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Journal of Nuclear Materials 274 (1999) 153–159

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Toward very high burnups, a strategy for plutonium utilization in pressurized water reactors

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Abstract

The aim of using plutonium more efficiently in pressurized water reactors has led to objectives of high and very high burnups. The reasons are not only economic, but also related to the optimization of the utilization of fissile material and to increased proliferation resistance. Here are presented the reflections that contributed to the definition of a R&D programme conducted by the Nuclear Reactors Division (DRN) of the Commissariat à l'Énergie Atomique (CEA) in this domain in the nineties.

1. Introduction

The very particular context of nuclear industry in France, which committed itself very early to reprocessing activities, has given rise to the fact that, at present, a very great quantity of plutonium is available. The implementation of fast breeder reactors intended to use plutonium is delayed. Moreover, the policy of the French utility, Electricité de France (EdF), consists in requiring the fuel cycle operator, Compagnie Générale des Matières (COGEMA), to only reprocess the amount of irradiated fuel needed for the fabrication of reloads for the 20 (soon 28) pressurized water reactors (PWRs) authorized to recycle Pu in 30% plutonium–uranium mixed oxide (MOX) core loading. This has led on the one hand to accumulate stocks of already reprocessed Pu, and, on the other hand, to encumber the storage sites of nuclear power plants as well as those of COGEMA.

Moreover, [1], keeping in mind that both fissile and fossil resources are far from being inexhaustible, saving our natural resources should be a growing concern. Current trends, related to public acceptance, are becoming stronger and this means considering the need to minimize the volume, quantity, and radiotoxicity of nuclear wastes. Finally, the instability of certain regions of the world is a further reason to reinforce our desire to increase resistance to proliferation. It has become obvious that the current situation of French nuclear industry does not exactly meet these criteria.

The conditions as described above argue in favour of a substantial extension of the cycle and therefore of high burnups with the very long 'once through' version, and direct permanent storage in geological formations using a 'rock-like' fuel concept (or a cermet fuel), which represents the final version of these concepts. Leaving the fuel longer in the core permits extracting more energy from the fuel, which favours the economic aspect, especially if the fissile material does not require ^{235}U enrichment, but uses Pu. This also allows deferring storage, which should not be considered as procrastination, but as providing more time to refine the studies required to treat the problem as a whole, and possibly constructing additional storage sites. The reduction of the amount of actinides could also represent an interesting point, but without prior definition of the type of fuel and the spectrum of the reactor, this cannot be considered as established; on the other hand, resistance to proliferation is assured, since in-core storage may be seen as the place where fissile material is best protected from external aggression.

This study presents a general review of possible actions with a view to taking this problem into account in the medium term and also propose a certain number of

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0022-3115/99/$ – see front matter
PII: S0022-3115(99)00084-7

research orientations that will better define the shape of extensive R&D programmes to come for the years 2000–2005.

2. The materials

The first field of possible action could be devoted to the intrinsic properties of the material used to manufacture the fuel. Although the current reference is the UO_2 ceramic, a new fuel should at least meet the specifications of today's fuels, and, if possible, go a step further with respect to safety and economics.

Present material limitations concern the release of fission gases. This phenomenon is in fact driven by two highly dependent factors: the first is linked to the nature of the ceramic, which fractures under irradiation and, as it evolves, has a tendency to offer increasingly numerous routes for gas migration. The second factor is connected to the operating temperature of the fuel, and particularly to the considerable temperature gradient inside the rod,

