CONCEPT AND EXPERIMENTAL STUDIES ON FUEL AND TARGET FOR MINOR ACTINIDES AND FISSION PRODUCTS TRANSMUTATION

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ABSTRACT

High activity long-lived radionuclides in nuclear wastes, namely minor actinides (Americium and Neptunium) are in large amount generated by current nuclear reactive.

The destruction of these radionuclides is a part of the French SPIN (Partitioning and Burning) programme consistent with the determination to send a minimum amount of harmful products for final storage.

Transmutation concepts are defined for Neptunium and Americium tacking into account fuel cycle strategies.

Neptunium destruction does not pose any major problems. It's a by product of uranium consumption, as plutonium and in despite of a slight gamma activity due to the protactinium 233 it's quite easy to handle.

Diluting Neptunium in the MOX should not be an obstacle for fabrication, in-pile behaviour and reprocessing either. Consequently we make the proposal of homogeneous mode of Neptunium in MOX PWR's fuel which should be soon explored in the experimental OSIRIS reactor and in FR's Phenix and Superphenix.

The analysis is more complex for multi isotope Americium. Its destruction is difficult because of gamma radioactivity which complicates fabrication.

Experiments in Phenix and calculation showed that FR offer a good potential for Americium incineration, but similar datas do not exist for PWR.

It will remain a well know difficulty for fabrication and reprocessing. In this case we have to put a real new face to the fabrication flow-sheet of Americium compounds and we propose to develop the heterogeneous mode.

Targets choice are defined in term of :
- safety, considering fuel reaction with cladding and water sodium,
- transmutation rate, limited by target behaviour, in FR's (Phenix), PWR's (OSIRIS) and HFR (Petten),
- reprocessing, checking the solubility of such targets by Purex process.

So, at the beginning of our programme the account has been on improving fuel and targets properties related to safety and fuel cycle.

In order respond to the public concern about wastes and in particular the long-lived high level ones, a French law issued on December 30, 1991 identified the major objectives of research for the next fifteen years, before a new debate and possibly a decision on final wastes disposal in Parliament (2006).

To comply with the requirements of the law, CEA has launched an important and long term R and D programme. A part of this programme called SPIN [1] is devoted to partitioning and transmutation of these wastes. Once separated, elements have to be mixed with fuel or manufactured into target elements for transmutation.

Present transmutation studies of Minor Actinides (M.A) and long-lived fission products (LLFP) are conducted on a wide range of options :
- fission reactors (FR and PWR),
- M.A. homogeneous or heterogeneous recycling,
- LLFP transmutation in fission reactors.

Fuel studies cover the following areas :
- Fuel fabrication and irradiation for homogeneous recycling Np and Am both in thermal and fast reactors.
- Inert matrices basic research, with fabrication and irradiation for targets in the heterogeneous recycling mode, in particular for Americium.

HOMOGENEOUS RECYCLING

The Superfact experiment has shown on the one hand the good metallurgic performance of M.A fuels under irradiation in the Phenix F.R and on the other hand that the transmutation affected the whole of the fuel without "rim effect" leading to the disappearance of Np and Am actinides of between 25 and 30 % for those burn-up limited to 6 atoms % [2].

Neptunium destruction does not pose any major problems : Neptunium is a by product of uranium consumption, as plutonium and in despite of a slight gamma activity due to the protactinium -233 it's quite easy to handle.

Diluting Neptunium in the MOX fuel should not be an obstacle for fabrication, in-pile behaviour and reprocessing [3] consequently we make the proposal of homogeneous mode with priority to Neptunium in MOX fuel, which will be soon explored.

Figures 1 and 2 show respectively :
- the irradiation capsule for a new SUPERFACT-2 experiment in Phenix FR aiming to reach higher burn-ups (> 10 at %) than the previous SUPERFACT-1 experiment,
- the ACTINEAU experiment related to transmutation studies in PWRs.

Demonstration NACRE experiments are foreseen in SUPERPHENIX which will involve 1 and later 4 full subassemblies, with fuel pins containing ~ 2 % Np homogeneously mixed to the standard MOX FR's fuel. Aim is to reach with M.A homogeneous fuel the same burn-up (at % or equivalent fuel power days) than for the standard MOX fuels.

HETEROGENEOUS RECYCLING

Recycling is more complex for multi-isotope Americium [3]. Its destruction is difficult because of gamma radioactivity which complicates fabrication. It will remain a well known difficulty for reprocessing. In this case we have to put a real new face to the fabrication flow-sheet of Americium compounds and we propose to develop the heterogeneous recycling.

In the SUPERFACT-1 experiment we have used a content of 45 % weight of M.A which represents the heterogeneous recycling.

