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Fast Breeder Reactor Fuel Performances [and Discussion]

R. Lallement, A. Tuzov and K. Q. Bagley

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Fast breeder reactor fuel performances

By R. Lallement

Commissariat à l'Énergie Atomique, 31–33 *Rue de la Fédération*, *Boite Postale* 510, 75015 *Paris*, *France*

Achieving high burn-ups of the fuel used in fast-neutron reactors is essential for the economy of the breeder system. The mixed-oxide fuel (UO_2, PuO_2), which is generally used, enclosed in metallic pins assembled together inside an hexagonal wrapper, suffers high irradiation damage in a very hostile environment (high temperature, severe temperature gradients, corrosive atmosphere) for long periods of time.

We review the physical phenomena that are in action during the lifetime of a fuel assembly and evaluate the predictable performances of mixed oxide breeder fuel.

Five factors are specially described: fuel swelling, fuel cladding interaction, clad swelling, fuel behaviour under transients and subassembly behaviour in a reactor core.

1. Introduction

The energy produced by the fast breeder reactors (FBRs) comes from the fission of fissile ^{239}Pu induced by fast neutrons. This type of reactor does not include a neutron moderator. Consequently, the energy of the neutron remains high (their speed is fast), and the fission cross section is thus smaller. To produce the fission reaction one must increase the concentration of fissile material.

Several important consequences arise:

(*a*) the fuel (generally uranium–plutonium dioxide) is highly enriched (15–18% ^{239}Pu compared with about 3% ^{235}U in a pressurized water reactor (PWR));

(*b*) the neutron flux is high and has sufficient energy to cause displacement damage;

(*c*) the rather high concentration of fissile material induces high specific power ratings and high heat flux to be removed from fuel elements (*ca.* 200 W cm^{-2} or three times higher than in a PWR);

(*d*) to carry away this high flux a liquid metal coolant, with high thermal conductivity, is required (sodium has been generally chosen);

(*e*) even though the efficiency of cooling is very good, the fuel temperature remains very high in the centre of the fuel pins (*ca.* 2000 °C in steady-state conditions, liable to exceed 2500 °C during operational transients);

(*f*) the fuel cladding temperature reaches 650 °C in steady state conditions, which severely restricts the choice of cladding materials (generally high-strength stainless steel).

Associated with these severe physical conditions are several advantages, which are among the great assets of FBR.

1. As there is a high concentration of fissile material in the fuel, the fuel can reach, under proper design, a very high burn-up.

2. The thermodynamic efficiency of an FBR plant is relatively high (40%) because of the high coolant outlet temperature (545 °C in Superphénix).

3. With proper density of the fuel and the provision of a uranium dioxide blanket, the reactor can 'breed' plutonium (i.e. produce plutonium through neutron captures by the fertile isotope ^{238}U).

The fuel specialist must find the design that enables the fuel to reach high burn-up without any fuel failure. Fuel failures have always to be avoided in any type of reactors to avoid contamination of the primary circuit. This concern is of utmost importance in FBR because sodium reacts with the chosen fuel (mixed oxide (MOX)).

This is a difficult aim and the specialist will need to understand and master the behaviour of fuel, fuel pins and fuel assemblies, under the extreme conditions of high temperature, high strain and high flux, which have been listed above.

More than two decades of prototype and demonstration FBR operation with high load factors and no major incidents have demonstrated that excellent fuel performances are achievable.

2. Main physical phenomena affecting the in-pile fuel behaviour

2.1. *Description of fuel pins and subassembly*

A fuel pin consists of a stack of cylindrical pellets placed in a sealed cladding tube, which forms the first barrier against possible dispersion of fissile material and fission products. Almost all fission gases formed are released from the fuel and trapped in plenum of adequate volume to limit pressure stresses (up to 7 MPa in the case of Superphénix). Other stacks of uranium dioxide pellets are placed in the blanket zones at either end of the fissile column. (See figure 1.)

Bundles of fuel pins are placed inside a hexagonal outer wrapper tube. The fuel pins are separated from one another to ensure adequate coolant flow by a helical spacer wire attached to the end plugs of each pin (as in Phénix and Superphénix), or by spacer grids similar to the ones found in PWRS (as in the Prototype Fast Reactor (PFR) design).

2.2. *Fuel pellet in-pile behaviour*

2.2.1. *Fuel restructuring*

The fuel pellet temperature in normal operation ranges from about 800 °C on the surface to over 2000 °C at the centre. The temperature and thermal gradient are such that they induce significant oxide restructuring. (See figure 2.)

