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# FIRST RESULTS AND FUTURE TRENDS FOR THE TRANSMUTATION OF LONG-LIVED RADIOACTIVE WASTES

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## INTRODUCTION

The study of the partitioning and transmutation of long-lived radioactive nuclei in order to reduce the long-term potential risk source, is one of the research directions foreseen by the French Parliament law on high level activity nuclear wastes.
At CEA the corresponding R and D activities are gathered in a coordinated program (SPIN program).
The present paper is related to the transmutation activities performed at the Nuclear Reactor Direction, which are performed in the frame of a unique project (PRIA : Actinide Burning Reactor Project) and which cover basic physics, core, fuel and material studies.
The basic physics studies are related to the understanding of the physical mechanisms of transmutation, basic data experimental validation and assessment.
The core and fuel studies are devoted to the analysis of the potential of standard and innovative fission reactor concepts, their fuel and the associated fuel cycles.
These are the studies which will be summarized in the present paper. Further studies are also presently devoted to more advanced transmutation devices, such as accelerator/subcritical blanket systems.
Finally, it has to be mentioned that most CEA studies are performed in the frame of international collaborations (KfK [1], Transuranium Institute Karlsruhe [2], PSI - Villigen, ECN - Petten, Obninsk Institute, Kurchatov Institute, PNC, CRIEPI, ENEA, etc.).

# 1 - THE CONTEXT AND OBJECTIVES

The objective of the transmutation studies can be simply specified as the reduction of the long term potential risk source by transmutation of radioactive nuclei with half-lives higher than a few hundred years into stable nuclei, either directly or after decay of nuclei of much shorter half-lives.

If one is concerned with the potential radiotoxicity source term inside a deep storage, the nuclei of interest are the actinides of the four α-families

Of course some long-lived fission products are of relevance if one is concerned with the return to the bio-sphere of the radioactive nuclei after solution and migration in the geological environment.

Since the main topic of the present paper is the status of the studies related to the actinide transmutation, we will briefly recall the main characteristics of the actinide production (for example in the frame of the present French nuclear park) and their contribution to the radiotoxicity source term.

## 1.1 - Actinide production

One can summarize the present situation as follows :

- First, one has to consider Pu (~ 200 g/MWe.y production).
  If losses at reprocessing are assumed to be ≈ 0.3 %, then the amount of Pu going to the wastes is approximately 30 Kg/y.
- Neptunium : 10 g/MWe.y production.
  For the long term, it is useful to add the quantities resulting from Pu-241, Am-241 and Cm-245.
  If the Pu losses are ≈ 0.3 %, the potential Np-237 going to wastes is approximately 800 Kg/y.
- Americium :
  *Am-241* : For a reprocessing 3 years after exit from reactor, the production is of 5 g/MWe.y (which means 250 Kg/y).
  *Am-242m* → 0.014 g/MWe.y (small production)
  *Am-243* → 3 g/MWe.y production.
  It represents the major source of Pu-239 in the wastes at $t = 10^4 \div 10^5$ years ($7 \div 8$ times higher than Pu-239 resulting from reprocessing losses).
- Curium :
  *Cm-242* ($T_{1/2}$ = 6 month). It decays into Pu-238.
  *Cm-243* ($T_{1/2}$ = 28.5 y). It decays into Pu-239, and represents a negligible amount with respect to Am-243.
  *Cm-244* ($T_{1/2}$ = 18 y). It decays into Pu-240 (~ 5 times higher than Pu-240 resulting from reprocessing losses).
  *Cm-245* ($T_{1/2} = 8.5 \times 10^3$ y). It decays into Np-237, but in small amount.

## 1.2 - Radiotoxicity source term

To each isotope it is associated an "ingestion danger" coefficient : (in SV/Bq).

For a unit mass of a given isotope, the radiotoxicity R(t) at an instant t is

given by :

$$R(t) = a \cdot \sum_i \frac{Q_i(t)}{Q_0(0)} \frac{D_i}{M T_i} \quad (SV/g)$$

- where is a constant ($= 1.322 \times 10^{16}$) ;
- The sum is over the "father" isotope and its progenies ;
- $Q_0(0)$ initial number of "father" nuclei ;
- $Q_i(t)$ number of nuclei of isotope i at t ;
- M molar mass of "father" isotope ;
- $D_i$ "ingestion danger" coefficient (SV/Bq) of isotope i ;
- $T_i$ radioactive half-live of isotope i.

