

# Criteria for the Development of Targets and Fuels in Nuclear Transmutation

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## Abstract

A variety of partitioning and transmutation schemes have been recently proposed, which treat an increasing number of radionuclides. Therefore criteria are discussed to select the processes and nuclides on which promising fuel and target development studies are based. A summary of the planned and ongoing studies at the European Institute of **Transuranium** Elements is given.

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## Introduction

The public attitude towards any type of **anthropogenic** waste has led to a ranking of the proposed options according to recent polls in Germany and Japan (Table 1).

Table 1: Public acceptance and classification of processes

<b>generating less waste</b>		
<b>recycle (and reuse) waste</b>	<b>P&amp;T scheme</b>	<b>EFR / IFR with P &amp; T</b>
<b>incinerate waste in ‘ special installations</b>	<b>U/Pu recycling</b>	<b>MAB, ADR</b>
<b>condition waste for geological disposal</b>	<b>direct storage</b>	

Any process which would generate less waste will - not surprisingly - meet the highest public acceptance. The option to recycle and possibly reuse waste would be the second choice. A partitioning and transmutation scheme of nuclear waste could be classified under this option as far as it applies to the fast type of reactor. Here minor actinides can be reused cost-effectively for energy production compared with other schemes relying on special minor actinide burners or accelerator-driven reactors where the energy balance is not so favorable. Therefore this option must be classified as waste incineration in special installation (which e.g. is used for household waste) still being preferred over the conditioning of waste for geological disposal. The direct storage concept for spent nuclear fuel could be classified under this less accepted category.

The situation of the backend of the fuel cycle in the European Community - though different in each of its states - is typically interim storage of spent nuclear fuel for some decades. During this period sites will be explored for geological repositories which are planned to be in operation after the year 2010. Despite the newly built reprocessing plants in UK and France there remains fuel which cannot be (or is not intended to be) reprocessed during this period.

Hence there is still time to see whether research activities in partitioning and transmutation over the next 15 years will reveal that this option could be added to the present fuel cycle. So far two states, Netherlands and France, conduct this type of studies which in the case of France should come to a conclusion by the year 2007. The European Commission supports studies in its indirect and direct action programme in this field.

In the European Institute for Transuranium Elements minor-actinide containing fuels have been developed and irradiated in cooperation with the CEA France in the PHENIX reactor [1, 2]. This activity will be extended to other fuel concepts. In a commercial agreement minor actinide fuel based on Zr-alloy is studied for CRIEPI [3].

### Fuel design constraints

The present discussion on the future of nuclear energy generation can be described by two extreme scenarios (Table 2).

Table 2: Scenarios

1. Phasing-out of nuclear energy production	Transmutation by LWR and/or ADR
2. Breeding of Pu for nuclear energy production	Transmutation by FRB

Under the first scenario one assumes that there is a phasing-out of the nuclear energy production and that it is successfully replaced by another technology. During this phase the fissile material used for military purposes has to be treated as well. If under such a scenario transmutation shall be employed it certainly would use the LWRS because funding to develop and deploy other reactor types would meet strong public reluctance. However LWRS have limited capabilities for transmutation of actinides and hardly any for fission products. Therefore accelerator-driven reactors (ADR) might be used. Some of them would have the ability to destroy the actinides even more rapidly than LWRs [4].

In the other scenario the generation of nuclear energy continues and because of limited U resources of low cost sooner or later breeding of Pu is needed to fuel future reactors. Since this breeding requires fast reactors, they could be used with advantage for transmutation purposes, too.

In this case the situation for the EC is quantified in Fig. 1, with assumed losses of 0.2% of Pu to the geological repository [5].

To develop a fuel for the purpose of transmutation, constraint have to be observed.

