ELSEVIER

Journal of Alloys and Compounds 271–273 (1998) 517–521

Journal of ALLOYS AND COMPOUNDS

Nuclear fuels for actinides burning CAPRA and SPIN programmes status

A. Languille*, P. Millet, J. Rouault, S. Pillon

Commissariat à l'Energie Atomique de Cadarache 13108 Saint Paul-Lez-Durance, France

Abstract

The on-going R&D programmes on Fast Reactors fuels (CAPRA, SPIN, . . .) are defined to explore a large variety of possible solutions in accordance with options adopted for management of Pu, Minor Actinides and others long-lived Fission Products. Innovative nonconventional fuels are studied in the frame of various international collaborations. A new area of research has become highly effective, in particular the study of nonfissile materials to be used as matrices for these fuels. © 1998 Published by Elsevier Science S.A.

Keywords: Fast reactors; Fuels; Transmutation; Matrices; U-free fuels

1. Introduction

Fifty years of Fast Reactor fuel development have seen fluctuations of the various fuel types; starting from oxide fuel in the first reactor of Fermi, to emphasis on metal fuels, thence to ceramics as the objective and the performance demands have changed. But the last two decades of prototype Fast Breeder Reactors operation have provided a sound basis of knowledge particularly on oxide fuel behaviour. In the early 1990's, the main objective of fuel development was the achievement of very high burnup from an economical point of view (reduction of the fuel cycle cost).

The French waste management R&D law of 30 December 1991 calls for research into actinide transmutation (one of the three research axes foreseen). For this purpose, the programmes CAPRA and SPIN are underway at CEA. New nonconventional fuels are studied in the frame of European and International collaborations.

In this paper, we will review the fuel design principles on the basis of the conventional FBR oxide fuel pin experience and by comparison we will describe the peculiarities of innovative fuels.

2. Survey of «classical» FBR fuel design principles

2.1. Generic features and specifications

A typical FBR core includes the nuclear fuel (mixed oxide), control rods, the uranium dioxide blanket which enables the reactor to breed plutonium (neutron capture by the fertile isotope ^{238}U to form fissile atoms, mainly ^{239}Pu) and the liquid sodium coolant. The fission cross-section is smaller than in a thermal reactor. The consequences of these features on the fuel itself are the following:

1. Higher enrichment in the range 15–20% Pu (compared with few % ^{235}U in a pressurised water reactor PWR),
2. high neutron flux which induces structural damage,
3. high specific power rating (~1500 W cm^{-3}) and high heat flux to be removed from the fuel pin (~200 W cm^{-3}),
4. high oxide temperature (~2000°C) even under steady-state operation.

Fuel pin and subassemblies must be designed in accordance with the requirements of the different steps in the whole fuel cycle (fabrication, in pile performance, handling and reprocessing). Safety requirements are also taken into account at the design stage. A fuel pin consists of a stack of cylindrical pellets in a sealed cladding tube which is the first barrier from the safety point of view against fissile material and fission products releases. Almost all fission gases produced are released from the fuel and trapped in the plenum to limit the pressure stresses in the pin.

2.2. Limiting phenomena

The phenomena which limit the performances of the fuel

*Corresponding author. Tel.: +33 4 42 25 72 65/27 22; fax: +33 4 42 25 47 47.

0925-8388/98/$19.00
PII S0925-8388(98)00139-X

are mainly the irradiation damage of the structures and the fuel-cladding interactions (chemical and mechanical) [1].

2.2.1. Irradiation damage on structures and fuels

Under fast neutron flux, the motion, annealing, coalescence and trapping of neutron induced defects (vacancies and interstitials) are responsible for the phenomena of swelling, irradiation creep and embrittlement. The swelling is due to diffusion speed difference between vacancies and interstitials; consequently vacancies gather together and tend to create bubbles which can grow later on. Irradiation creep is more complex: it is a temperature independent phenomena (irradiation-induced creep) which plays a great role in stress relaxation. Irradiation defects strengthen the lattice which induces embrittlement.