Using the present recommendation for the $D_i$ values, issued by the IPSN (Nuclear Installations Protection and Safety Institute), we can evaluate the radiotoxicity source R (t) in the repository at different times after storage and making different hypothesis.

Tables 1 and 2 give the contribution to R (t) of the different actinides for an irradiated PWR fuel (33.000 MWd/t), cooled for three years.

Table 1 is related to the "open cycle" scenario (i.e. all Pu and minor actinides are stored) and Table 2 to a "closed cycle" - type scenario, in which all minor actinides go the repository together with the losses of Pu at reprocessing (assumed to be 0.3 %) (however the Pu recycling is not explicitely accounted for).

In the open cycle scenario (table 1), the radiotoxicity source term is obviously dominated by the contribution of the different Pu isotopes.

Table 2 shows that in the case of Pu reprocessing there is a global reduction of the radiotoxicity source term of approximately a factor of 10 (at all times). Moreover the contribution of the different isotopes shows the relevance of Am-241 for $t \approx 10^2 \div 10^3$ years, the strong contribution of Am-243 (via the formation of Pu-239) at $t \approx 10^4 \div 10^5$ years, and the not-negligeable contribution of Cm-244 (via the formation Pu-240) at $t \approx 10^4$ years. As far as Np-237, its contribution is dominant at $t = 10^6$ years (the Am-241 contribution is related to its decay into Np-237).

Finally in terms of radiotoxicity source, the fission product contribution is small, their relevance being enhanced in the scenario of a possible return to the bio-sphere after solution and migration in the geological environment.

## 2 - FISSION REACTOR STUDIES

The following studies have been performed parametrically :

- Minor Actinides transmutation (mainly Np and Am) :
  homogeneous / heterogeneous } recycling modes in FRs and PWRs.

- Evaluation of the influence of { fuel type (MOX, UOX, metal, ...), reactor size / spectrum, moderator / fuel ratio.

- Evaluation of the consequences on the core neutronics parameters.
- Evaluation of the consequences on the fuel cycle (neutron sources, activity, decay heat,...).
- Fission Products transmutation :
- use of moderated S/A at the periphery of a fast reactor (mainly for Tc-99 and I-129).

The major results are summarized in tables 3 and 4, respectively for fast reactors and for light water reactors.

2.1 - Fast Reactors

The superior neutron economy of fast reactors and the better fission/capture ratios for most actinides, results in favourable transmutation performances. The fuel type (oxide or metal) has a moderate influence (mainly related to the higher fission/capture ratios in a metal core spectrum). In the case of an homogeneous multiple recycling of Np and Am, the maximum amount of minor actinides is fixed in order to avoid any significant increase of the positive sodium void coefficient in the most penalizing situation (i.e. at end of cycle) and to avoid a Pu-238 build-up in the fuel beyond a ~ 5 % value (for fuel fabrication and reprocessing purposes). The decrease of the reactivity loss over the cycle is a favourable effect, which can be used for a further core optimization.

In the heterogeneous recycling of minor actinides in dedicated subassemblies at the periphery of the core, the maximum amount of minor actinides targets, is fixed by the need to avoid strong flux gradients and their evolution during the irradiation cycle. Moreover, the flux level at core boundary and the transmutation rates are such that, even in case of long ($> 3 \div 5$ years) irradiation cycles, a multiple recycling strategy is necessary, which implies to implement a separate and specific fuel cycle.

Different elements can be treated in different ways and one can say on the basis of the present results, that an homogeneous recycling of Np (together with Pu) seems the most promising strategy, while Am due to fabrication and fuel cycle specific problems can be transmuted separately, possibly in an heterogeneous mode, using the high neutron flux at the periphery of a fast reactor, and making use of subassemblies with a moderator materials, in which Am bearing targets are introduced, in order to increase the transmutation rates.

As far as the consequences on the fuel cycle, the homogeneous mode gives rise to relatively small increases of the core decay heat and of the neutron sources and fuel activity in the out-of-pile fuel cycle. In the case of an heterogeneous recycling, the specific fuel cycle has to be defined in detail before drawing conclusions.