Because of reactor safety criteria the concentration of minor actinides in any fuel matrix has to be limited for several reasons. For instance during irradiation a linear power increase will occur because of the build-up of fissile material with high fission neutron cross-sections. If the present cladding type is employed its interaction with the new fuel has to be studied. In case of cladding rupture the minor actinide content of the fuel may change its compatibility with the coolant. In order to optimise the fuel fabrication one has to consider the advantages and

the disadvantages of the homogeneous and heterogeneous minor-actinide fuel concept e.g. to minimise the occupational dose. Already at an early stage the impact of a new fuel type on the existing reprocessing schemes has to be assessed. In the past there are several examples where fuel was developed and even used but which later turned out to be too difficult to be reprocessed (thorium-oxide fuel, carbide fuel, etc.). Since the success of any partitioning and transmutation scheme is strongly bound to the reprocessing losses one has to pay the greatest attention to these problems. Finally the aspect of nuclear material safeguards has to be evaluated to see whether the new fuel concept would increase the diversion risk.

Table 3: Constraints for fuel design

Reactor safety	linear power, fci, coolant compatibility, etc.
Fuel make-up	cost, occupational dose, etc.
Reprocessing	cost, losses, toxic waste, etc.
Nucl. Mat. Safeguards	diversion resistance, etc.

### Example of criteria for fuel development

In the following some of the major criteria in developing a minor actinide mixed oxide fuel for fast reactors will be described. The considerations are limited to the performance of the fuel in the reactor.

The objective is: to design a nuclear fuel which under normal or ramp operating conditions of a power station shows no rupture of the cladding. If such an incident occurs the fuel should not be dispersed through the coolant in the reactor.

In order to assess the fuel behaviour there are two possibilities

- to compute the fuel behaviour from its basic thermal physical data, ,
- to simulate the fuel behaviour under artificial experimental conditions:

Usually both ways are considered. The basic data needed for the fuel are: the phase diagrams of the fuel (Fig. 2), the oxidation potential of the fuel (Fig. 3) and the thermal conductivity (Fig. 4) [61].

The following discussion is based on the four fuel types irradiated in the SUPERFACT irradiation experiment for which the composition is given in the headline of Table 4. In case of a fuel rupture the fuel itself must resist a reaction with sodium. A crucial experiment of the fuel specimen therefore was to expose the fuel under reactor operation conditions to the Na coolant. In Fig. 5 the example of a  $(U_{0.5}Am_{0.5})O_x$  and a  $(U_{0.6}Am_{0.2}Np_{0.2})O_x$  fuel with varying O/M ratios is given. In the case where the fuels have a stoichiometry of 1.884 and of 1.990 one observes a strong attack of sodium which leads to a disruption of the fuel pellet. From this experience and taken into account the reduced thermal conductivity of the Na resistant UAmO fuel of an O/M ratio of 1.83 the irradiation of  $(U_{0.5}Am_{0.5})O_x$  having the standard fuel dimension of this fast reactor was rejected. To adjust the oxidation potential of any fast reactor fuel so that it does not react with sodium the fuel is sintered in argon containing 5% hydrogen. This results in an O/M ratio as given in Table 4 [6].

