TRANSMUTATION OF MINOR ACTINIDES IN PWRs : PREPARATION OF THE "ACTIIWEAU" EXPERIMENT

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ABSTRACT

This paper deals with the experimental work that is performed at the CEA to study the metallurgical problems that need to be overcame in order to transmute neptunium and americium in PWRS. After recalling the experience gained on transmutation under thermal flux, the preparation of the experimental irradiation ACTINEAU is described. This experiment, to be carried out in the OSIRIS reactor, will test both homogeneous and heterogeneous recycling with different inert matrices and different mass fractions of actinide in the rods. According to prediction, a transmutation rate of over 50 % is expected after 600 days of irradiation.

INTRODUCTION

In the framework of the CEA SPIN programme on the partitioning and transmutation of long-lived radiotoxicity products, metallurgical studies are in progress in order to assess the feasibility of transmuting minor actinides (neptunium and americium), to identify the problems from the material point of view, and to propose the best options for fuels and targets in agreement with the different strategies [1]. Both homogeneous and heterogeneous recycling are considered, as well as two types of reactors : Pressurized Water Reactors (PWRs) and Fast Reactors (FRs).

Thanks to the SUPERFACT experiment in PHENIX [2], experimental results, related to fabrication problems and to the in-pile behaviour of pins containing Np and Am, have been obtained for transmutation in FR conditions.

For transmutation under thermal flux, the only experiment performed in the CEA aimed to produce ^{238}Pu for medical application (pacemakers). Experience of the implementation of neptunium on a medium scale (20 kg) was thus acquired. But this irradiation performed in the CELESTIN reactors was carried out in conditions too different from those of a PWR to allow assessment of in-pile behaviour.

It was therefore decided to perform an experimental irradiation, called " ACTINEAU", in order to obtain information on the metallurgical problems that arise while transmuting neptunium and americium in PWR conditions.

After summarizing the experience gained in the CELESTIN reactors, this paper describes the preparation of the ACTINEAU experiment.

I. Transmutation of neptunium in the CELESTIN reactors.

In the 1970s, a large scale neptunium transmutation programme was carried out in the Celestin reactors, at Marcoule, with the aim of producing ^{238}Pu for pacemakers.

In the first experiments, the neptunium was in the form of an Np - Al plate (modelled on the standard Celestin fuel consisting of U - Al plates). Use of this material however was soon abandoned as the intended medical application imposed a very low 236Pu content ($< 110^{-6}$), and (α,n) reaction with aluminium had a detrimental effect from this point of view

Subsequently, the entire experiment was concentrated on Np02 - MgO targets, generally with a Np02 mass fraction of 50% (or approximately 25 % of the volume).

From 1973 to 1976, 26 target elements, each containing approximately 700 g of neptunium, were fabricated at the Plutonium Technology Workshop at Cadarache. Thus almost 20 kg of neptunium were implemented.

This fabrication was carried out using standard powder metallurgy in glove boxes with lead panels : by decay, ^{237}Np (which has a very long half-life : 2.14 10^6 years) produces ^{233}Pa whose half-life is only 27 days and which is therefore in equilibrium with its parent after a few months. ^{233}Pa emits 40 % of its radiation in the form of 310 keV gamma radiation. A biological shield is therefore necessary whenever large quantities of neptunium need to be handled.

The fabrication cycle was based on the fuel fabrication procedure : calcination in air, powder mix, lubrication, sintering for 4 hours at 1600°C in Ar - H2 gas. The columns produced were 50 cm long, clad in zircaloy and assembled in clusters each containing 16 pins.

These clusters were then irradiated during about 100 days in a flux of 2 to 31013 $n.cm^{-2}.s^{-1}$ (a highly thermalized flux as the reactors are moderated by heavy water). The conversion rate of Np to Pu was around 6 % with a ^{238}Pu isotopic abundance greater than 90%. Increasing the irradiation time would have provided a higher Np to Pu conversion rate but at the cost of the ^{238}Pu isotopic abundance, which was undesirable.

This proganu-ne therefore showed that it is possible to implement significant quantities of neptunium and to transmute a not inconsiderable proportion during a short term irradiation.

No cladding breach occurred but the irradiation conditions (coolant temperature, linear power, residence time, burnup, etc...) were not representative of those required for the actinide burning programme.

11. Preparation of the ACTINEAU experiment.

A. Objectives of the experiment

The ACTINEAU experiment (ACTinides Incinerated in water - EAU - reactor) aims at :

- assessing the feasibility of transmutation in water reactors,

- studying the metallurgical behaviour of fuels and targets and the thermomechanical behaviour of the rods with a view to their subsequent optimization,

- comparing the efficiency of actinide burning in a thermal flux with that in a fast flux, thanks to the SUPER-FACT 1 results [2].