2.2 - Pressurized Water Reactors

The major drawback of using a minor actinides homogeneous recycling is due to the tighter neutron economy in this type of reactors. A significant increase ($> 20$ % in relative) of the U-235 enrichment is needed for about 1 % minor actinide mixed to the fuel. The reactivity coefficients are

worsened but in tolerable way. The consequences on the fuel cycle are much more severe than in the case of fast reactor transmutation (e.g. the neutron sources are increased by a factor of at least 30).
Moreover, the high capture cross-sections of the minor actinides in a thermal spectrum, give rise to the production of significant quantities of higher atomic mass actinides.
The heterogeneous multiple recycling can be a better option and an example is given in a companion paper presented at this conference by Puill et al. However, the associated specific fuel cycle impact has to be evaluated in detail.

2.3 - Consequences on the radiotoxicity source term

To evaluate the benefits of the transmutation in fission reactors indicated above, simple evaluations have been made. It is necessary to make hypothesis on the partitioning efficiency in the reprocessing steps. If one

| | | TIME (y) | | | | | |
|---|---|---|---|---|---|---|---|
| ACTINIDES | MASS (g) | $10^2$ | $10^3$ | $10^4$ | $10^5$ | $10^6$ | $10^7$ |
| Np 237 | 10040. | | | | 1. | 17. | 12. |
| Am 241 | 5187. | 9. | 8. | | | 8. | 6. |
| Am 242m | 14. | | | | | | |
| Am 243 | 2954. | | 1. | 3. | 2.5 | | |
| Am | | 9. | 9. | 3. | 2.5 | 8. | 6. |
| Cm 243 | 10. | | | | | | |
| Cm 244 | 768. | | | | | | |
| Cm 245 | 38. | | | | | | |
| Cm | | 0. | 0. | 0. | 0. | 0. | 0. |
| Pu 238 | 4343. | 17. | | | 4. | 6. | |
| Pu 239 | 137771. | 5. | 17. | 58. | 78. | 3. | 39. |
| Pu 240 | 52840. | 7. | 22. | 58. | | | 3. |
| PU 241 | 33297. | 61. | 51. | | 7. | 53. | 38 |
| Pu 242 | 130029. | | | | 7. | 13. | |
| Pu | | 90. | 90. | 96. | 96. | 75. | 80. |
| Radiotoxicity (SV) | | $7.37\ 10^9$ | $2.07\ 10^9$ | $4.75\ 10^8$ | $2.6\ 10^7$ | $2.57\ 10^6$ | $1.90\ 10^5$ |

Table 1 : Contribution of each isotope to radiotoxicity (%) irradiated PWR fuel (33.000 MWd/t) 3 years cooling - 100 % Pu, Np, Am, Cm (open cycle)

| | | TIME (y) | | | | | |
|---|---|---|---|---|---|---|---|
| ACTINIDES | MASS (g) | $10^2$ | $10^3$ | $10^4$ | $10^5$ | $10^6$ | $10^7$ |
| Np 237 | 10040. | | | 1.5 | 29. | 65. | 63. |
| Am 241 | 5187. | 88. | 81. | | 15. | 33. | 32 |
| Am 242m | 14. | | | | | | |
| Am 243 | 2954. | 3. | 12. | 75. | 50. | | |
| Am | | 91. | 93. | 75. | 65. | 33. | 36. |
| Cm 243 | 10. | | | | | | |
| Cm 244 | 768. | 5. | 3. | 13.5 | | | |
| Cm 245 | 38. | | | 1.5 | | | |
| Cm | | 5. | 3. | 15. | 0. | 0. | 0. |
| Pu 238 | 13. | | | | | | |
| Pu 239 | 413. | | | 4. | 5. | | |
| Pu 240 | 158. | | | 3. | | | |
| Pu 241 | 100. | 2. | 2. | | | | |
| Pu 242 | 39. | | | | | | |
| Pu | | 2. | 2. | 7. | 5. | 0. | 0. |
| Radiotoxicity (SV) | | $7.5\ 10^8$ | $1.9\ 10^8$ | $1.9\ 10^7$ | $1.3\ 10^6$ | $6.5\ 10^5$ | $3.9\ 10^4$ |
| Fission Products | | | | | | | |
| Tc 99 | 17405. | $3.7\ 10^3$ | $3.7\ 10^3$ | $3.6\ 10^3$ | $2.7\ 10^3$ | $1.4\ 10^2$ | 0. |
| I 129 | 4026. | $1.9\ 10^3$ | $1.9\ 10^3$ | $1.9\ 10^3$ | $1.9\ 10^3$ | $1.9\ 10^3$ | $1.2\ 10^3$ |
| Cs 135 | 9768. | $7.9\ 10^2$ | $7.9\ 10^2$ | $7.9\ 10^2$ | $7.8\ 10^2$ | $5.9\ 10^2$ | $3.9\ 10^1$ |