**Table 4: Fabrication data of MA fuel to be irradiated in PHENIX**

Fuel composition	$U_{0.74}Pu_{0.24}Np_{0.02}O_2$	$U_{0.74}Pu_{0.24}Am_{0.02}O_2$	$U_{0.55}Np_{0.45}O_2$	$U_{0.6}Np_{0.2}Am_{0.2}O_2$
thermal treatment of GSP particles	4000C / 2h argon 400°C / 2h air  850°C / 2h air 850°C / 2h Ar5%H <sub>2</sub>	400°C / 2h argon 400°C / 2h air  900°C / 4h air 1050°C / 2h Ar5%H <sub>2</sub>	400°C / 1h argon 4000C / 4h air  865°C / 3h air 865°C 14h Ar5%H <sub>2</sub>	400°C / 1h argon 4000C / 4h air  900°C / 2.5h air 900°C / 3h Ar5%H <sub>2</sub>
pressing	5.1-6.4 t/cm <sup>2</sup>	4.4- 5.6 t/cm <sup>2</sup>	6.1 t/cm <sup>2</sup>	6.1 t/cm <sup>2</sup>
free density	58.6% TD	68.0% TD	55.6% TD	56.4% TD
sintering	1620°C / 6h Ar5%H <sub>2</sub>	1620°C / 6h Ar5%H <sub>2</sub>	1620°C / 6h Ar5%H <sub>2</sub>	1620°C / 6h Ar5%H <sub>2</sub>
pellet $\phi$	5.363 ± 0.052 mm	5.417 ± 0.029 mm	5.518 ± 0.027 mm	5.434 ± 0.013 mm
pellet density	.97.5 * 0.8% TD	96.8 ± 0.790 TD	95.1 ± 0.6% TD	95.9 ± 1.0% TD
D/M	1.973	1.957	1.996	1.928
X-ray diffraction parameter	5.4578 ± 5	5.4570 * 5	5,4545 * 5	5.4735 ± 5 5.4696 ± 5

The examples show how the operating condition of the fuel determines its final composition and demonstrate that e.g. the irradiation of an  $(U_{0.5}Am_{0.5})O_x$  is not feasible. Of course there are other constraints to be observed such as fuel swelling which is difficult to simulate other than under irradiation. Under the assumption made for the four fuel types experienced, the fuel behaved as expected under the irradiation conditions as shown in Fig. 6. No unusual fuel swelling or fuel-clad-interaction was observed.

## Conclusion

The design of minor-actinide containing fuel is an interdisciplinary task where the impact of the minor actinide on the reactor operation as well as the material performance of the fuel itself and with its cladding have to be seen together with the unavoidable need to reprocess and remake the material. In the case of the example given here of minor-actinide containing fuels in an oxide matrix this task has been solved so far successfully.