Micrographic examination of an irradiated fuel element cross section shows the following modifications:

(*a*) the fuel is cracked radially, with the cracks annealed at the centre;

(*b*) the central pellet zone structure consists of highly elongated (columnar) grains;

(*c*) the initial pellet-cladding gap has largely disappeared, and a chimney has formed at the centre of the pellet stack.

The high temperature of the fuel results in an almost complete release of fission gases. Significant material transfers may even occur by sublimation in the annular fuel column when the core temperature exceeds 2400 °C.

2.2.2. *Fuel swelling*

Under steady-state conditions, the MOX fuel swells, at a rate estimated to be about 0.7 % per atom percent fissioned.

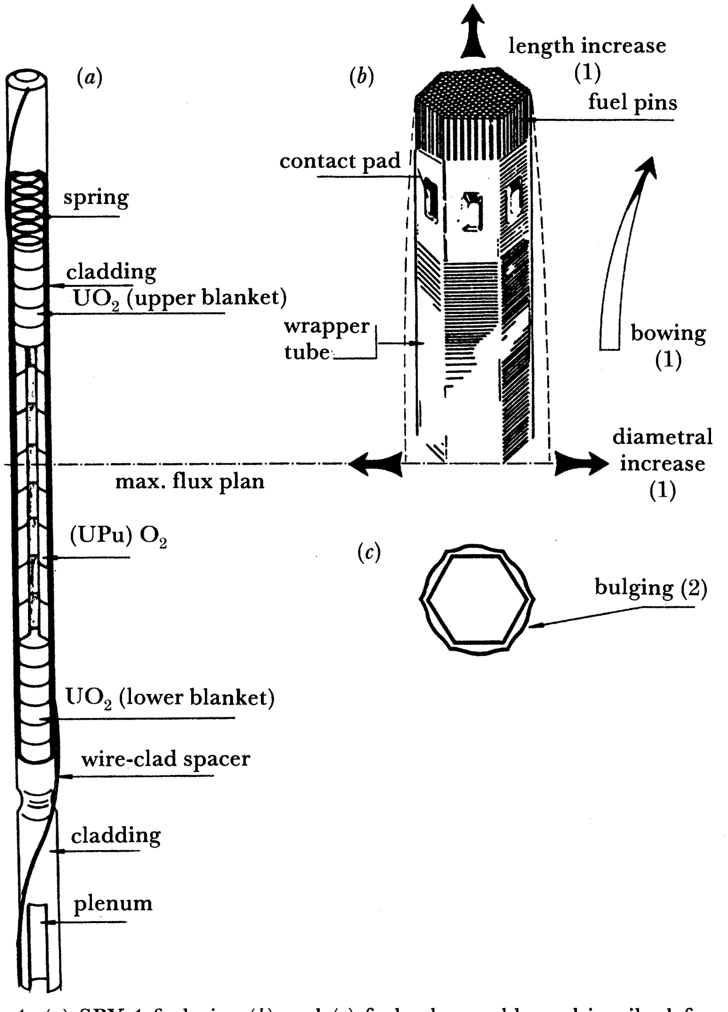

FIGURE 1. (*a*) SPX 1 fuel pin; (*b*) and (*c*) fuel subassembly and in-pile deformations: (1) by swelling, (2) by irradiation creep.

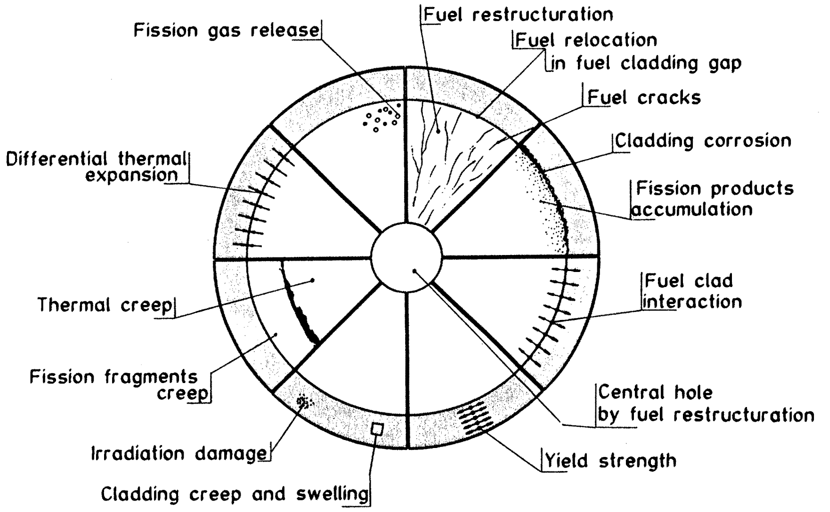

FIGURE 2. Phenomena affecting the in-pile fuel behaviour.