Table 2 : contribution of each isotope to radiotoxicity (%) irradiated PWR fuel (33.000 MWD/T) 3 years cooling - Losses = 0.3 % Pu, 100 % Np, Am, Cm to the wastes

| | HOMOGENEOUS RECYCLING | HETEROGENEOUS RECYCLING |
|---|---|---|
| Consequences on the core parameters | Na void coeff. ↑ (BOC)<br>Na void coeff.~const (EOC)<br>Doppler coeff.↓<br>Δρ/cycle ↓ | Control of power distributions at the core outer boundary |
| Pu-238 content (Pu-238/Pu) | ~ 5 % | ~ 80 % |
| Envisageable MA Content | ~ 2.5 % (EFR-type)<br>~ 5 % (PRISM-type) | ~ 50 % (Np)<br>(less for Am) |
| Spectrum, fuel type, reactor size effects | Small | - |
| Waste radiotoxicity reduction | Factor ~ 10 to ~ 30 | Factor ~ < 10 to 30 |
| Ratio FR/Park | ~ 20 % | ~ 30 % |
| Recycling mode | Multiple recycling | Multiple recycling |
| Consequences on the fuel cycle | Decay heat : + 20 + 50 %<br>n source : + factor 2 | Specific fuel cycle to be evaluated |
| Experimental validation | SUPERFACT experiment | Foreseen |

Table 3 : Minor actinide burning in fast reactors

| | HOMOGENEOUS RECYCLING | HETEROGENEOUS RECYCLING |
|---|---|---|
| Consequences on the core parameters | Reactivity coefficients (case of MOX fuel) | Control of power and its evolution during the cycle |
| Pu-238 content (Pu-238/Pu) | ~ 20 % | ~ 80 % |
| Maximum MA content | 1 %<br>(UOX, less with MOX) | < 50 % Np<br>~ 20 % Am |
| Extra enrichment | 1 % (case of UOX) | 1 % (case of Np) |
| Spectrum, fuel type, reactor size effects | To be defined for MOX fuel | - |
| Waste radiotoxicity réduction | Factor ~ 10 to ~ 50 | Factor ~ 10 to 25 |
| Ratio dedicated PWRs/Park | ~ 20 % | ~ 30 % |
| Recycling mode | Multiple recycling | Multiple recycling |
| Consequences on the fuel cycle | n source ↑ (Factor 30)<br>Activity ↑ (Factor 15)<br>(case of UOX) | Specific fuel cycle to be evaluated |
| Experimental validation | Foreseen | Np→Pu-238 production at CELESTIN reactor<br>More foreseen<br>(see § 3) |

Table 4 : Minor actinide burning in PWRs

assumes that the losses during reprocessing are of the order of 0.3 % for Pu and 1 % for the minor actinides, one can achieve a reduction of a factor ~ 100, with respect to the open cycle option, of the radiotoxicity source term in the repository. This factor is consistently similar for all times, if, and this is an important point, Cm (essentially Cm-244) is continuously separated during the multiple recycling, to avoid the build-up of Pu-240 in the wastes. The Cm problem has to be further investigated ; however its temporary storage outside the reactor can result in the production of Pu, which can be re-used after a few decades.
The radiotoxicity reduction of approximately a factor of 100 can be achieved with practically all fission reactors (and recycling modes). As an example figure 1 gives the time evolution of the radiotoxicity source R (t) in the case of a homogeneous recycling of 2.5 % Np and Am in a large oxide-fuelled fast reactor of the EFR type, with the hypothesis of partitioning process losses indicated above.