## References

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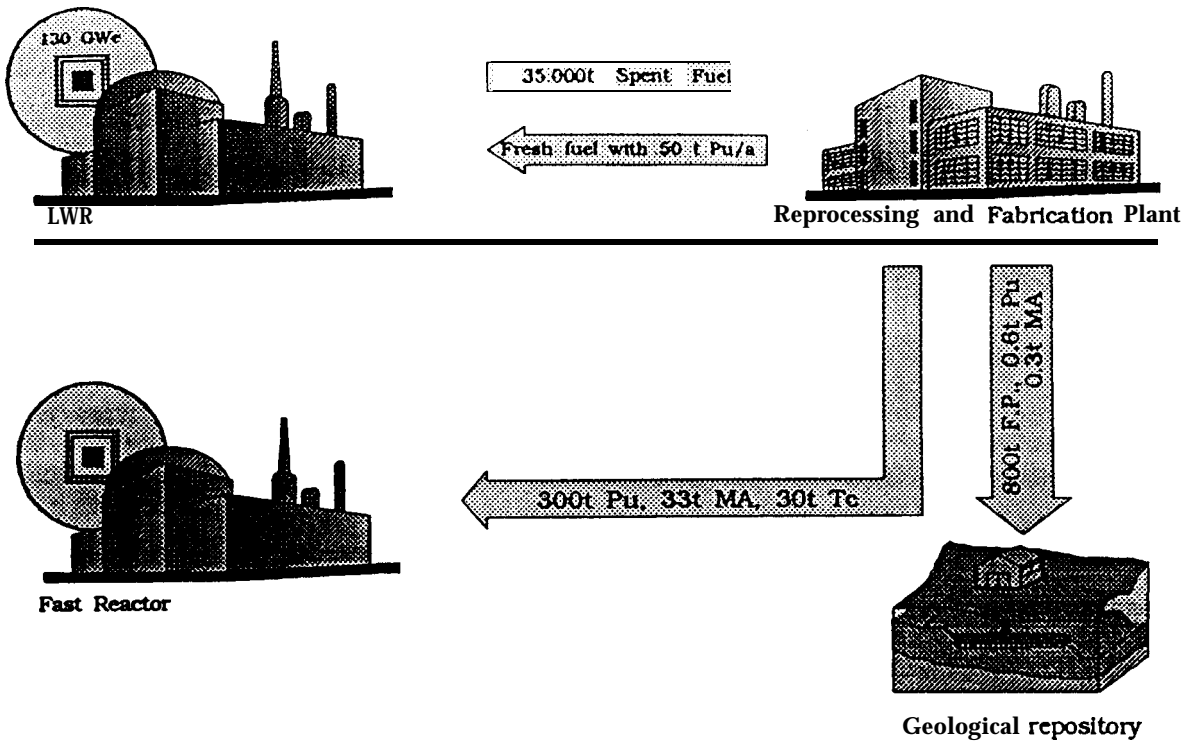
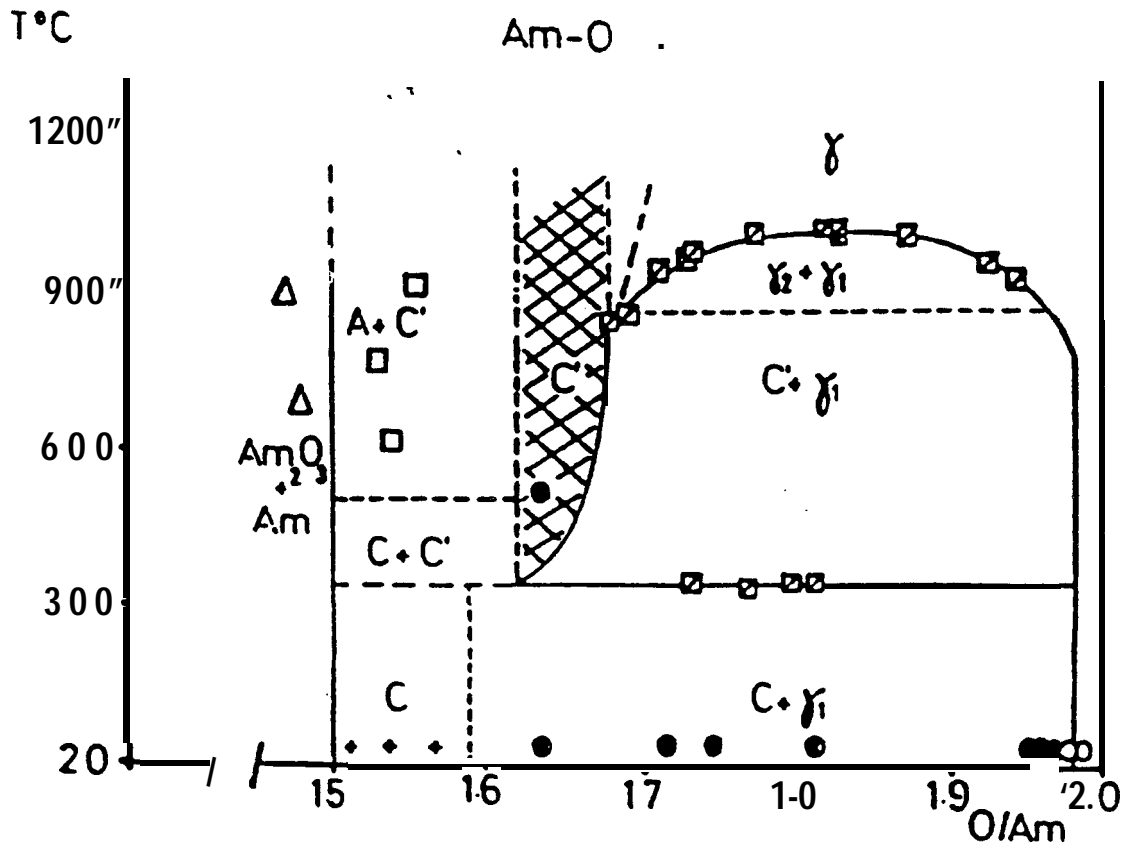