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This is a result of the difference between the volume of fissile plutonium and the volume of the solid fission products. This is also due to the gas bubbles produced by the coalescence of gaseous fission products. The first term is difficult to overcome. The other term can be minimized by the choice of a proper microstructure of the fuel and depends also on temperature and time.

2.2.3. *Fuel creep*

When stress is applied to a lattice, the lattice tends to deform through creep. Deformation becomes easier when atoms can move easily. The existence of vacant sites helps the displacement of atoms from one site to another. An atom can jump from one place to the other easily if there is an adjacent vacant site. Generally, vacant sites are created through heating. Normal creep is temperature dependent. (See figure 3.)

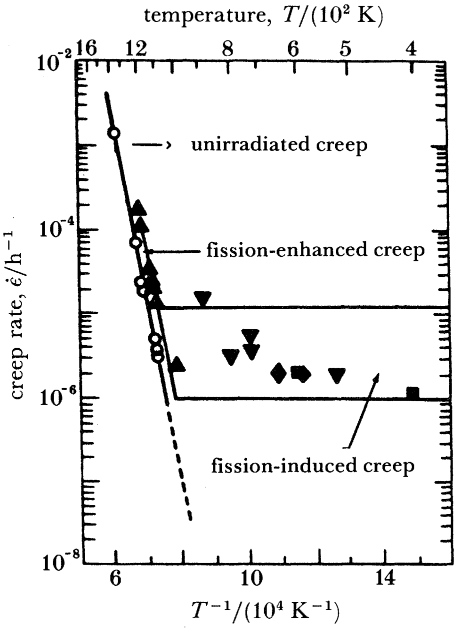

FIGURE 3. In-pile creep of oxide fuel. Creep rate normalized to σ = 24 MPa and $F = 1.2 \times 10^{13}$ fissions cm^{-3} s^{-1}. (From Olander 1976.)

However, irradiation (particularly fission fragments), creates many vacancies so that an irradiated material is similar, as far as the concentration of defects is concerned, to a material exposed to high temperatures. Consequently, even at low temperature, irradiated material can creep easily. This phenomenon is called temperature-independent irradiation creep (irradiation-induced creep), and plays an important role in fuel and fuel cladding deformation.

Fuel pellets are thus submitted to thermal creep in the centre of the pellet and to irradiation creep at the periphery.

2.4. *Fuel changes of composition*

When plutonium is fissioned, each fissile atom gives birth to two different atoms. Those atoms have a size different from the initial plutonium atom (which gives rise to swelling), but have also a different valency.

When a careful calculation is made of the number of existing atoms after fission, taking into account their nature and solubility in the matrix, one finds that the mean valency of the dissolved fission products is lower than that of fissioned plutonium. So the valency of the remaining plutonium increases to maintain the electrical neutrality of the crystal. Fission induces a continuous increase of oxygen potential, which is of utmost importance when considering chemical interaction between fuel and cladding.

2.3. *Cladding in-pile behaviour*

2.3.1. *Irradiation damage*

Metallic structures tend to swell and become brittle when subjected to a fast neutron flux. Up to now the swelling phenomenon in metals has been the main limiting factor in the performance of fuel, especially with regard to high burn-up.

As the neutron energy exceeds the atom displacement threshold, each initial fast neutron impact results in a chain reaction of interatomic collisions, the total effect of which depends on the neutron flux and energy spectrum and thus on the core size.

As a consequence of these collisions many atoms are displaced from their equilibrium lattice positions. (The average number of displacements from a lattice position suffered by each atom is used to characterize the level of material damage and is given as displacements per atom (DPA).) Vacant sites are created and atoms are left in interstitial positions. The resulting defects (vacancies and interstitials) have a tendency to move to restore a more stable state. They either diffuse towards existing defects and recombine and so disappear, or collect to form more extensive defects.

The motion of irradiation-induced defects, their annealing, coalescence or trapping, are the fundamental reasons for the major phenomena encountered in metallic structures and fuels, namely embrittlement, swelling and irradiation creep.

Embrittlement. Irradiation-induced defects coalesce and form microstructural changes that considerably strengthen the lattice. This lattice hardening reduces plastic deformation and induces embrittlement. However, the precise type of defect (dislocation loops, dislocation structure, voids) is strongly dependent on the irradiation temperature. In addition, in some alloy systems such as stainless steels or nickel-based alloys, the irradiation-induced precipitation of second-phase gases and the precipitation of transmutation-produced gases such as helium must be considered. So, the overall effect on material ductility is dependent on several parameters as well as the neutron flux, i.e. temperature, time, stress and metallurgical parameters.