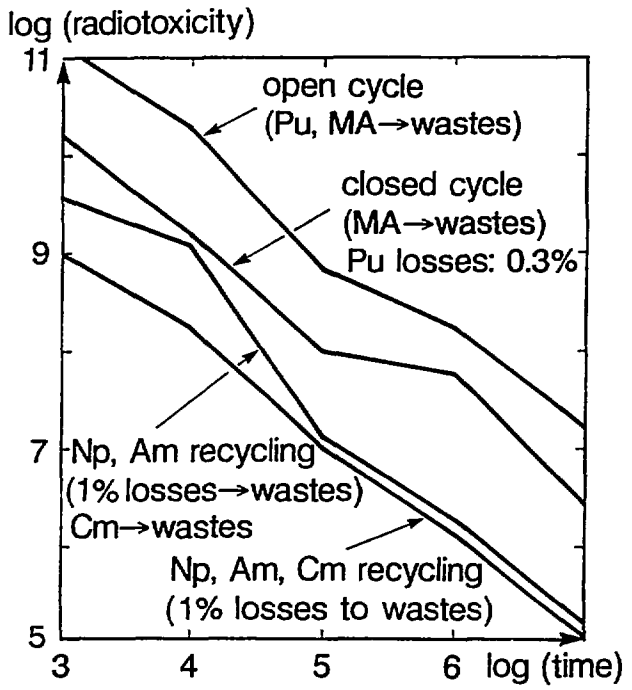


Figure 1 : Homogeneous actinide recycling in an EFR-type reactor
Radiotoxicity reduction

## 3 - FUEL AND TARGET STUDIES

3.1 - Results

The purpose of this research into fuels and targets with a minor actinide base is to confirm that they are eliminated in significant quantities by transmutation in existing PWRs and FRs, with a good in pile behaviour.
Two types of experiment were conducted under conditions similar to those prevailing in the reactors mentioned above although they were not entirely representative of burning-type irradiation.
The minor actinides - Am and Np - were implemented in the form of fuel (with uranium or uranium-plutonium supports) or targets (with inert oxide supports).
The encouraging results obtained for design, fabrication and behaviour under irradiation allowed us to establish a program directed towards high transmutation levels in OSIRIS (PWR) SUPERPHENIX and PHENIX (FR) reactors.

*3.1.1 - Fast reactors*

After separation, long-lived minor actinide type waste can be eliminated using existing technologies. We have seen that the destruction of significant quantities of actinides Np and Am is theoretically possible in fast neutron reactors.
The SUPERFACT experiment, prepared by TUI (Institute of Transuranium Elements - Commission of European Communities, Joint Research Center, Karlsruhe, FRG) and the DEC (Fuel Research Department, Nuclear Reactor Direction, CEA, France) is aimed at demonstrating the feasibility of fabricating and irradiating in FR the minor actinides 237Np and 241Am originating in waste present in spent fuels. Transmutation, that is to say transformation of a long-lived element to a shorter-lived element, is performed by recycling actinides in a so-called "homogeneous" mode. The minor actinides are (Figure 2) :

- either added in large quantities (40 % and 45 %) to $UO_2$ fuel, (type A),
- or mixed in small quantities (2 % by weight) to $(UPu)O_2$ fuel, (type B).

The specifications of PHENIX standard fuel were applied to fabrication of americium and neptunium-based fuels. The wet route GSP (Gel Supported Precipitation) solgel process was chosen for the fuel $(UPu)O_2$ [2] [3]. It avoids manipulations of powders and circulation of dusts which are deposited on glove box surfaces and expose personnel to radiation.
PHENIX specifications were complied with.
The SUPERFACT capsule is a PHENIX standard one (Figure 3) with 19 cladded pins in helium atmosphere and made of cold-worked steel with 15 % Cr and 15 % Ni and added titanium (diameters 6.55 x 5.65 mm and 1793 mm long). The capsule is a subassembly of 19 pins placed in a standard fuel assembly. The actinide-based pins contain fissile columns approximately 400 mm long for type A and 850 mm long for type B as for standard pins.

About metallurgical behaviour, on pin n° 7 a corrosion zone less than 40 microns thick was observed.
The section taken at the same level on the standard pin shows a much larger central hole as a result of the higher linear power and burnup levels (Table 5).
The radial section of high actinide content pins shows no central hole for pins but large porous zones with coalescence towards the center of the pellet - the first indication of the central hole forming. The cracks in the fuel show notable differences. In pin n° 6 they are fine and irregular, but tend to be wide and axial in pin n° 5. Such indications would suggest that the temperature during irradiation was not very high, though higher for pin n° 6 where restructuring is more marked. The gap between the fuel and the cladding (approximately 60 microns) is greater than for 2 % actinide fuels and identical for the two pins observed. No trace of corrosion was observed.

*3.1.2 - Pressurized Water Reactors*

SUPERFACT has provided experimental results under representative conditions (limited burnup) for the behaviour of actinide-incinerating pins under irradiation in a fast reactor. Such results do not exist at the moment for PWRs. The only experiment providing information about neptunium transmutation in thermal flux is an old experiment involving 238Pu production for heart pacemakers. An experimental irradiation was therefore set up (ACTINEAU) in order to fill in the gaps of knowledge.