2.2.2. Fuel-cladding chemical interaction

The pin cladding may be corroded locally by caesium and tellurium (both produced by fission). The corrosion reaction is controlled by the oxygen potential of the irradiated oxide and is dependent on local temperature but also of the burn-up.

2.2.3. Fuel-cladding mechanical interaction

The fuel swells with B.U mainly due to the difference between the volume of solid fission products produced and the volume of plutonium atom fissioned; the retained fission gas contributes to a minor part of the swelling. The volume increase estimated for an oxide is about 0.7%/ha%. The resulting cladding stress remains low under steady state conditions thank to thermal and irradiation fuel creep and also cladding deformation if sufficient void is available (smear density influence). But during power transients, the stresses may reach much higher values which are liable to exceed the cladding yield stress and consequently lead to a pin failure.

Table 1 shows the limiting phenomena and variables to be taken into account for the main design parameters: Linear rate, cladding temperature, burn-up and dose.

2.3. Fuel pin performance

2.3.1. Fuel thermal performance

The radial temperature distribution can be calculated in a cylindrical element, neglecting axial heat flow; the thermal conductivity of the fuel is temperature dependent. The maximum linear rate is determined as a design limit in order to prevent fuel melting in any situation (taking into account uncertainties). Descriptive laws have been established on the basis of the experience gained in reactors.

At beginning of irradiation, the mixed oxide is restructuring due to temperature gradient effects inside the fuel. The pelleted fuel, for example, is cracked radially very early, the central zone is modified in elongated grains (columnar zone) and the closure of the pellet-cladding gap occurs. The result of these modifications is the formation (or the increase) of the central hole.

2.3.2. Fuel pin mechanical design

The pin is subjected to a complex system of stresses: Primary stresses resulting from pressurisation by fission gas released from the fuel (linearly increasing with B.U) and secondary stresses (thermal stresses, swelling gradient stresses, bundle-duct interaction, fuel-cladding interaction).

In the past, the damage fraction was used to evaluate the pin reliability (cumulating thermal creep damage and instantaneous plastic deformation). Currently, the mechanical design of pin and S/A is based on the RAMSES rules (mechanical criteria).

2.4. Limit of the conventional fuel

The extensive worldwide irradiation experience on mixed oxide fuel $(UPu)O_2$ provides a good confidence to reach the required high burn-ups. In fact, no intrinsic limitation has so far been identified and today the main limiting factor is still the irradiation damage on the structures. A large part of the current research programs is devoted to the optimisation of wrapper and cladding materials able to reach high doses without excessive swelling while keeping acceptable mechanical properties.

Different classes of structural materials are currently studied: The optimised austenitic steels (130/150 dpa), the advanced austenitic steels as low chromium steels (150/180 dpa), the ferritic-martensitic steels ($>$200 dpa).

For a Fast Breeder Reactor, the objective was to reach safely and reliably a peak burn-up of 20% ha with a fuel residence time of more than 1500 EFPD (~5 years)

Table 1
Relationship between in-pile phenomena and fuel design

Design parameter	Limiting factor	Main variables
Linear rate	Fuel melting	Fuel type
	BOL FCCI	Pellet geometry
Cladding temperature	Cladding creep	Cladding material
	FCCI	Fuel type
Burn-up	Fuel swelling	Fuel type
	FCCI, FCMI	Smear density
Dose	Cladding and Wrapper Tube deformation-S/A bowing	Cladding and Wrapper materials

representing a structural damage of 180 dpa. This objective is assessed for the fuel but not for the structures.

3. New trends in FR fuels from CAPRA and SPIN programmes

3.1. The CAPRA programme

The objective of the CAPRA programme is to assess the feasibility of a Fast Reactor optimised to burn as much plutonium as possible but which also contributes to the destruction of Minor Actinides. The achievement of an attractive plutonium burning rate needs the *use of a fuel with a high Pu content* that necessitates a drastic fuel inventory decrease in a given core volume (dilution concept) [2].