Swelling. Interstitials and vacancies do not have the same speed in the lattice. Interstitials move and disappear quicker. Consequently, vacancies gather together and tend to create bubbles that induce swelling in the cladding. (See figures 4 and 5.)

This swelling problem has been the great scientific and technological challenge during the past 15 years. It seems that some solutions have been found which minimize it, through careful choices of specific materials (stabilized stainless steels, high-nickel alloys, ferritic steels). Nevertheless experiment has shown that swelling was a threshold phenomenon, with an incubation dose. As long as you have not reached the target dose, you cannot be sure that your chosen alloy, with good initial performance, may not start to swell at higher-damage dose.

Creep. The same phenomena, which have been described above for the fuel pellet, are valid

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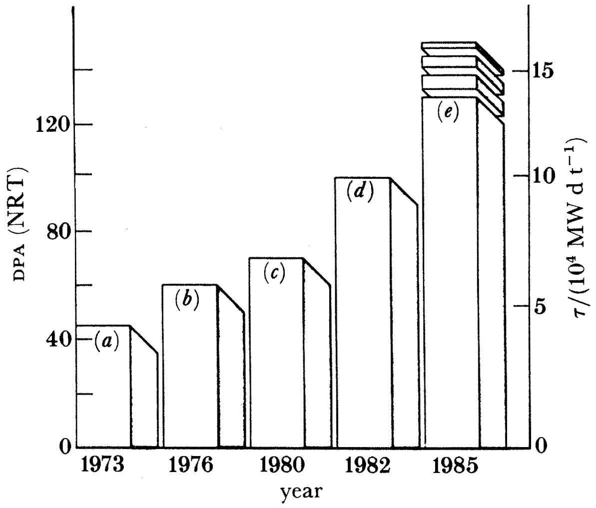

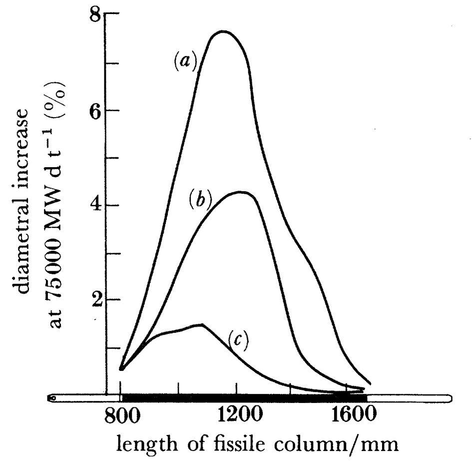

FIGURE 4. Improvement of Phénix driver fuel element cladded with stainless steel. (*a*), (*b*) Solution treated; (*c*) cold worked; (*d*) stabilized; (*e*) optimized. The project year is 1973, and the first core was introduced in 1976.

FIGURE 5. Reduction of cladding deformation by modifying metallurgical specifications of material. (*a*) 316 solution treated (1976); (*b*) 319 cold worked (1978); (*c*) 316 stabilized (1980).

for the metallic cladding, which undergoes thermal creep and irradiation creep, due, in this case, to the defects created by atomic displacements.

2.4. *Fuel cladding interactions*

2.4.1. *Cladding corrosion*

External corrosion by sodium. Because sodium is used as the reactor coolant, reactor materials must be able to withstand contact with sodium at temperatures of up to 700 °C. The choice of materials is limited to stainless steels (austenitic and ferritic alloys) and high-nickel alloys, which sustain only slight dissolution weight losses at a rate low enough to prevent problems.

Corrosion by fuel and fission products. The pin cladding may be corroded locally by caesium and tellurium produced by fission. These elements tend to migrate and are concentrated around the fuel periphery, mainly in the cold part of the fuel column. The principal corrosion reaction is:

$$Cs_2Te + (UPu)O_2 + (O_2)_{fuel} + (M)_{cladding} \rightarrow Cs_2(UPu)O_4 + MTe.$$

This reaction is controlled by the oxygen potential of irradiated mixed oxide. So it is dependent on the local fuel surface temperature and also on burn-up. The burn-up effect is fundamental, because, as we have said above, fission induces a continuous increase of oxygen potential. (See figure 6.)

Owing to this increase of oxygen potential with burn-up, the corrosion by tellurium becomes possible as irradiation proceeds. Post-irradiation examination of irradiated fuel elements has shown an increasing corrosion depth with burn-up, reaching 30% maximum of the cladding thickness in some cases. But it should be emphasized that cladding corrosion is not systematic, although it becomes increasingly likely at higher burn-ups.