The experiment will investigate the homogeneous concept (U02 fuel with 2 % Np02 or AmO_2) and the heterogeneous concept (target rods with a higher actinide content within an inert matrix - without uranium).

ACTINEAU is planned to take place in the OPERA loop of the OSIRIS reactor at SACLAY and should begin when this loop comes into operation at the end of 1994. In this loop, the thermohydraulical conditions will be similar to those of a PWR (P = 155 bar, T_{water} = 300°C to 330°C).

This experiment is prepared in cooperation with the TransUranium Institute of Karlsruhe, which will fabricate and study the materials containing americium.

B. Choice of the matrices and definition of the heterogeneous pins.

As it is not possible to irradiate pure neptunium or americium ceramics, the targets for testing heterogeneous recycling have to be a composite material of actinide ceramic with an inert matrix. The first stage consisted in choosing the most suitable materials for inert matrices. Among the different types of possible ceramics and metals, safety considerations led us to choose oxides : the only ones which do not increase hydrogen production in the case of an accident.

Among the different oxides, a preselection was made with the following criteria :

- low neutron cross-section of the element (lower than ^{238}U), and low generation of activation products,

- good compatibility with water up to 350°C (to avoid an actinide leak in case of cladding breach), and with the zircaloy cladding,

- high melting, phase change, or eutectic with the actinide compound, temperature (preferaMy higher than 1900°C),

- properties suitable for irradiation : good thermal conductivity (better than UO_2), thermal expansion similar to that of U02, etc...

- good behaviour under irradiation : the matrix must be able to withstand the neutron flux and the recoil of fission products with neither excessive swelling nor loss of its properties,

- ability to he fabricated,

- ability to be reprocessed.

Unfortunately, there is no material that perfectly fits all these criteria, and for many points there is a patent lack of knowledge. Several possibilities were eliminated for different reasons : for exemple MgO, the only material for which there was manufacturing and reprocessing experience, had to be discarded because our tests in an autoclave showed that, in water at 350°C, it was rapidly changed into a fine $Mg(OH)_2$ powder. ZrO_2 was discarded because of its poor thermal conductivity. BeO, despite its very good thermal conductivity, was not suitable because of the (α,n) reaction (fabrication problem), and poor behaviour under irradiation.

By this process of elimination, the only remaining possibilities among the oxide ceramics were :

* simple oxides : Al_2O_3, CeO_2, Y_2O_3,

* complex oxides : spinel ($MgAl_2O_4$), and YAG ($Y_3Al_5O_{12}$)

Among these different oxides, our first choices were spine] and alumina : spinel has the best behaviour under irradiation (up to a fluence of 2 1022 n. cm-z) ; alumina is also quite a well-known ceramic ; although its properties are altered by irradiation, its swelling remains of the same order as that of U02. By means of autoclave tests we verified that these two oxides were fully compatible with water ; we also verified however - and this is the main drawback of $MgAl_2O_4$ and Al_2O_3 - that they are not soluble in nitric acid. At the beginning of this programme, we did not lend too much weight to this reprocessing criterion because we had not excluded the possibility of reaching such a high transmutation rate that the rods could be sent directly to storage ; unfortunately this now appears rather utopian : even if very high transmutation rates are obtained, the fission rates will not be high enough to significantly decrease long term radiotoxicity.

It was therefore decided on the one hand, to continue preparing the experiment with these two oxides, spinel and alumina, on the other hand, to seek a solution to this reprocessing problem. This research is twofold and will involve either changing the reprocessing process (or finding a way to dissolve the actinide oxide without dissolving the matrix), or using other less well-known matrices. Our studies are therefore continuing on other oxide ceramics : Y_2O_3, Ce02, $Y_3Al_5O_{12}$ have been fabricated and characterized ; they are being tested in water and nitric acid, and they will be irradiated alone at high fluence in an other experiment.

The percentage of actinide to be introduced into these heterogeneous targets results, on the one hand, from the following consideration referring to an industrial scale programme : the higher the percentage, the fewer the pins needed to recycle a given amount of actinide ; but also the more difficult it will be to reach a high transmutation rate as the material will have to be submitted to greater damage. On the other hand, referring now to the special case of the ACTINEAU experiment, this percentage is limited by the linear power, the aim being to maintain this linear power at a level not exceeding that of standard PWR fuel pins.