Fig. 1 MA + F.P. Transmutation by transition from LWR to FR



A = hexagonal  $Am_2O_3$ , C = low temperature bcc  $Am_2O_3$ ,  
 C' = high temperature bcc  $Am_2O_3$ ,  $\gamma$ ,  $\gamma_1$ ,  $\gamma_2$  = fcc  $AmO_{2-x}$ ,  
 □ = results of DTA measurements

Fig. 2 low temperature Am-O phase diagram

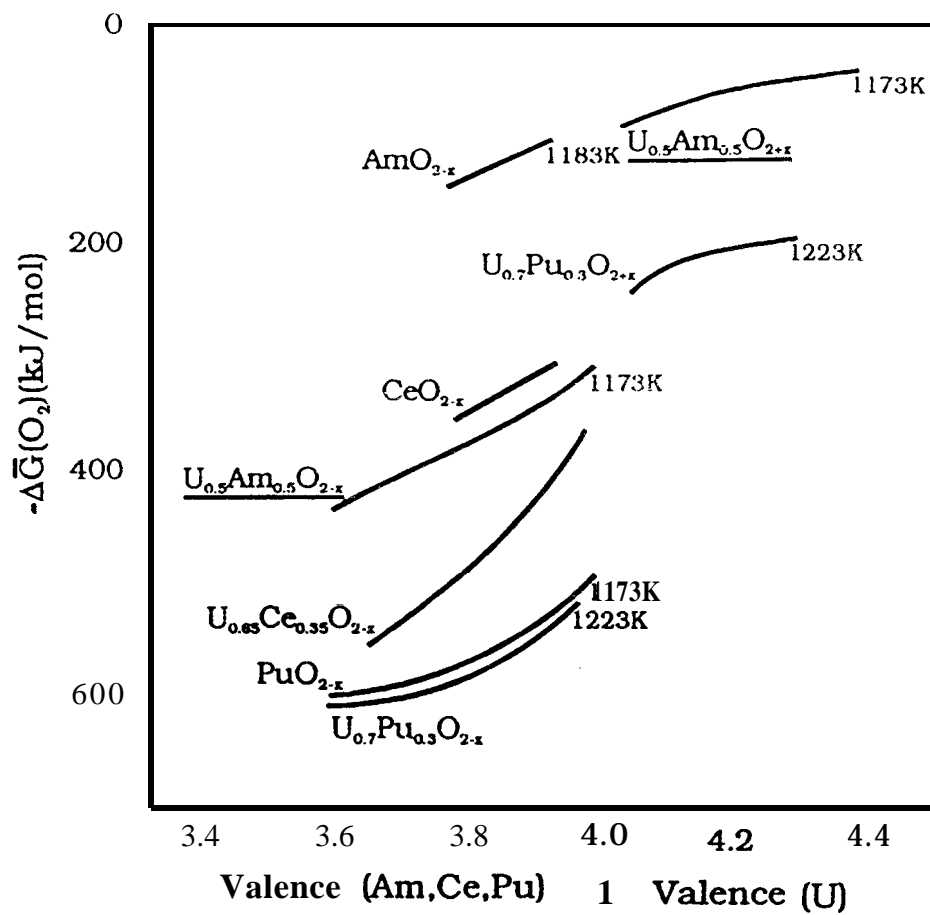


Fig. 3 Oxidation-potential of  $\text{U}_{0.5}\text{Am}_{0.5}\text{O}_{2-x}$  compared to other oxides