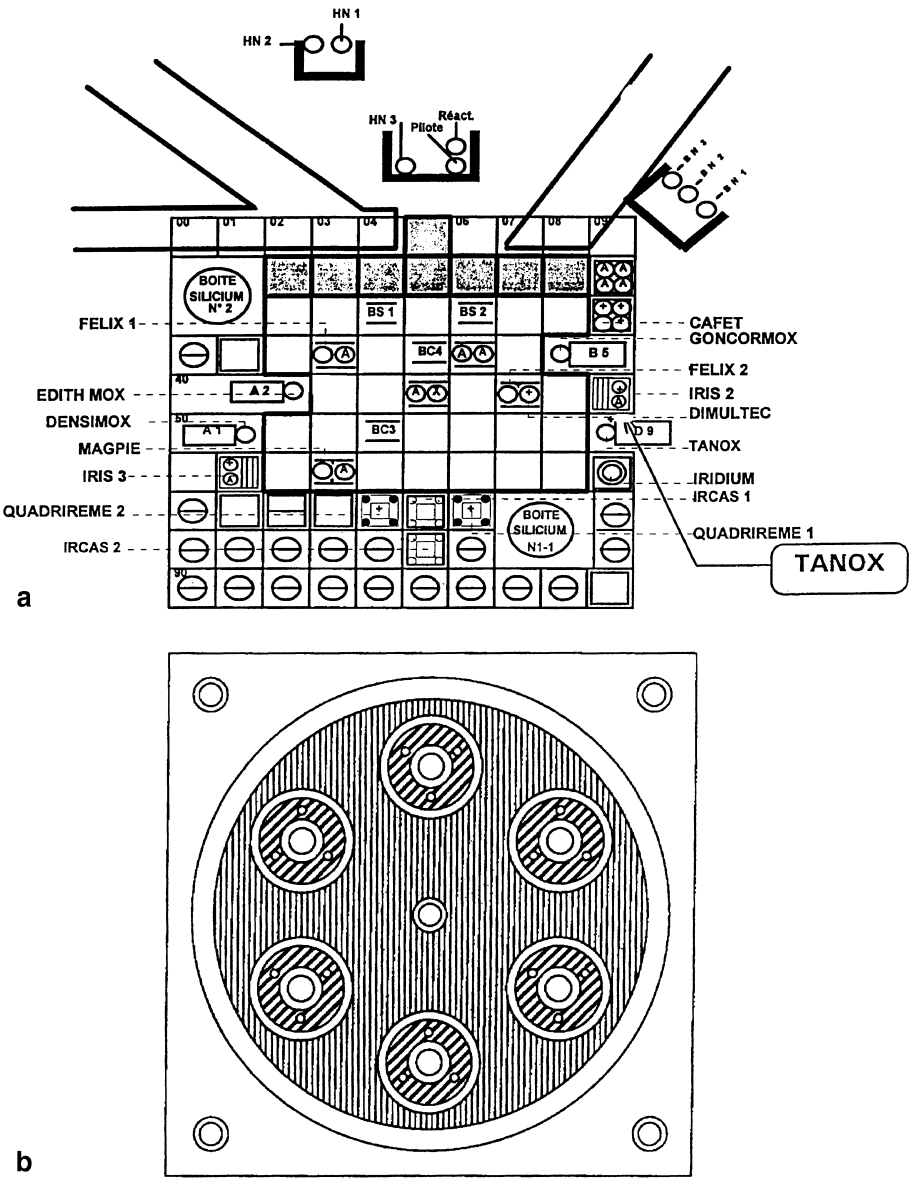


Fig. 1. SILOE core and TANOX location (a) SILOE. (b) Fuel per location.

which increases gas release. The combination of these two phenomena defines the conditions of pellet cladding interaction (PCI), which is a factor that limits not only operating conditions, but also the achievement of high burnups.

It was with the aim of providing solutions to this type of problem that research on advanced microstructures was initiated. This research, performed by the CEA Grenoble, has contributed to the determination of microstructures that improve significantly the gas retention by the ceramic. Overall experimental requirements in this field, linked particularly to the achievement of high burnups, resulted in the definition and the installation of the TANOX experimental device [2] (Fig. 1) in the SILOE research reactor at Grenoble. At the same time, work on composite fuels was oriented towards cercers, heterogeneous mixtures of two types of ceramic materials, one containing the fuel and the other being a neutronically inert matrix [3–6]. They feature a pre-microcracked ceramic fuel (see Fig. 2) to permit better resistance under irradiation, while a gap is arranged between the ceramic matrix and the spheres so that the gases are able to remain in the gap area.

The second type of material to be studied were metals, which have three essential advantages for the achievement of high burnups. Metals are potential barriers against gas diffusion and therefore ensure better fission gas retention. This also relieves mechanical stress on the cladding and thus the matrix becomes the first safety-related barrier. Better thermal conductivity means a lower operating temperature for the fuel and less thermal fatigue for the materials, and becomes a safety factor insofar as the potential energy stored in the fuel is decreased, which is a favourable factor in the event of a severe accident.