*Transmutation of Neptunium in CELESTIN Reactors [4]*
In the 1970 a large-scale neptunium transmutation program was carried out in Celestin reactors, with the aim of producing 238Pu for pacemakers.
In the first experiments, the neptunium was in the form of an Np-Al alloy plate (modelled on the standard Celestin fuel consisting of U-Al plates). But use of this material was rapidly abandoned as the intended medical application imposed a very low 236Pu content ($< 1 \times 10^{-6}$), and ($\alpha$, n) reactions with aluminium had a detrimental effect from this point of view.
Subsequently, the entire experiment was concentrated on $NpO_2$ - MgO targets generally with a $NpO_2$ 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 by standard powder metallurgy in glove boxes with lead panels : by a decay, 237Np (which has a very long half-life of $2.14 \times 10^6$ years) in fact becomes 233Pa (whose half-life is only 27 days and which is therefore in equilibrium with its "parent" after a few months). It 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.

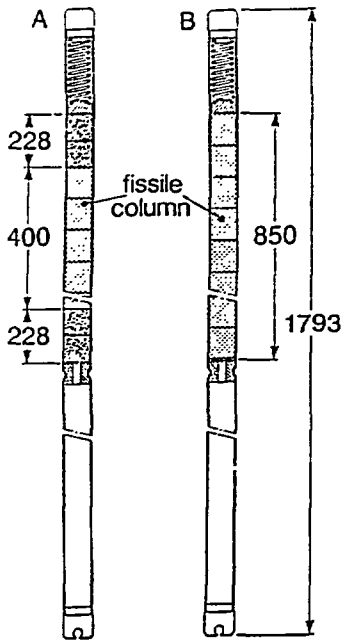


Figure 2
Design of the PHENIX
irradiation homogeneous pins

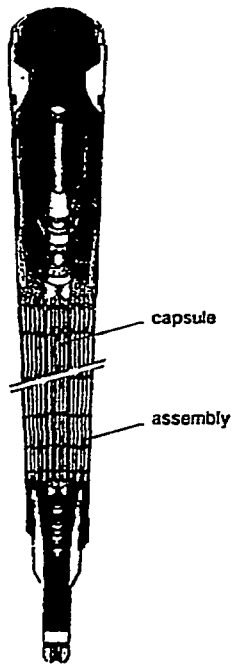


Figure 3
PHENIX irradiation device

| pin n° | fuel | Linear power rating (kW/m) beginning | & end of life | maximum burnup (at%) |
|---|---|---|---|---|
| 4 - 16 | 2 % Am | 38.0 | 32.5 | 6.8 |
| 7 - 13 | 2 % Np | 38.0 | 32.5 | 6.8 |
| 6 - 14 | 20 % Am 20 % Np | 17.4 | 27.3 | 4.3 |
| 5 - 15 | 45 % Np | 20.6 | 28.3 | 4.6 |
| 8 | standard | 43.0 | 37.0 | 8.5 |

Tableau 5 : Irradiation of actinide pins

| pin / gas | Kr + Xe % | He % |
|---|---|---|
| standard | 100 (235 cm³) | 100 (10.3 cm³) |
| 4 | 65 | 387 |
| 5 | 60 | 222 |
| 6 | 57 | 6127 |
| 7 | 65 | 140 |

Tableau 6 : Volume of gases in the plenum

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-$H_2$ gas. The columns produced were 50 cm long, clad in zircaloy and assembled in clusters each containing 16 pins.
These clusters were then irradiated in Celestin reactors for a duration of two cycles, or approximately 100 days in a flux of 2 to $3 \times 10^{13}$ n/cm$^2$/s (a highly thermalized flux as the reactors are moderated by heavy water). The conversion rate of Np to Pu was around 6 % with a Pu-238 isotopic abundance greater than 90 %. Increasing the irradiation time would provide a higher Np to Pu conversion rate but at the cost of the Pu-238 isotopic abundance, which is undesirable.
This program therefore showed that it is possible to implement significant quantities of neptunium and to transmute a significant proportion during a short-term irradiation.
No cladding breach occurred but the irradiation conditions were not very representative of those required for the objectives of the actinide burning program, where the highest possible transmutation rate imposes much stricter conditions of residence time, power and fuel temperature.