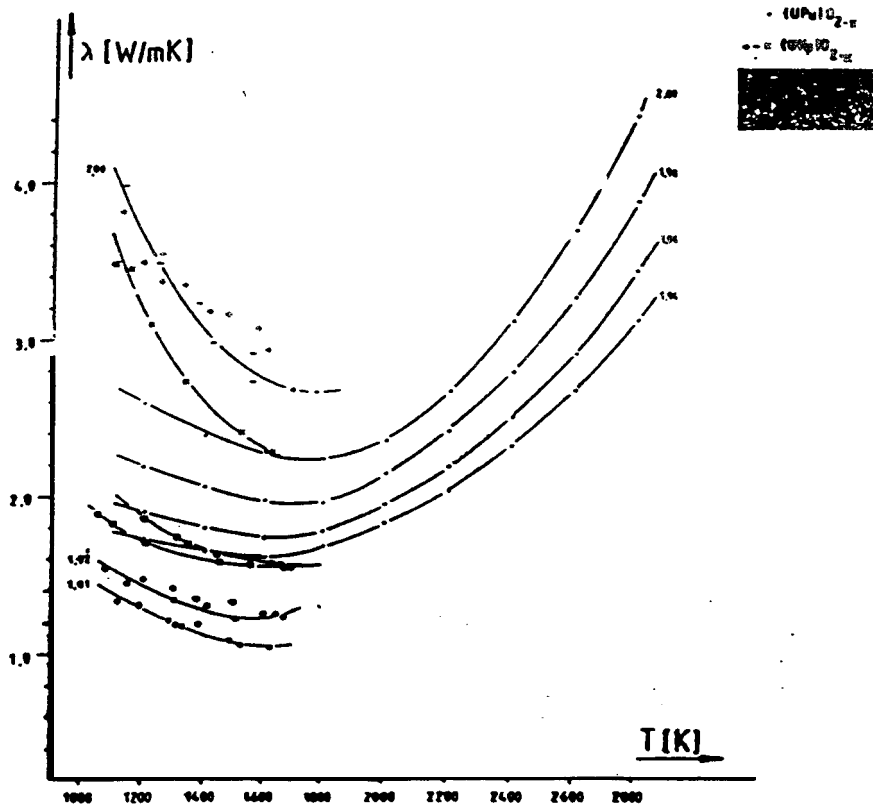
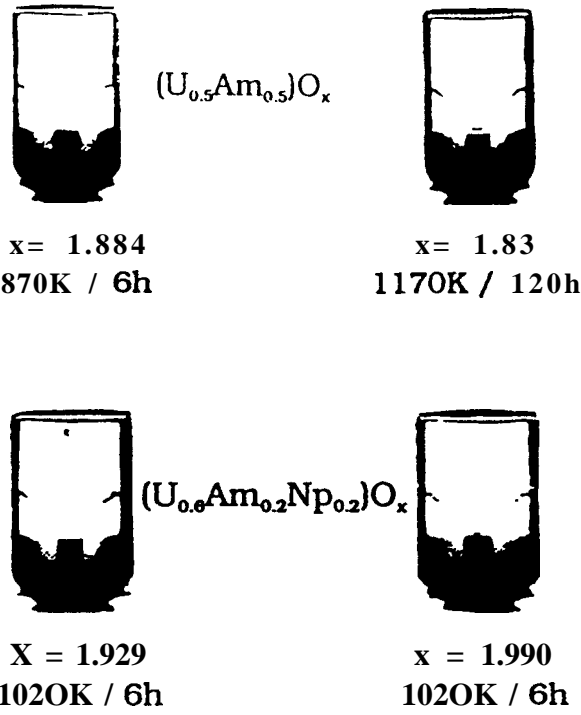