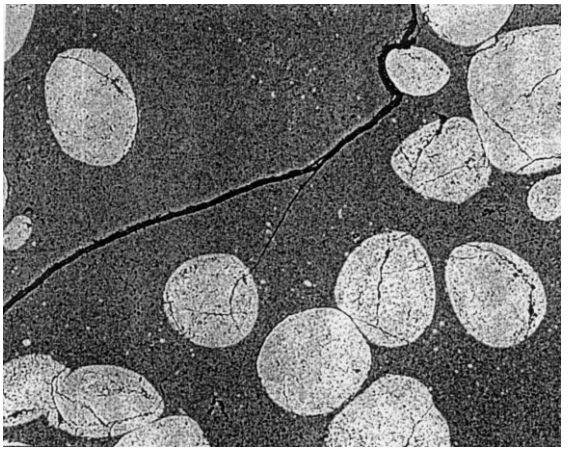


Fig. 2. Pre-cracked UO_2 grain in cercer composite. Clear gray: UO_2, dark gray: spinel, black: crack.

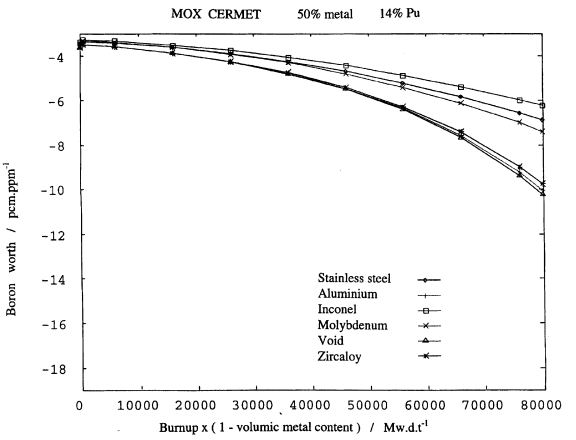


Fig. 3. Differential soluble boron worth for a MOX cermet.

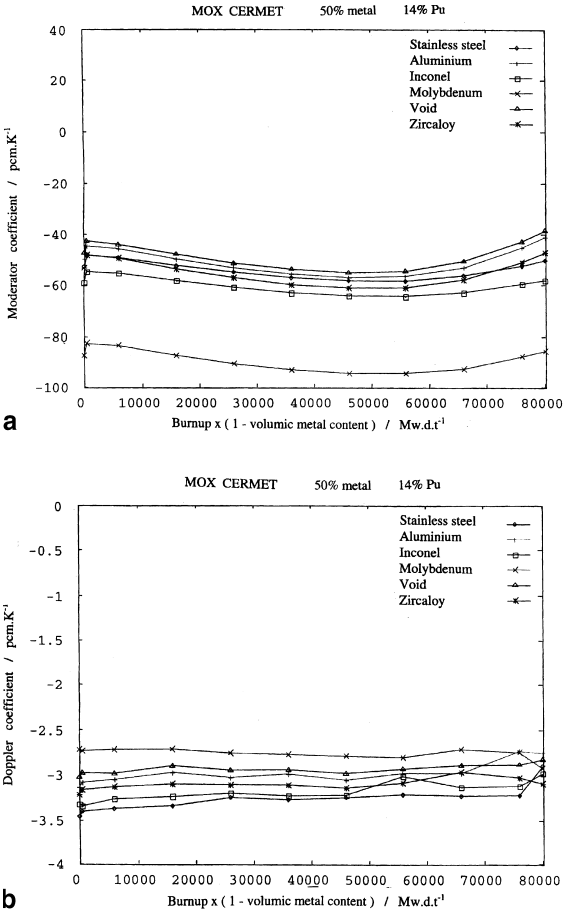


Fig. 4. Moderator and Doppler coefficients for a MOX cermet, geometric parameters. (a) Moderator coefficient. (b) Doppler coefficient.

Another solution using materials for the 'rock-like' concept based on zirconia or yttrium aluminate is being studied and followed up, but will not be addressed here.

3. The fuel rods

The TANOX device, installed in the SILOE reactor, and the TANOXOS installed in the OSIRIS reactor, Saclay, have proved to be determining tools for the rapid accumulation of high burnups (0.5 GW d t^{-1} per full power day, i.e. ten times more than in a PWR), which is an essential factor insofar as the achievement of a burnup of 100 GW d t^{-1} represents ten years in-core residence, a condition that can only be applied to final test specimens and not to those whose optimization is currently being sought.

The first series of experiments resulted in the comprehension of certain mechanisms linked to the transport and release of fission gases and selective irradiation tests led to the selection of advanced microstructures for the fabrication of fuel rods now under irradiation in PWRs.