Since 1993, the greater part of the effort is focused on the mixed oxide reference option for which an attractive Pu burning rate of ~70 Kg/TWeh can be reached; the need of such a high Pu content fuel has led to some peculiarities of the fuel pin design (see Table 2).

1. The choice of a maximal plutonium content of the oxide of 45% Pu is based on the search for a compatibility with the current fuel cycle technology (dry route fabrication and PUREX process reprocessing),
2. a small diameter fuel pin containing fuel pellets with a large central hole (low smear density).

The properties of mixed oxides are fairly well known for Pu contents of up to 30% and have been extrapolated up to 45% Pu. An out-of-pile programme is underway (for example, diffusivity has been measured which shows a good agreement with the extrapolated data).

For an application in a large core size (1500 MWe), it is necessary to reduce the fuel inventory by a factor 2 (which leads to a reduction of the fuel residence time). For that, heterogeneous bundles with a large number of pins (of which one third are empty of fuel and filled with an inert material) and unfuelled diluent subassemblies in the core are required. This heterogeneity brings also a large flexibility (from the safety point of view, for accommodation of a large range of plutonium qualities, . . .).

U-free fuels are considered for a maximum level of Pu burning (i.e ~110 Kg/TWeh) for the use of very degraded Pu vector qualities in a dedicated reactor. The required reduction of fuel inventory leads to the use of innovative designs in which the compound is diluted within a new material either in the form of a dispersed fuel or of a solid solution.

3.2. The SPIN programme

In the SPIN programme, the possibility in the long term, for the minor actinides (Np, Am, Cm) to be recycled and burnt in reactors is studied. There are two methods to burn minor actinides in nuclear reactors (see Table 2) [3].

—The first one is to mix them directly within the standard fuel itself, this is called «homogeneous recycling». Only a small content of MA's can be loaded in the reactors because of the characteristics of the core (reactivity loss reduction over the cycle, degradation of the fuel temperature coefficient, . . .).

—The second one is to concentrate the MA's in targets (high content of MA's) in specific locations of the core, for example S/As in the periphery of the core, with a small impact on the core characteristics. The so-called «heterogeneous recycling» is currently the recommended way for the highly active americium. Two options can be foreseen: The multirecycling where the americium is burnt by successive irradiations in targets which are reprocessed and the «once-through recycling» where more than 90% of the americium is transmuted in a single irradiation.

Table 2
Main features of fuel elements for breeder and burner FRs

Reactor / Parameters	FBR PHENIX	FBR SPX	FBR EFR	Pu BURNER CAPRA	Once through SPIN	Multirecycling SPIN
Fuel element						
Fuel type	$(UPu)O_2$	$(UPu)O_2$	$(UPu)O_2$	$(UPu)O_2$	AmO_2+MgO	AmO_2+MgO
Pu (Am) wt.%	21.3–27.0	15.0–18.8	21.0	43.0–44.7	10.0–20.0	20.0–40.0
Pellet diam. (mm)	5.42	7.14/2.0	6.94/2.0	5.27/2.16	5.42	5.42
Fuel height (mm)	850.0	1000.0	1000.0	1000.0	(1000.0)	(1000.0)
Smear density (%)	88.0	83.0	84.0	75.0	70.0–90.0	70.0–90.0
Core conditions						
Max lin. Rate ($W\ cm^{-1}$)	450.0	470.0	480.0	500.0	100.0–200.0	40.0–100.0
Nom. clad. Temp (°C)	650.0	620.0	630.0	630.0	<550.0	<550.0
Max Burn—up (%ha)	14.0	8.0	20.0	20.0	70.0–90.0	30.0–40.0
Max dose (dpa)	90	83.0	190.0	124.0	130.0–200.0	~200.0
Residence time (j)	690.0	640.0	1500.0	855.0	>3000.0	~1500.0

The actinide incineration performances are expressed in terms of rate of consumption where the element is burnt by fission and by neutron capture during the irradiation period. The fission rate is quite high in a fast neutron spectrum, from 20 to 30% (an intrinsic advantage of FRs in comparison with PWRs). In both cases, the targets containing minor actinides require the development of new designs where the americium compound is dispersed in an inert matrix (a CERCER type compound) as for the CAPRA U-free fuels.