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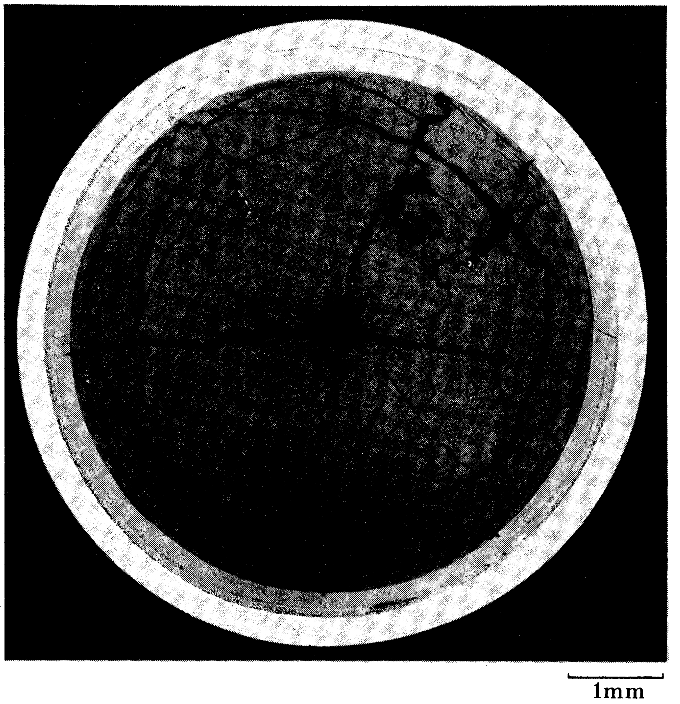

Figure 6. Example of large localized corrosion at high burn-up.

2.4.2. *Fuel cladding mechanical interaction*

Two types of interaction are liable to occur. Under steady-state conditions, the mixed oxide fuel swells as fission causes a volume increase estimated to be about 0.7 % per at. % burned. The resulting cladding stress level remains low owing to thermal and irradiation fuel creep and cladding swelling.

During fast power transients the stresses may reach much higher values able to exceed the elastic limit of cladding material, resulting in cladding failure.

2.5. *Thermal resistance of the fuel–cladding gap*

The thermal flux and the total thermal resistance between fuel and cladding determines the centre temperature of the fuel.

One can easily see how difficult it is to calculate it precisely because of restructuring, corrosion of the cladding, accumulation of fission products at the periphery of fuel, differential swelling between fuel and cladding, and possible degradation of the thermal conductivity of the fuel with burn-up.

2.6. *Sodium–MOX interaction*

When compared with other fuel (like metallic or carbide fuels) MOX has the great disadvantage of reacting chemically with sodium. This fuel would have been rejected for that reason, had it not the great advantage of having a very low swelling rate.

When there is a cladding failure, sodium enters the pin, reacts with fuel, which induces change of composition (formation of sodium uranates), of volume and temperature with risk of fuel escaping into the coolant. As a consequence, fuel failures have to be detected quickly and failed fuel has to be transferred to the periphery of the reactor where lower sodium temperatures

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can be found, which minimize the reaction rates, or if necessary has to be discharged from the reactor, which impose a shutdown.

It is obvious that avoiding failure is a major challenge. In the long run, mixed carbide or nitride might be preferred to mixed oxide, for that reason.

2.7. *Subassembly behaviour and design*

The pin bundle concept using helical spacer wire has been widely adopted because of its simplicity and reliability. The basic design principles are as follows.

Differential deformations due to differential swelling should be limited between pin and spacer wire. If the spacer wire deformation is higher than the pin deformation, the sodium coolant flow can be reduced and consequently the fuel pins overheat slightly. On the other hand, when the pin deformation is higher than that of the spacer wire, there is a mechanical interaction and the fuel pin undergoes a helical deformation. The designer can avoid both of these extreme situations: the correct design is currently obtained by a proper choice of materials, mainly by using a low-swelling material.

Vibrations and mechanical interaction between the pin bundle and wrapper tube should be prevented by limiting subassembly clearance tolerances.

Wrapper deformation should be limited: subassembly handling procedures, notably for removal or relocation, should not involve excessive loads.

Irradiation-induced void swelling and creep can product different types of wrapper deformation during reactor operation (see figure 1). At the centre of the core, subassemblies are expected to remain perfectly straight with an elongation and an increase of distance across flats. But at the periphery, subassemblies may bow outwards due of the differential void swelling induced in the opposite faces of the wrappers as a consequence of the neutron flux gradient. The combination of void swelling and creep induced by the internal sodium pressure produces dilation and rounding of the wrappers faces.

2.8. *Status of our knowledge*

Today one can say that we control the swelling of the cladding but that we are not sure of being able to avoid the embrittlement of clad materials. Thus because it can induce clad failures, mechanical interaction must be avoided by careful control of the fuel smeared density in the pin. Cladding corrosion is of concern especially for high burn-up.