But in M.A-UO_2 fuels the disappearance of M.A is counter-balanced by the appearance of Pu isotopes throughout the fuel pellets.

Working from this base, we have expanded research on the transmutation of M.A using inert matrices as support in place of UO_2 in order to reduce the production of plutonium.

Targets choice are defined in term of :
- safety, considering fuel reaction with claddings, water (PWR) and sodium (FR),
- transmutation rate, limited by in-pile target behaviour studied in Phenix-FR, HFR Petten and OSIRIS-PWR [4],
- reprocessing, checking the solubility of such targets by Purex process.

Figure 3 shows the irradiation device for inert matrices test : MATINA experiment in Phenix FR launched in 1994.

LLFP TRANSMUTATION

An experiment of Tc-99 irradiation called ANTICORP 1 will start in a subassembly of PHENIX (core periphery) using a moderator to increase the epithermal captures. Figure 4 shows the irradiation device for the ANTICORP experiment.

The irradiation of 3 Tc pins, identical to the samples of the HFR-Petten irradiation [4], will be undergone in 1995 [5]. The fabrication by ITU of the bars should be completed by the beginning of 1995 after a grain structure study of as-casting Tc. For a long irradiation a refined grain structure is researched by increasing the cooling rate during the metal solidification.

On other hand a Tc alloy metallurgy study is launched to stabilize a cubic crystallographic phase by light addition of a cubic metal (Nb, Ti, Mo, V, Cr) in place of the hexagonal Tc system.

In the both case the aim is to avoid an anisotropic swelling during irradiation, well knowed phenomena occurring for coarse grain structure and anisotropic system either.

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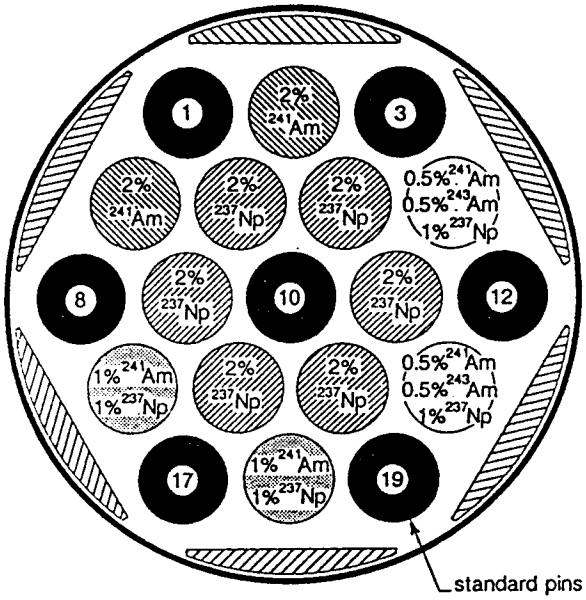

Fig. 1 Neptunium and Americium Homogeneous Concept. SUPERFACT 2 Experiment in PHENIX

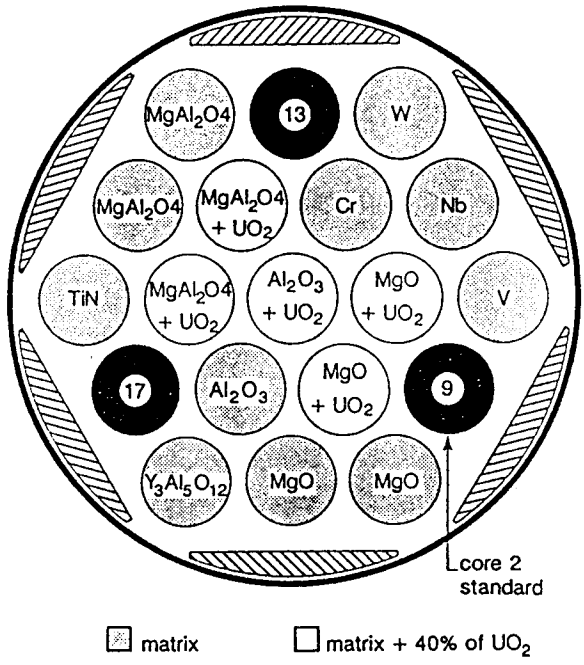

Fig. 3 Heterogeneous Concept. Behaviour of the inert matrix. MATINA Experiment in PHENIX