3.3. Studies and validation of the fuels options

3.3.1. On the CAPRA oxide reference fuel

The oxide reference fuel must be compatible with the PUREX process. This has been demonstrated on fresh 45% Pu content fuel but with slow dissolution kinetics; this solubility must be confirmed by tests on irradiated fuels.

With respect to the irradiation behaviour, thermomechanical calculations have been performed on a CAPRA oxide fuel pin; the results show a satisfactory behaviour of the fuel in comparison with a conventional fuel. Nevertheless, some points connected with this 45% Pu fuel need further confirmation.

The Pu redistribution: The thermal gradient in the pellet leads to a radial redistribution of the U and Pu in the fuel as the result of two phenomena: Evaporation/condensation in the pores in the beginning and solid state diffusion during the irradiation period. This Pu redistribution depends on the initial stoichiometry (O/M) and also on the Pu content of the fuel.

The large central hole: Although this gives advantages for thermal behaviour, it is necessary to preserve the fuel stack integrity in the beginning when the fuel cracking occurs. The good thermomechanical behaviour of the fuel at BOL must be confirmed.

The inner clad corrosion: The most important factors affecting the internal corrosion are mainly stoichiometry, clad strain and irradiation conditions; the effect of plutonium content remains of secondary concern but we must check the acceptability of the expected corrosion.

The heterogeneous design of the bundle with a great number of pins is also a point to be validated. The thermal–hydraulic behaviour of such a S/A must be checked.

An important irradiation programme has been launched to provide the first results necessary for the evaluation of the fuel options taken for the oxide reference option. The reactors involved are SILOE, HFR, PHENIX, SUPER PHENIX and BOR60 [4]. A few irradiation experiments are completed and first results are available. On the IFOP experiment, a single pin containing 45% Pu content fuel was irradiated at the end of 1994 in the SILOE reactor for a short time at 1.15 ha%; the PIE results show a global good behaviour, in particular concerning the integrity of the fuel column. A second experiment, TRABANT1 in the HFR reactor, has been completed. Only the first nondestructive results are available today.

3.3.2. On CAPRA U-free fuels and SPIN targets

Unlike the oxide option for CAPRA, these innovative options are at the exploratory stage and a first selection of the potential candidates has been required to restrict the R&D studies to a reasonable level. This selection is based on criteria involving their neutronic, physical and chemical properties, their fabrication possibilities, their irradiation behaviour and their reprocessing capabilities [4].

About the choice of the compound itself, for the FRs application, the more relevant plutonium compounds considered are the plutonium oxide and the plutonium nitride, the best one remaining PuN (PuO_2 is incompatible with the PUREX process and it reacts with sodium). Very few data are available for americium compounds and basic knowledge does not allow us to assess oxide (AmO_{2-x} or Am_2O_3) and nitride (AmN) compounds for the FRs application.

The selection of inert matrices according to a certain number of criteria has been made. For FRs, oxide matrices (MgO, Y_2O_3, Al_2O_3, $MgAl_2O_4$, $Y_3Al_5O_{12}$) and nitride matrices (TiN, YN, ZrN, and CeN) seem to be good potential candidates.

Combining the compound and matrix candidates, one can define different classes of U-free fuels and Am targets: Pure actinide compounds (PuO_2, PuN, AmO_{2-x}, Am_2O_3, AmN), solid solution materials (PuZrN for example), the solid solution being formed by a chemical reaction between the compound itself and the inert matrix, and two-phase materials (CERCER or CERMET) of oxides or nitrides.