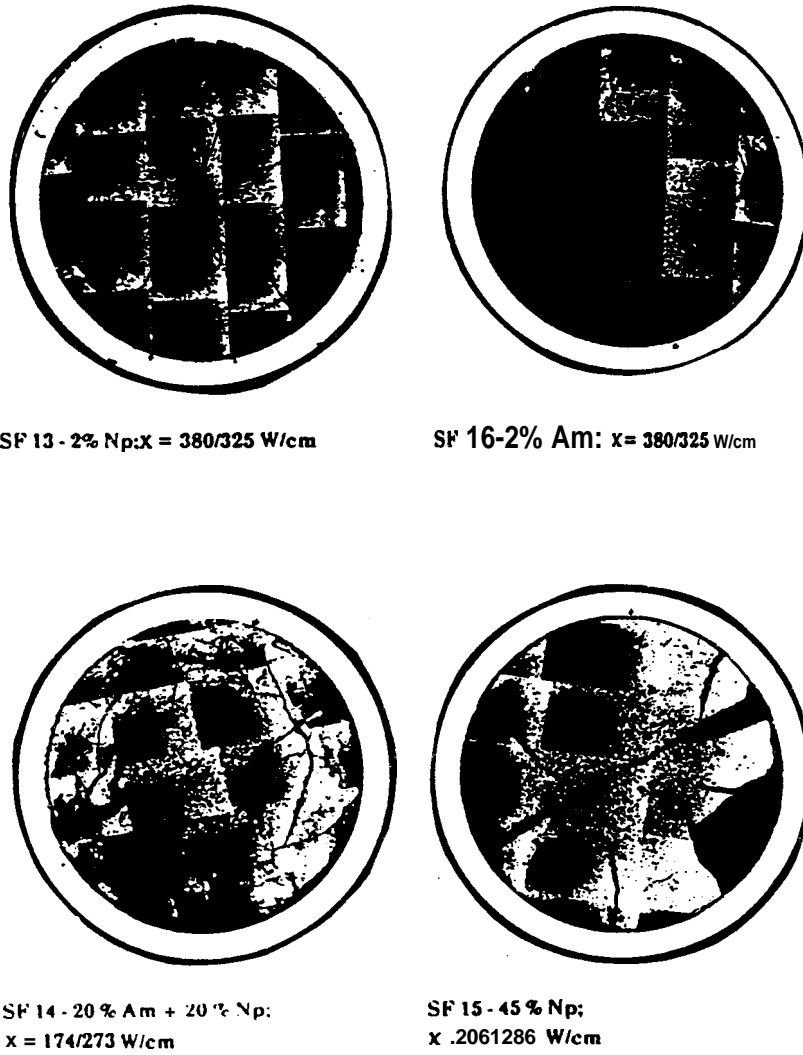