The proper choice of wrapper and cladding materials, taking into account their swelling rate and mechanical properties at operating temperatures and high irradiation doses, leads to a satisfactory in-reactor behaviour of the subassemblies in the existing test reactors.

3. Performance of fuel and specifications for a large fast reactor

3.1. *Specifications*

Twenty years of research (or more) have provided a wealth of knowledge on core and core behaviour, which has been briefly described above. This knowledge has been integrated in models, codes and design rules which are necessary to design properly the fuel for a given reactor.

A good design has to fulfil the following requirements:

(*a*) it must take in account the technological limits linked to problems described above;

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(*b*) it must meet the requirements of the designers.

The requirements of the designers are the results of several trends.

1. The request for a burn-up as high as possible, because the burn-up has a great impact on cycle cost: doubling the mean fuel discharge burn-up nearly halves the fuel cost.

2. The requests to fulfil the requirements of the utilities.

3. The operational request of the utilities are easy to understand but impose severe conditions on the designers and on the fuel: (i) grid load following and unscheduled cycling; (ii) handling requirements; (iii) long fuel-in-reactor time; (iv) no fuel failure or good failure detection systems.

4. The request of the safety authorities, in particular, (i) tests of criticality incidents; (ii) tests of one subassembly melting by lack of cooling.

Among the safety authorities' requirements one is of utmost importance. As some of the phenomena which lead to failure and which have been described above are of statistical nature (corrosion, swelling, damage), the safety authorities (and the utilities and designers) do not consider any result as demonstrated and guaranteed if this result has not been statistically obtained. In France, a statistically acceptable result must be based on around 10 subassemblies (or *ca.* 2000 pins).

All those considerations being taken into account, the requirements which have to be met for the next European Fast Reactor are the following: (i) burn-up, 150000 MW d t^{-1}; (ii) damage, 180 DPA NRT; (iii) lifetime, 1500 full power days; (iv) statistical basis, 10 fuel subassemblies?; (v) design basis accident, melting of one subassembly?; (vi) oxide smeared density, 85%.

3.2. *Results*

We in Europe have not yet demonstrated and reached all the objectives given by the utilities and design companies.

It should be stressed that one of the main difficulties is that there are no experimental reactors that reproduce exactly the conditions that prevail in a large reactor. For example, small test reactors (like DFR, Rapsodie, KFK) gave high burn-ups but low damage dose. Statistical demonstration requests bigger reactors like PFR or Phénix or Superphénix.

The results are the following.

(*a*) *Burn-up*. The European and world results show that high burn-ups are easy to achieve; as demonstrated in figure 7.

(*b*) *Damage*. (See figure 8.) However, high doses are more difficult to obtain as illustrated in table 1. On the whole, we have not yet reached the target. As a consequence several cladding materials have to be tested as part of the European programme.

(i) The British develop the high-nickel alloys (PE 16). They have reached 140000 MW d t^{-1} and 120 DPA.

(ii) The French and Germans develop the optimized 15–15 steels. They have reached 140000 MW d t^{-1} and 135 DPA.

(iii) The Belgians develop (as a back-up) the oxide-strengthened alloys. Good results have been reached on specimens and on few pins (87400 MWJ t^{-1} and 91 DPA NRT).

(*c*) *Fuel density and reactivity decrease during the life of a core*. High burn-ups are required for economical reasons. Supposing that adequate damage-resisting cladding is found, there is an easy way to reach high burn-ups without trouble, by decreasing the smeared density of fuel, to minimize swelling and fuel–cladding interaction.

FIGURE 7

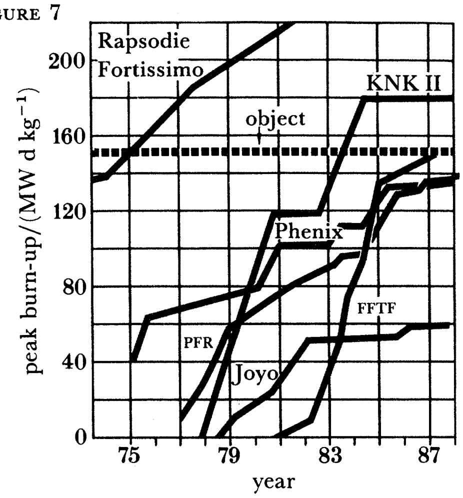

FIGURE 8

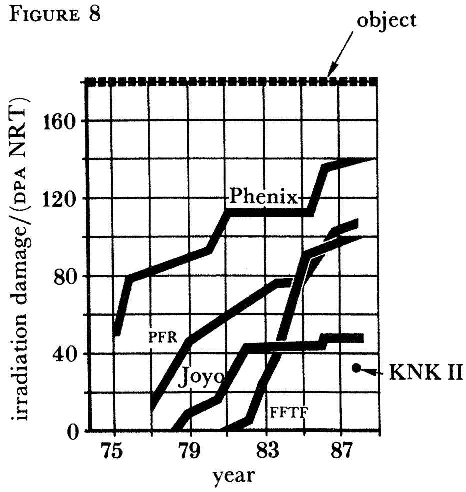

FIGURE 7. Fuel performances of some FBRS.
FIGURE 8. Irradiation damage on structural materials of some FBRS.