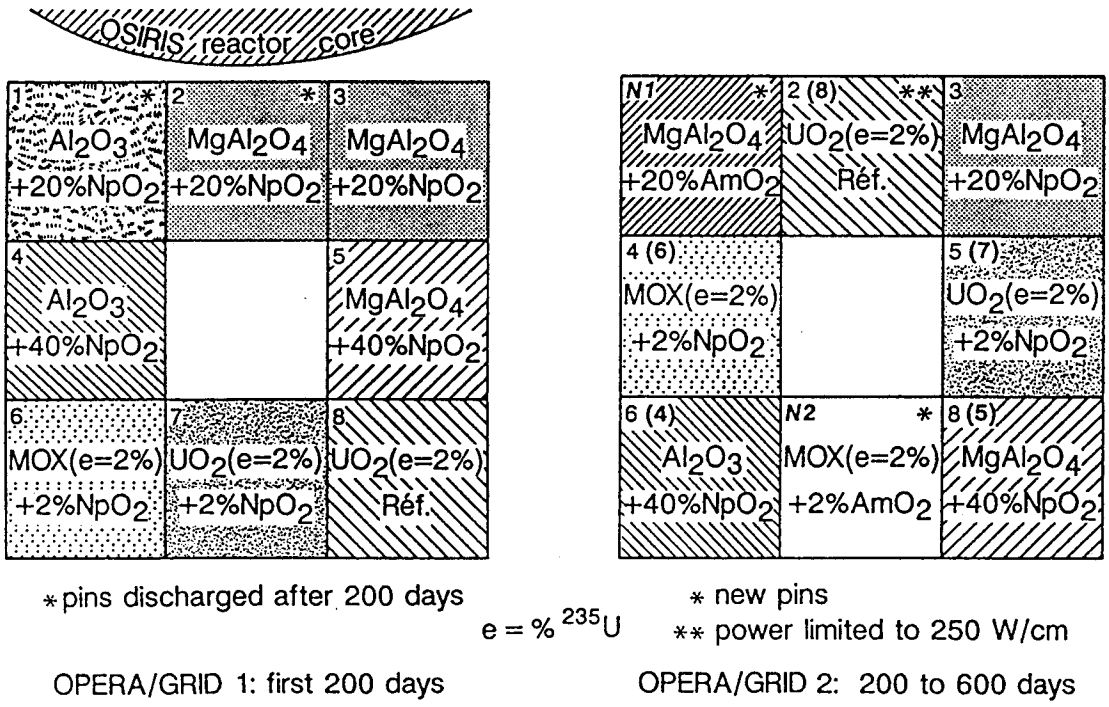

Fig. 2 Neptunium and Americium Heterogeneous and Homogeneous Concepts : ACTINEAU Experiment in the OPERA loop of OSIRIS Reactor. Pins positions in grid 1 (Np) and grid 2 (Am)

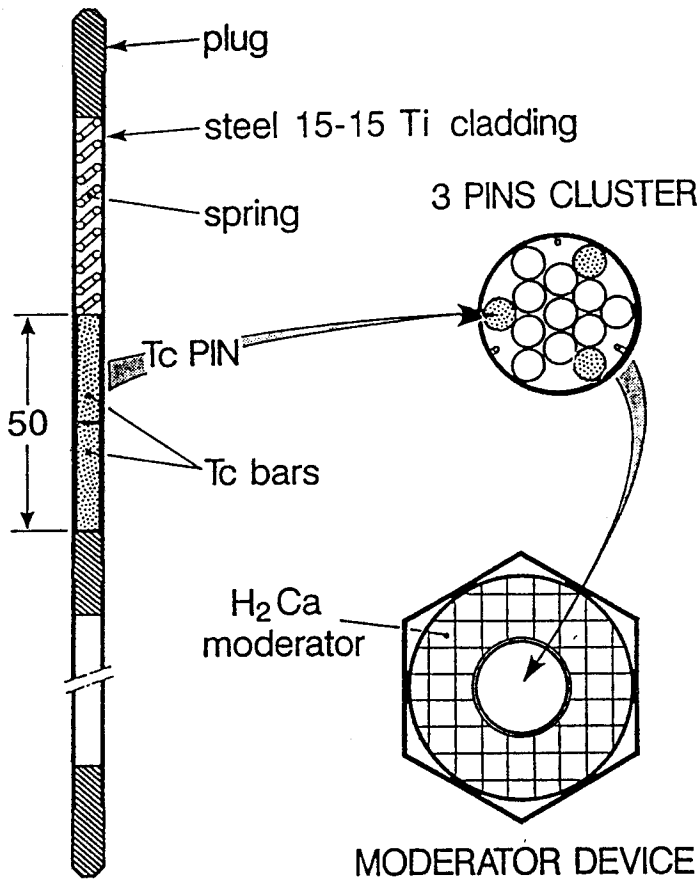

Fig. 4 ANTICORP 1 Experiment in PHENIX