Fig. 4 Thermal conductivity of various mixed actinide oxides as function of temperature

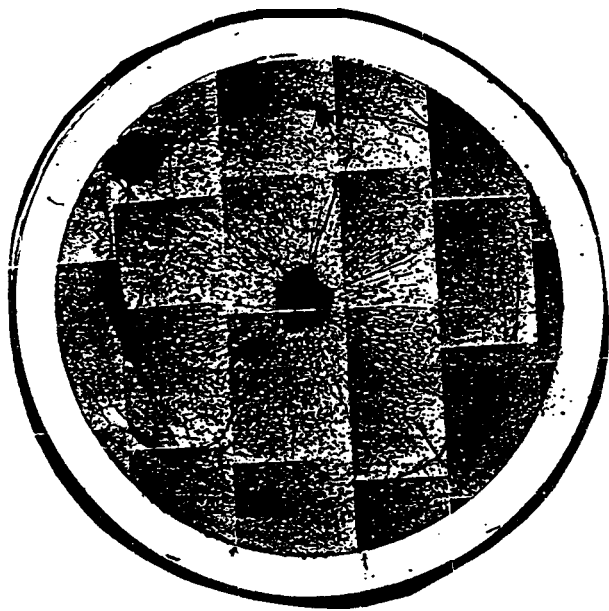




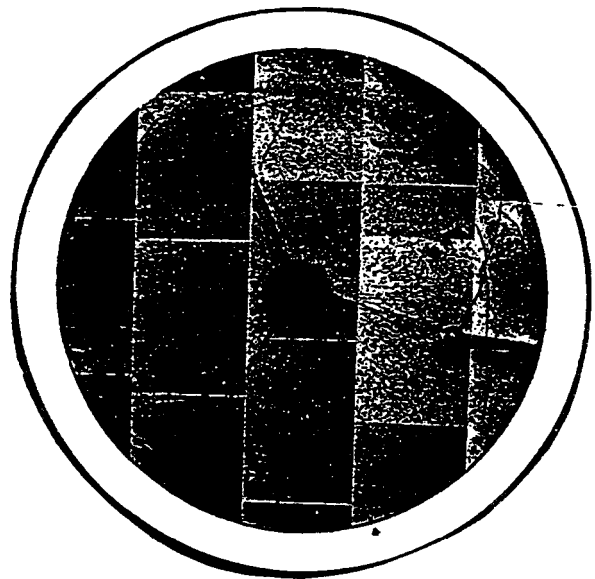
**Fig. 5 Simulated compatibility experiments between mixed Am oxides and Na**



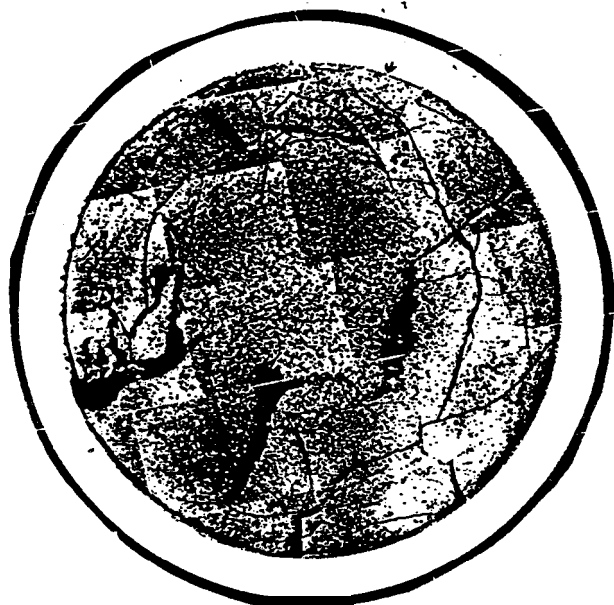
**Fig. 6 Microscopy of the SUPERFACT fuels**



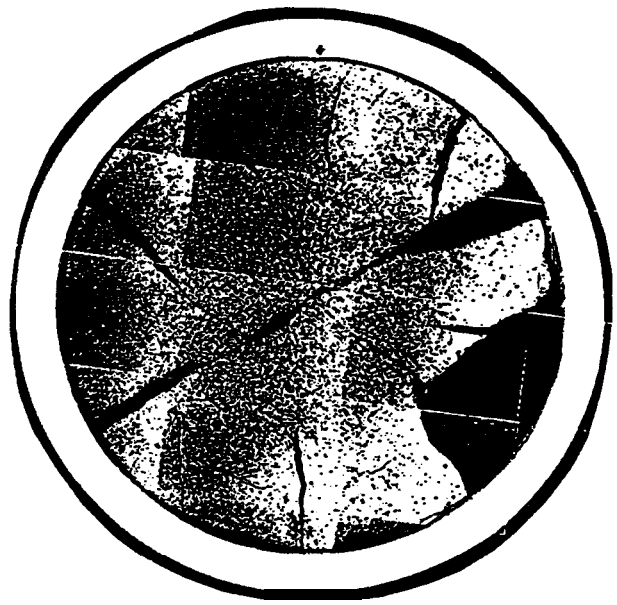
SF 13 - 2% Np;  $x = 380/325$  W/cm



SF 16 - 2% Am;  $x = 380/325$  W/cm



SF 14 - 20% Am + 20% Np;  
 $x = 174/273$  W/cm



SF 15 - 45% Np;  
 $x = 206/288$  W/cm

Fig. 316