Neutronic calculations have shown that, in the conditions of the OPERA loop and with standard geometry rods, the highest possible mass fraction of Np02 is 40 % : it gives a linear power of about 250 W/cm after 600 irradiation days ; we therefore decided to test two percentages of NpO_2 : 20 % and 40 % for each of the two inert matrices Al_2O_3 and $MgAl_2O_4$.

C. Content of ACTINEAU : grid of the experiment.

8 pins can be irradiated in the OPERA loop. The following parameters are those we wish to investigate :

- concept : homogeneous and heterogeneous ; a reference rod (U02) is also necessary in order tc

calibrate the analysis after irradiation,

- minor actinides : neptunium and americium,

- inert matrices : alumina and spine],

- percentages :20 and 40 % (weight).

Taking into account the fact that the neptunium rods will be available sooner than the americium rods, the following grid (Table 1) was decided for the first loading :

↑ towarc core

$MgAl_2O_4$ + 20 % Np02 (1	$MgAl_2O_4$ 20 % Np02 (2	Al_2O_3 +20 % Np02 (3
$MgAl_2O_4$ + 40 % Np02 (4		Al_2O_3 + 40 % Np02 (5
UO_2 (2 % ^{235}U) (6	Al_2O_3 40 % Np02 (7	J02 (2 % ^{235}U) +2 % Np02 (8

Table 1: grid 1 for the first loading (O to 200 days).

After 200 irradiation days, two pins (number 2 and 7) will be unloaded for destructive examination, the two americium pins will be loaded, and some changes of position will be made to take into account the flux gradient in the cluster. The grid presented in Table 2 will then be loaded for the next 400 days of irradiation.

D. Rod design.

To meet the objectives of the experiment, rod design would have to remain similar to that of standard PWR rods. This is possible as far as diameter, helium pressure or maximum linear power are concerned.

The OPERA loop is designed for 75 cm long rods (in OSIRIS the flux is about 60 cm high). In order to spare neptunium and americium however, we use fuel or target columns 25 cm long. As a result, a large plenum is available. Hafnium pellets are arranged at each end of the stack in order to avoid a power increase at this level.

↑ towards core

MgA1204 + 20 % Np02 (1)	$MgAl_2O_4$ + 20 % Am02 (2)	Al_2O_3 +20 % Np02 (3
UO_2 (2 % ^{235}U) (4)		UO_2 (2 % ^{235}U) +2 % Np02 (5
MgA1204 + 40 % Np02 (6	UO_2 (2 % ^{235}U) +2 % Am02 (7	Al_2O_3 + 40 % Np02 (8

Table 2 : grid 2 for the second loading (200 to 600 days).

E. Neutronic calculations.

These calculations were made using diffusion and evolution codes with constants derived from results of the APOLLO code with the loop in the closest position to the core. The thermal flux, without perturbation, on the first pins equals 21014 $n.cm^{-2}.s^{-1}$.

1. Linear power evolution,

In homogeneous fuel, the only constraint is to keep the linear power at a reasonable level : therefore the enrichment was calculated so as not to exceed a linear power of 250 W/cm at beginning of life. With a standard rod diameter, in the OPERA flux, this corresponds to an enrichment of 2 % ^{235}U. The linear power decreases slightly during irradiation.

In neptunium heterogeneous fuel, the power, which is initially very low, gradually rises mainly due to the fission of the ^{238}Pu and ^{239}Pu isotopes created by neptunium transmutation (see an example of evolution for pin 1, with 20 % Np02, in Figure 1). A further addition to this power comes from the fission of ^{238}Np which has an extremely high fission cross-section but a very short life (half-life of 2.3 d). This explains the discontinuities in Figure 1 which are caused by the generation of ^{238}Np during each 20 day run and its almost complete disappearance after each 10 day inter-run.

Calculation shows that in rods containing 20 % Np02 linear power reaches a plateau after about 500 irradiation days : plutonium consumption by fission

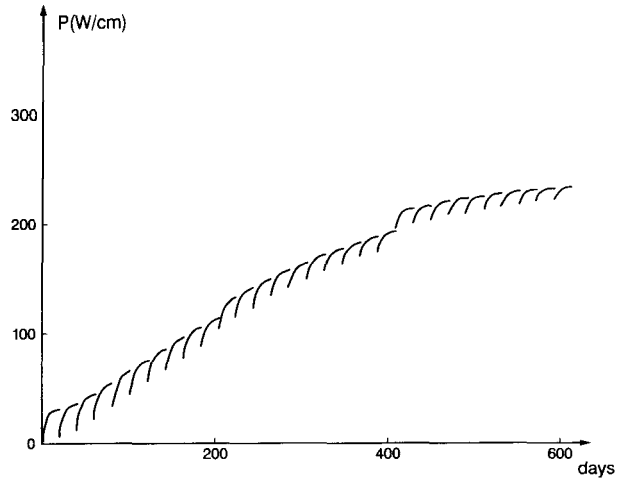

Figure 1 : Evolution of linear power in the rod containing MgA1204 + 20 % Np02, during the 600 days of irradiation.

almost equals its production by neptunium transmutation, wherein in rods with 40 % Np02, the power still increases after 600 days.