TABLE 1

	burn-up	dose/DPA	comments
Rapsodie	200000	100	small reactors dose too low
KNK II	180000	40	
PFR	140000	120	below the dose target
Phénix	140000	135	
FFTF	150000	100	dose too low target
EFR	150000	180	

But if the fuel density is too low, and if the burn-up is high, the reactor might have not enough reactivity to operate as requested.

Our best calculations lead to a required density of 85 %. This is a very high density for very high burn-ups, which is probably near a technological limit. We all have opportunities to check that question because PFR and FFTF operate with an 80 % smeared density fuel, when Phénix operates with an 88 % smeared density.

The longest lifetimes are Phénix (1050 days); PFR (600 days); FFTF (600 days); KNK II (400 days).

This is clearly not enough in view of the risks of corrosion by chemical attack and of radiation embrittlement, which could occur during the target lifetime of 1500 full power days.

(*d*) *Statistical basis*. Phénix, PFR and later SPX, gave the statistical basis for all the future fuels. This statistical basis is very important, taking into account that the guaranteed performance chosen by the designers are much lower than the maximum performances obtained in test reactors (on a few subassemblies).

As an example, the French have reached 140000 MW d t^{-1} in Phénix. But the safety authorities have given only very recently the authorization to operate Superphénix at 100000 MW d t^{-1}.

As a matter of fact the requirements for EFR implies that our best R&D results go as far as 200000 MW d t^{-1} and 220 DPA.

(*e*) *Operational results and safety*. A good basis for safety behaviour can be found in the Treat, Cabri, Mol and Scarabée tests. Those tests might need to be extended, taking into account the high performances requested for commercial breeders, especially high burn-ups, in more or less damaged claddings.

Conclusion

The European R&D community research for fuel development has reached a very high level of understanding of the basic phenomena involved in fuel behaviour as well as of the industrial requirement to be fulfilled by a well-designed fuel. (See table 2.)

Table 2. Target and present results of FBR fuel

	target	present results
experimental results	200000 MW d t^{-1}	140000 in Phénix and PFR
corresponding industrial limit	150000 MW d t^{-1}	—
experimental results	220 DPA	135 DPA in Phénix (stainless steel) 125 DPA in PFR (P. 16)
corresponding industrial limit	180 DPA	—
lifetime	1500 days	1050 days in Phénix 600 days in PFR
density	85%	88% in Phénix (too high?) 80% in PFR (too low?)
statistical base	requirement of 10 subassemblies or 2000 pins	more than needed for stainless steel coming in PFR for PE16
safety	criticality melting of subassembly	Cabri, Treat Scarabée, Mol 7C

The results are promising. Nevertheless, some difficult problems are not yet fully solved and need further work. The future of the breeder, through the lowering of the cycle cost, is linked to the success of the fuel research.

I am greatly indebted to Mr H. Mikailoff of the fuel division of the Commissariat à l'Énergie Atomique for his helpful discussion and comments.

Reference

Olander, D. R. 1976 Fundamental aspects of nuclear reactor fuel elements. Technical Information Center, Office of Public Affairs, Energy Research and Development Administration.

Discussion

A. Tuzov (*Institute of Physics and Power Engineering*, *Obninsk*, *U.S.S.R.*). What are the impacts of the target burn-ups of 20% heavy atoms for the EFR on the design of fuel assemblies, in particular, on the size of the fuel pin gas plenum and the method of spacing the pins (grid or wire wrap)?

R. Lallement. This is a challenging target given an overriding need to avoid clad failures. Many aspects of the design are not yet resolved as the testing programme has not reached the stage at which all the essential information can be provided to the designers. A 100% gas release from the fuel will be assumed in sizing the plenum. The choice between grid or wire wrap will not be made until statistical data has been built up on behaviour under normal conditions including cycling and shutdown and in-fault conditions.

