roughly the number of targets necessary to burn these actinides, we should look at tab 5 presenting the Am+Cm production of such scenarios.

From the previous chapter, we know that the highest mass consumption per S/A is of around 100 g/TWeh for targets placed in core and 26 for those placed in the first blanket region. For the fisrt scenario, as the fast reactor is self-sustaining (breeding ration ≈ 1), we can not use the blanket region positions for irradiation purposes : they are dedicated to fertile S/A. For the second one we can use the 78 S/A placed in this region because the fast reactor are in this case Plutonium burners. Thus, a first estimate of the necessary number of targets in core is :

$$N_{100\%EFR} = 3.84/0.98 \approx 39(\mathbf{30})$$

and

$$N_{UOX+EFR} = (7.02 - 78 * 0.026)/0.098 \approx 51(\mathbf{35})$$

In parenthesis is given the expected numbers of targets with the theoretical design.

4.2 Complete calculation with optimized realistic target

4.2.1 100%EFR

Table 6 shows the final incineration results for this scenario. 39 targets were placed in the core.

Position	Irradiation time (*jepp*)	Σdpa_{max} (NRT)	C_{Am+Cm} (kg/TWeh)	τ_F (%)
Core	2300	210	3.8	**89**

Table 6: Incineration of Am+Cm : 100%EFR case.

We clearly see that we are very close to what was expected with the optimisation procedure. Nevertheless, to have some more degree of freedom (especially concerning the transmuted mass) we should have added some more target S/A in the core

4.2.2 UOX+EFR

Table 7 shows the final incineration results for this scenario.

Position	Irradiation time (*jepn*)	Σdpa_{max} (NRT)	C_{Am+Cm} (kg/TWh)	τ_F (%)
Blanket (78 targets)	8700	189	2.0	89.7
Core (53 targets)	2300	210	5.0	89.0

Table 7: Incineration of Am+Cm : UOX+EFR case.

Both scenarios will reduce the radiotoxicity of the waste by a factor 20-30 with respect to the open cycle. Pu recycling only (with no Minor Actinide recycling at all) would reduce the waste radiotoxicity by a factor ≈ 6 with respect to the open cycle [6]. Such a limited extra reduction ($\approx 4 - 5$) with Am+Cm burning is due to the residual actinide content in the spent target S/A , e.g. 10% of their initial content if the burn-up is 90% : this emphasizes the need to reach extremely high burn-ups, at least 90%, for once-through moderated targets.

5 Conclusions

This study shows that a very high fission rate ($\tau_F \approx 90\%$) can be obtained with moderated targets. The theoretical target concept has got the highest mass consumption with an acceptable maximum linear power. If we look at more realistic targets with a well-known technology (pins, spacer wires, ...) we end up with an optimized target that allows to transmute the desired Am+Cm actinides in the two scenarios. This target S/A respects all the technological limitations : clad thickness, pressure drop, temperature in pins, mechanical sustaining.

Nevertheless, one major aspect of this study is that the best target solution should be the one where actinides are diluted over the S/A to obtain correct linear power. The theoretical target was going in that direction but with a non-realistic design based on pins. One hint of solution is to look at particle bed fuel type S/A. This will permit to be closer to the neutronic optimization by having low linear powers.

Furthermore, another aspect worth looking at concerning this kind of scenarios is the impact of the moderated S/A on the core concentration equilibrium and in the mixed PWR/FR case on the proportion of fast reactors. This study, still going on, has got two major aspects :

- diode concepts to trap thermal neutrons escaping from the targets and producing power increase in neighbouring S/A,
- more precise equilibrium calculation using a realistic 3D hexagonal model of the fast core.

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