In americium heterogeneous pins, there will again be a continuous increase of linear power (Figure 2) if the americium is pure 241 Am as it is planned for this experiment. If americium is a mixture of isotopes 241, 242 and mainly 243, as will be the case if the operation is performed on an industrial scale, the evolution of linear power is different : there is only a slight increase.

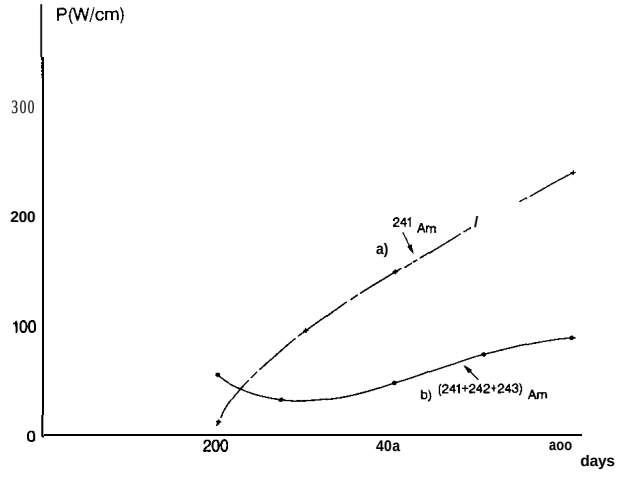

Figure 2 : Evolution of linear power in the rod containing MgA1204 + 20 % Am02, during its irradiation (a- with pure 241 Am, b- with a mixture 27.5 % ^{241}Am, 0.5 % 2^{4}2 Am, 72 % ^{243}Am).

2. Transmutation rates

The calculated transmutation rates of neptunium and americium are given in table 3 for the different pins (referring to grid 2 in Table 2) after 200, 400 and 600 days of irradiation.

↑ towards core		
$MgAl_2O_4$ + 20 % Np02 200d: 26% 400d: 46% 600d: 62%	MgA1204 + 20 % Am02 200d: 70% 400d: 89%	Al_2O_3 +20 % Np02 200d: 26% 400d: 45% 600 d : 61%
UO_2 (2 % ^{235}U)		UO_2 (2 % ^{235}U) +2 % Np02 200d: 20% 400d: 37% 600d: 50%
MgA1204 -t 40 % Np02 200 d : 18% 400d: 32% 600d: 44%	UO_2 (2 % ^{235}U) +2 % Am02 200d: 48% 400d: 75%	Al_2O_3 + 40 % Np02 200 d : 18% 400 d : 31% 600d: 43%

Table 3: Transmutation rates calculated in the different pins after 200, 400, and 600 days of irradiation.

After 600 irradiation days, the calculated transmutation rates of neptunium are 44 %, 62 %, and 51 % for the rods containing 40 %, 20%, and 2 % Np02 respectively. For 241 Am, the transmutation rates are even higher : almost 90 % in the heterogeneous rod after 400 days.

At the end of the irradiation, the burnup of the homogeneous rods will be about 22000 MWd/t, whereas heterogeneous rods will reach a burnup of about 40000 MWd/t.

An assessment of the radial depression of flux and the transmutation rate was also made in order to verify that the autoprotection at the periphery of the pellets was not too high. Calculations predict a factor 2 between the periphery and the center in americium pellets, and a lower gradient in neptunium pellets. Such gradients are not high enough to be an obstacle to the objectives of the experiment.

F. Fabrication.

Fuels and targets containing neptunium are fabricated at Cadarache, while those containing americium are manufactured at the TransUranium Institute of Karlsruhe.

Np02 (and Am02) forms with U02 a solid solution very similar to (U, $Pu)O_2$. Therefore the fabrication of U02 pellets containing 2 % (weight) of Np02 can be performed in a similar way to the fabrication process of MOX fuels ((U, $Pu)O_2$ for PWR) : powder mixing by co-milling, pelletizing at about 400 MPa and sintering at 1700°C for 4 hours.