K. Q. Bagley (*UKAEA*, *Risley*, *U.K.*). Experience suggests that the behaviour of oxide fuel is relatively benign. Swelling and restructuring do not stress the clad severely under steady-state conditions because volume changes are taken up by irradiation creep of clad or fuel, although rapid changes in reactor power can lead to high stresses. The most damaging feature of oxide fuel relates to the internal corrosion capability. French irradiation experiments have indicated that internal corrosion is significantly reduced in the axially heterogeneous pin design. Other performance benefits, such as reduced dose at target burn-up, also appear to derive from the axially heterogeneous pin design. Would Mr Lallement comment on the implications of these factors on permissible burn-up targets?

R. Lallement. Clad corrosion cannot be avoided but efforts must be made to minimize it. Some experimental work has indeed indicated that it is reduced in the axially heterogeneous core. Fission products are trapped in regions away from the clad interface at the core centre.

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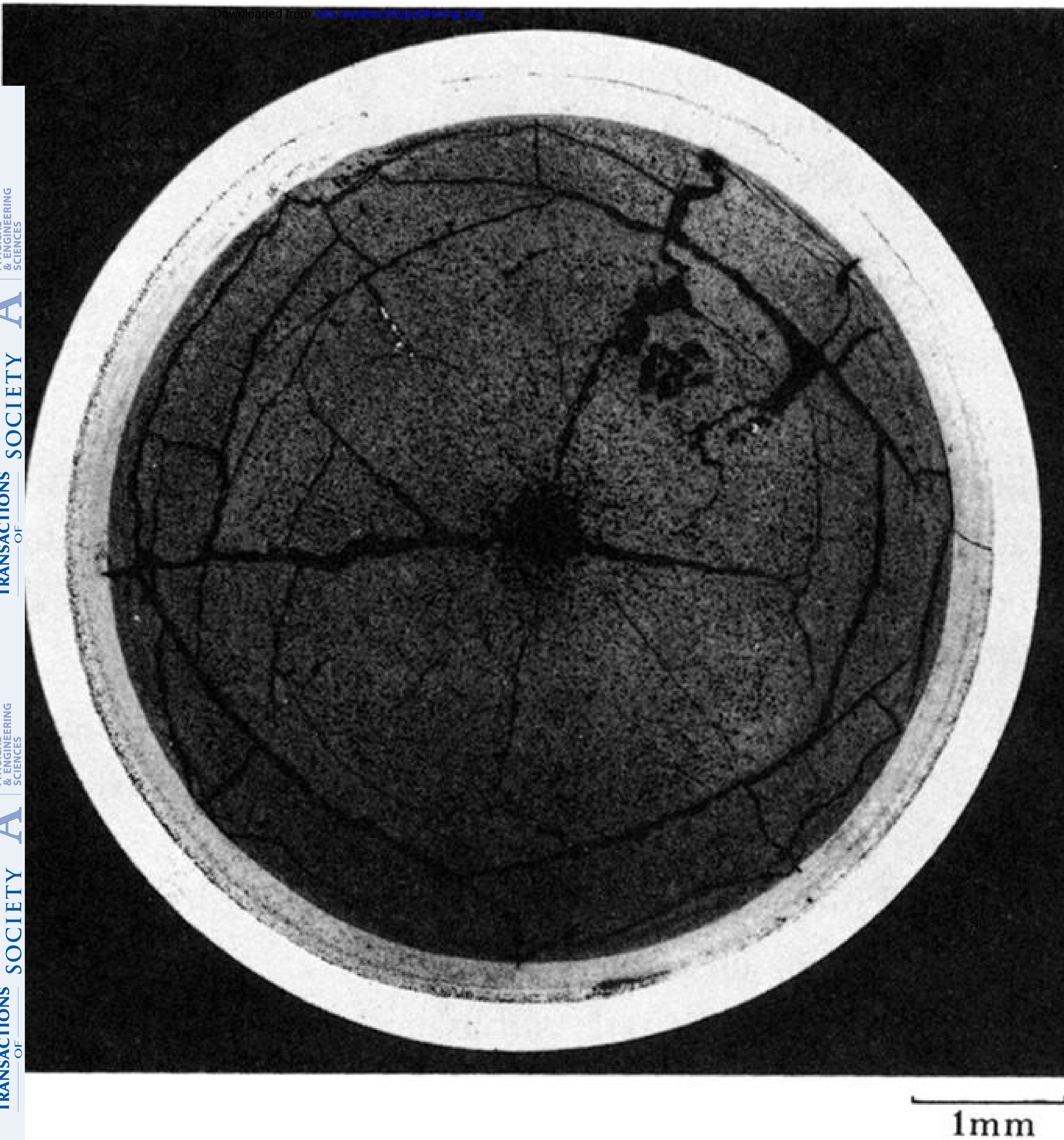

FIGURE 6. Example of large localized corrosion at high burn-up.