The following series of tests, TANOX 2, was devoted to the study of composite fuels for high burnup objectives. The following determining information was drawn from these series of tests:

- A disappointing behaviour of the spinel under irradiation
- A cermet (64 vol% Mo, 36 vol% UO_2) core temperature of about 793 K for a linear power density of 350 W cm^{-1}, which reduces to 693 K under PWR conditions
- Very limited release for the cermet (comparable to UO_2 at 16 GW d t^{-1}).

Cermet therefore appears to be a very serious candidate for high burnups, insofar as it minimizes several factors usually limiting burnup

- It increases thermal conductivity, which results in a decrease in the fuel temperature and hence decreases gas release and thermal-mechanical stresses
- It increases safety since the matrix becomes the first safety barrier and, moreover, the fuel decreases stored potential energy
- It limits, or even precludes, the occurrence of PCI as a result of the metal-to-metal contact of the cladding and the matrix.

4. The assemblies

If one considers, albeit a little arbitrarily, that high burnups begin at 100 GW d t^{-1}, representing some ten years in the reactor, experimental data has to be obtained in this domains. This is why the objective of the second series of tests in SILOE, TANOX CCE (erbium composite fuel) was to reach a minimum of 100 GW d t^{-1}. As the implemented fuel has to simulate plutonium, 2 wt% Er_2O_3 was introduced into the fuel ceramic (80% Mo–20% UO_2, 40% ^{235}U enriched UO_2), since Pu fuel in an inert matrix would require the addition of a poison to ensure the reactivity control [7–10].

This fuel was manufactured and loaded into the TANOX device for the first test series in the SILOE reactor in 1997 with the aim of performing an irradiation test of a technological nature. In addition, following the EROÏNE experiments in the EOLE research reactor (CEA Cadarache) in 1996, which aimed to measure erbium negative reactivity values at time 0, since the achievement of high burnup favours Er consumption, a series of dose measurements was planned in order to obtain a ^{166}Er to ^{167}Er ratio for the validation of erbium combustion kinetics.

At the same time, neutron studies [11,12] were performed in order to determine the conditions in which assemblies using these fuels in an inert matrix could

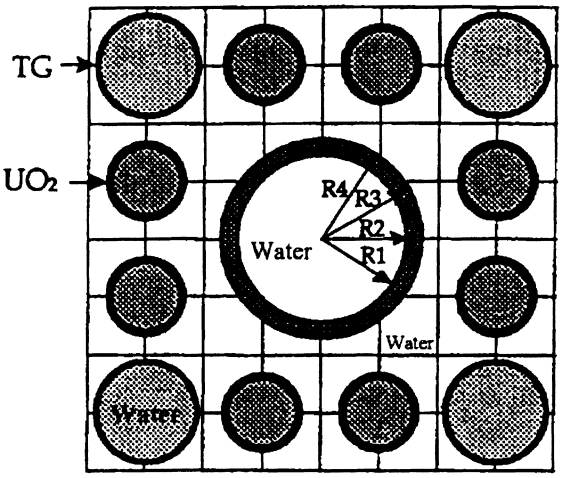
Table 1
Moderator and Doppler coefficients for a Pu–Zr cermet

	Tf: 550 ⇒ 300		Tf: 300 ⇒ 280		Tf: 280 ⇒ 120	
		Tm: 300 ⇒ 280		Tm: 280 ⇒ 120		Tm: 120 ⇒ 20
Step 0	−1.07		−1.07		−1.11	
		−35.48		−19.90		−8.49
Step 1	−1.09		−1.07		−1.11	
		−31.48		−15.49		−5.44
Step 2	−1.17		−1.15		−1.16	
		−31.90		−13.59		−3.00
Step 3	−1.30		−1.24		−1.26	
		−32.46		−10.50		+1.05
Step 4	−1.52		−1.49		−1.39	
		−29.08		−2.06		+10.16

Note: T – temperature (°C); index f – fuel; index m – moderator; 1 step = 2 yr fabrication + 5 yr irradiation + 5 yr cooling = 12 yr.

form part of a PWR core load. It was demonstrated that no particular problems were posed for uranium dioxide and uranium-plutonium mixed oxides cermets, that is to say that the values of the kinetic coefficients were acceptable for cycles of 12, or even 18 months (see Figs. 3 and 4 and Table 1).