3.2 - Fuel and target developments

The SUPERFACT experiment has shown that ITU and CEA laboratories are capable of fabricating fuel pins and the first measurements have not revealed any particular problem relating to behaviour under irradiation, even for fuels with high Np and Am actinide content, which are quite different from standard (UPu)$O_2$ fuel. From the examinations carried out, it can be concluded that under irradiation, the behaviour of fuels with an Am or Np minor actinide base is similar to the standard mixed oxide, at least for a burnup of up to 5 atom %.
The high level of helium production attributed to the production and decay of Cm-242 from americium 241 in the fuel does not alter the behaviour of the pins. Nevertheless, the high pressure levels which occur with prolonged irradiation would certainly affect the mechanical behaviour of the cladding and this point has to be taken into account in pin design (Table 6).
The transmutation program is currently being pursued by studying incinerating reactors [5] in conjunction with the studies of minor actinide-based fuels [6] [7].

*3.2.1- Minor actinides burning demonstration in FR SUPERPHENIX*

Thanks to SUPERFACT operating experience, a Np demonstration on a near-industrial scale has been planned for 1995 in the SUPERPHENIX core.
Irradiation of 150 homogeneous oxide fuel pins (i.e. a half assembly) containing about 2 % Np has been planned for Superphenix core 2.
In the same way, for Superphenix core n° 3 further experiment could be carried out on 4 similar assemblies of Np-based pins for 10 kg Np total amount. 1 or 2 more fuel 2 % Am pins could be also irradiated. Another

option could be irradiaton of homogeneous Np,3 subassemblies and one Np target subassembly representative of the heterogeneous mode by using magnesia as a support structure of Np oxide. The neptunium concentration would be approximately 40 % by weight. This solution would, in fact, require a precursor experiment on fuel pins within the core of PHENIX, in order to determine the metallurgic behaviour of this kind of target under irradiation.

*3.2.2 - Irradiation experiment in PWR (OPERA loop of OSIRIS*)*

The ACTINEAU experiment (ACTinides INcinérés en réacteur à EAU - Actinide Incineration in Water Reactors) is designed to demonstrate the feasibility of minor actinide incineration in PWRs and to study the metallurgic behaviour of the rods with a view to their subsequent optimization.
This irradiation involves the participation of teams from the Departments of Fuel Research, Mechanics and Technology, Experimental Reactor and Reactor Studies of the Nuclear Reactor Direction and of the Transuranium Institute either. It is planned to take place in the OPERA loop of the OSIRIS reactor at SACLAY and should begin when the loop comes into operation in 1994.

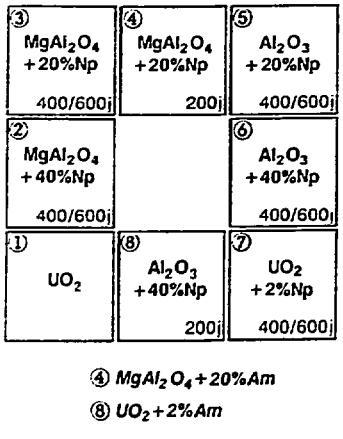


Figure 4
PWR fuel and target study in OSIRIS pile

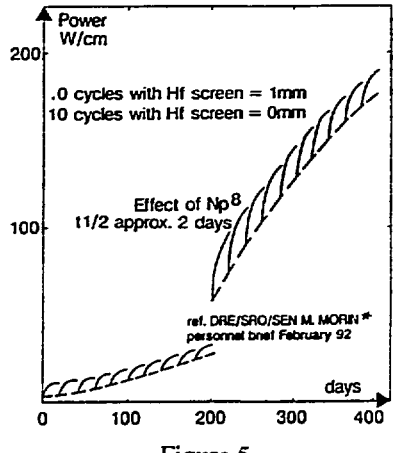


Figure 5
Power evolution $NpO_2$ pin

* Experimental Reactor Department - OSIRIS Reactor Section.