The fabrication of heterogeneous type pellets has first begun with U02 simulating Np02. According to the phase diagrams, the matrix phase (corundum or spinel) is in equilibrium with U02 (and probably with NpO_2) : no volubility of one into the other. Fabrication tests aimed at achieving pellets as similar as possible to the standard U02 pellets manufactured for irradiation in PWRS : the same geometry, a bulk density higher than 90 % of theoretical density, and an impurity level of less than 2500 ppm. Moreover it is necessary to obtain a regular distribution of the actinide particles and, if possible, the centerless grinding step should be avoided.

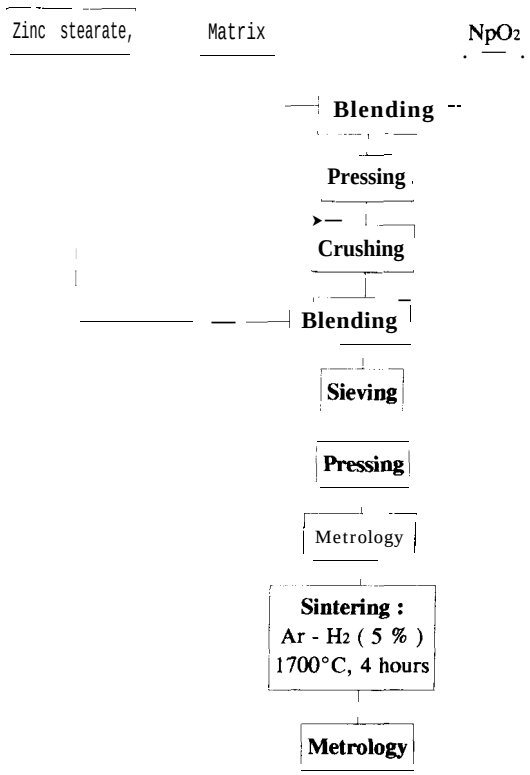

Figure 3 : Flowsheet of the fabrication of pellets devoted to heterogeneous recycling.

Based on the same flowsheet for both matrices (Figure 3), the first fabrication tests were performed in order to select the best alumina and spinel powders and to assess their compaction behaviour and sinterability under the specific conditions imposed by the fabrication of actinide oxides. Examples of densification results obtained for (80 % matrix, 20 % actinide) oxides are ven in Table 4:

Matrices	Powder	Pressure (MPa)	Density (% T. D.)
A1203	Baikalox CR 15	300	94. I
Al_2O_3	Cerac A 1124 200		94.9
$MgAl_2O_4$	Baikalox S 30 CR	300	95.3
$MgA120_4$	Cerac M 1117	500	89.9

Table 4 : Example of densities obtained depending on the powder used and pelletizing pressure.

Fabrication tests are in progress with Np02 powder. As the amounts of neptunium oxide implemented for the fabrication of these experimental fuels and targets are low (maximum some tens of grams for each batch), the gamma activity of Np remains in an order of magnitude which allows the utilisation of the laboratory unshielded equipment devoted to the fabrication of (U, $Pu)O_2$ fuels.

G. Assessment of behaviour under irradiation.

In the homogeneous concept, the content of Np02 or Am02 is too low (2 %) to significantly modify the behaviour of U02. Therefore the in-pile behaviour of homogeneous pins will probably be similar to standard U02 rods irradiated at the same linear power.

Main uncertainties and potential problems are linked to the irradiation of the composite materials testing the heterogeneous concept.

The matrices will be damaged by the neutron flux (up to 2.4 1021 $n.cm^{-2}$ fast neutrons). According to literature, the swelling generated by this fluence will be negligible for MgA1204 and less than 2 % for Al_2O_3. The damage generated by the recoil of fission products is hard to assess due to a lack of representative experiments : this is why the destructive examination of two pins is planned after 200 days of irradiation.

The calculation of temperatures inside the heterogeneous targets at the end of irradiation is difficult as several data and models that would be necessary are unknown : evolution of thermal conductivity under irradiation, restructuring of the pellets by cracking and swelling, gap closure, gas release, etc... Because of the low value of the fast flux in the OPERA loop, the effect of irradiation creep of the clad under external pressure will be lower than in a PWR. With a reasonable hypothesis concerning the unknown phenomena, central temperatures ranging from 900 to 1200°C are calculated for end-of-life in the highest loaded rods, depending upon the assumptions that are made.

CONCLUSION

To conclude, at the preparation stage of the ACTINEAU experiment, the actinide burning operation in PWRS seems feasible. Transmutation rates of about 50 % seem to be attainable within 600 days both in homogeneous and in heterogeneous recycling, provided the inert matrices behave well under irradiation.

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