Neutronic calculations have been performed with modules from the ERANOS code system [5] and the very first results show different trends depending on the neutron spectrum. Comparing for example a target ($AmO_x + MgO$) containing 20 wt.% Am in a moderate spectrum and in a fast spectrum, the evolution of parameters during irradiation such as Am transmutation, Pu and Cm build-up, linear rate, are quite different.

With respect to material studies, the peculiarities of designing such a target or U-free pin concern different aspects:

First, there are different sources of damage in such a composite: fission products and a-decay cause damage inside the matrix very close to the actinide compound, and the neutronic flux damages the whole matrix. The contribution of these component damages depends on the spectrum type (moderate/fast), the material itself, and the mode of dispersion of the particles. The size of the actinide compound particles is also of great importance. This evaluation of damage is a completely new field of activity to be developed in the short term.

Secondly, the composite thermal behaviour of such a dispersed fuel must be evaluated; first thermal calculations have been performed using the 2D (R, θ) finite elements code CASTEM 2000. Many parameters are sensitive in this evaluation among them the actinide content in the composite, the distribution and size of the actinide particles, the thermal conductivity of the damaged matrix, the conductance between the actinide compound and matrix, ...

Thirdly, as the fission rates are very high for the actinide transmutation, helium and fission gases will be produced in large quantities in the pin which must be designed specifically (sufficient gas plenum volumes to have acceptable primary stresses).

Finally, high cladding doses corresponding to long residence times are required in the range of 130 up to 200 dpa to reach high fission rates (effect more pronounced in case of a fast spectrum). This high doses objective requires the development of new swelling resistant cladding materials such as ferritic steels or ODS (oxide strengthened materials).

Various studies are underway in order to better understand the irradiation behaviour of such fuels or targets: theoretical studies, out-of-pile tests, simulation tests, material characterisation but also an irradiation programme. First simulation tests on inert material samples irradiated in the accelerator GANIL (Caen/FRANCE) simulating FP damage show clearly large differences of behaviour between various matrices. The irradiation programme has been launched to provide the first results necessary for a restriction of the selected candidates and their relevant optimisation. A very few irradiation experiments have been completed and first results are available. In particular, two pins after 60 EFPD of irradiation in PHENIX of the MATINA experiment (test of different matrices with or without fissile particles (UO_2)) have been unloaded and the PIE are in progress.

4. Conclusion

The R&D programmes on FRs CAPRA and SPIN fuels have been defined for a few years and very first results have been currently obtained. We have seen that some objectives of conventional FR fuels are still required for actinides burners and a continuous effort on generic fuel studies has to be made in this area. But complementary R&D studies have to be performed for these new nonconventional fuels and new areas of research must be undertaken (damages in inert matrices, . . .). An important programme (irradiation, simulation, theoretical studies) aiming at qualifying some options or screening the more innovative ones is underway. The first significant feedback is expected by 2003–2004 (before 2006 corresponding to the French Parliament debate foreseen in the 91 law).

References

[1] R. Lallement, H. Mikailoff, J.P. Mustellier, J. Villeneuve, Fast breeder reactor fuel design principles and performance, Nuclear Energy 28(1) (1989) 41–49.
[2] A. Languille, J.C. Garnier, P. Lo Pinto et al., CAPRA core studies—The oxide Reference Option, Global'95, September 11–14, 1995, Palais des Congrès, Versailles, France pp. 874–881.
[3] Viala, M.S., The SPIN program, Global'95, September 11–14, 1995, Palais des Congrès, Versailles, France pp. 118–131.
[4] N. Cocuaud, E. Picard, R.J.M. Konings, A. Conti, Hj. Matzke, Inert matrices, U-free Pu fuels and Am targets—Synthesis of CAPRA, SPIN and EFFTRA studies, Global'97, October 5–10, 1997, Yokohama, Japan.
[5] N. Chauvin, J. Tommasi, V. Georgenthum, G. Ritter, Neutronic and fuel studies for americium target design, Global'97, October 5–10, 1997, Yokohama, Japan.