The experiment will investigate the homogeneous concept ($UO_2$ fuel pins enriched with 2 % $NpO_2$ or 2 % $AmO_2$) and the heterogeneous concept (target rods with a higher actinide content contained within an inert matrix).
The first stage consisted in choosing materials best adapted for the inert matrix of the heterogeneous pins. A preselection of oxides, based on cross-sections, material properties and, insofar as it is known, behaviour under irradiation resulted in the following matrices being chosen : Mg $Al_2O_4$ and $Al_2O_3$ (reference materials), Ce $O_2$, $Y_2O_3$ and $Y_3Al_5O_{12}$ (alternative ones). The choice can only be confirmed by the experiment as there are still a number of uncertainties concerning the properties of these materials and their behaviour under irradiation. Moreover, as these matrices pose problems for reprocessing it would be especially advantageous if it provided an incineration rate sufficiently high for the pins to be sent directly for storage.
Work was begun on perfecting the fabrication of $MgAl_2O_4$ - $UO_2$ and $Al_2O_3$ - $UO_2$ pellets (as $UO_2$ has similar properties to $NpO_2$, it is possible, as a first step, to simulate the latter). On the basis of these fabrications, characterization of this composite material can be started and its properties determined.
The percentage of actinides to be introduced into these heterogeneous targets results from preliminary calculations that have been carried out by the OSIRIS Reactor Section of the Experimental Reactors Department on the behaviour of standard-geometry rods (pellet diameter = 8.2 mm) irradiated in the OPERA loop of OSIRIS. The results show that for this type of rod, the mass fraction of $AmO_2$ and $NpO_2$ can scarcely exceed 40 % without posing long-term behaviour problems due to high linear power. Consequently, the grid shown in Figure 4, which is the subject of current neutronic calculations, is being designed for the eight pins used in this experiment.
Figure 5 shows an example taken from the initial development calculations carried out with a $NpO_2$ mass fraction of 20 %. In this example, the first 200 days have been calculated with a hafnium screen which is then removed for the following 200 days. This explains the discontinuity which appears on the curve.
Power, which is initially very low, gradually grows as it results mainly from fission of 238Pu and 239Pu isotopes created by transmutation of neptunium. A further addition to this power comes from fission of 238Np which has an extremely high fission cross-section but very short life (half-life of 2, 3 days). This is shown by the discontinuities on Figure 5 which are caused by the almost complete disappearance of 238Np after each 10-day interrun.
After 400 days, the linear power of the 20 % $NpO_2$ pins remains within the field explored by standard PWR rods and the neptunium transmutation rate is already 40 %. These results are therefore very encouraging : subject to their good response under irradiation, it can be hoped that these composite materials will attain a transmutation rate of 50 % for the first experimental irradiation and if all goes well it should be possible to design a rod on the basis of these results which can reach an even higher transmutation rate.
As part of this experiment it is also intended to irradiate rods containing americium. These will be fabricated at the Institute of Transuranium Elements in Karlsruhe.

## CONCLUSION

The transmutation program of minor actinides and long-lived fission products has expanded considerably at the Nuclear Reactor Direction. This expansion has been accompanied by close cooperation with teams from the Fuel Cycle Direction working on chemical separation.
Research at the Nuclear Reactor Direction is being conducted concurrently in two areas : "reactors and other transmutation systems", and "fuels and targets". Concept and performance validation apply to PWRs and FRs and to the minor actinides, neptunium and americium and fission products.
Encouraging results have been obtained so far which indicate promising openings :

- in FR particularly Np homogeneous recycling,
- Am homogeneous or may be better heterogeneous recycling.

However more studies are necessary in the fields of fuel and target development and performance.

## REFERENCES


[1] H. Küsters, M. Salvatores et al. "Recent investigations in France and Germany on the Transmutation of Actinides and Fission Products in Reactors" Proc. Int. Conf. ANP'92, Nov. 1992.

[2] C. Prunier, A. Chalony, M. Boidron, M. Coquerelle, J.F. Gueugnon, L. Koch, K. Richter, Transmutation of minor actinides : behaviour of americium - and neptunium - based fuels under irradiation. FR'91 Int. Conf. on Fast Reactors and Related Fuel Cycles - Kyoto - Oct. - Nov. 1991.

[3] G. Cogliatti, P. Gerontopoulos, K. Richter, Gel-Suppported Precipitation of $(UPu)O_2$ pellets. Transaction of the ENC 1979. Conf. of ENS, ANS Trans 31, 175 (1979).

[4] B. Chevalier, Etude de cibles au 237Np pour une production de 238Pu industriel dans les réacteurs "Célestin", Mémoire CNAM, 26 mars 1975.

[5] M. Salvatores, C. Prunier, P. Bergeonneau, A. Zaetta, Long lived wastes transmutation studies at CEA, Winter Meeting ANS, Nov. 10-14 1991, San Francisco - USA.

[6] L. Koch, R. Wellum Editors, Proceedings of the workshop on partitioning and transmutation of minor actinides CEC, Nuclear Science and Technology - Report EUR 13347 EN, J.R.C., ITU 1991.

[7] L. Koch, R. Wellum, Workshop on partitionning and transmutation of minor actinides. CEC, JCR, ITU - June 1991 Meeting.