On the other hand, without aiming for high burnups, changes in standard assemblies have been sought in



R4 (mm)	=	11.5	
R3 (mm)	=	11.0	} 1.261 mm
R2 (mm)	=	9.739	
R1 (mm)	=	9.239	
$VPuO_2/Vcomb$	=	0.23	

Local moderation ratio = 5.96 (annular pin)
Global moderation ratio = 3.47

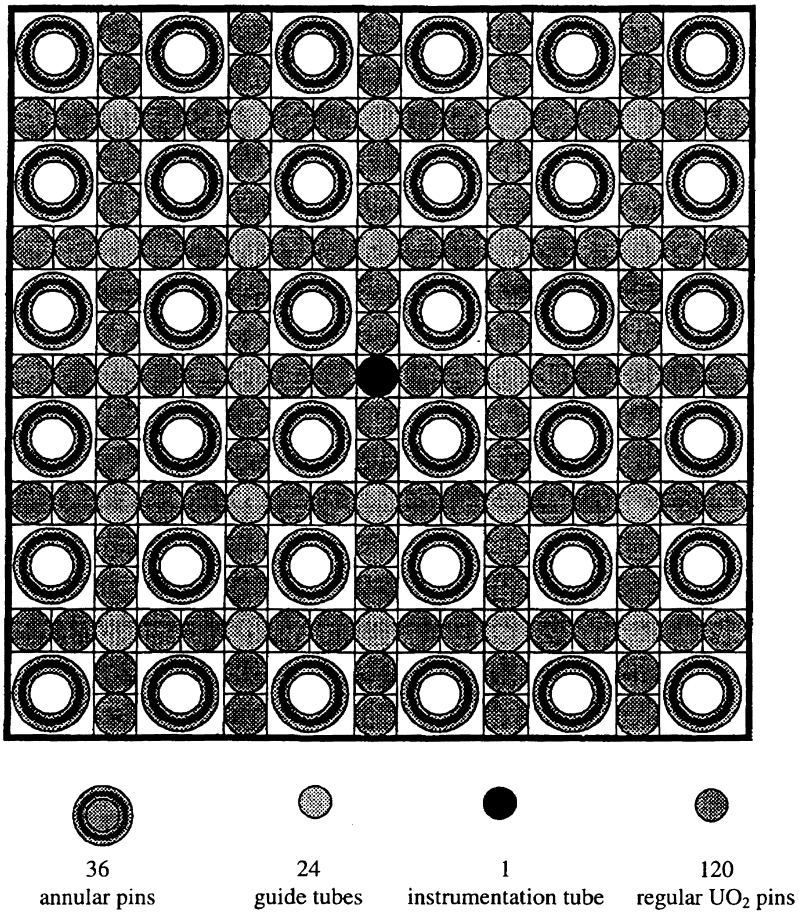


Fig. 5. Advanced plutonium assembly (APA).

Table 2
APA assembly global balance for Pu and minor actinides (kg yr^{-1})

Step	1	2	3	4	5	6	7
Fissile Pu content at loading (%)	63.7	49.4	45.8	44.2	43.3	42.8	42.4
^{235}U enrichment in 120 fuel rods (%)	0.53	2.59	3.17	3.43	3.57	3.66	3.71
Unloaded Plutonium (tons)	6.774	8.041	8.415	8.605	8.709	8.772	8.813
Fissile Pu content at unloading (%)	29.6	24.9	22.5	21.0	20.2	19.6	19.4
Pu consumption (%)	59	51	49	48	47	47	46
33 PWR – U +12 PWR – APA							
Pu	−300	900	1330	1520	1630	1690	1730
^{237}Np	570	615	627	632	635	637	638
Am	1396	1558	1604	1626	1639	1646	1653
Cm	393	487	535	562	578	588	594
Pu + MA	2359	3620	4096	4340	4482	4561	4615

Pu	^{237}Np	Am	Cm	Pu+MA
45 PWR – U once through				
12 813	741	758	108	14 420

Note: Fuel balance in 12 APWR – APA (5 yr cooling); 1 step = 2 yr fabrication + 5 yr irradiation + 5 yr cooling = 12 yr; Global balance (remaining Pu and minor actinides/kg yr^{-1}); 65 GW – 437 TWh.

order to obtain both better control and better kinetic coefficients for fuels using large proportions of plutonium.

The APA as presented in Fig. 5 with the data from Table 2 [10,13–15], is a typical model of this research. It should be observed that the preliminary version presented here uses a $(Pu,Ce)O_2$ fuel material, but that the nature of the fuel and the geometry of the assembly can be modified for a specific optimization related to a given fuel and strategy.

5. Conclusion

High burnups are not an end in themselves, but a means of progressing towards a better utilization of resources and a reduction of the plutonium inventory in France. At the present stage of our reflection and studies, it seems that, although all the problems related to the fundamental aspects described here – materials, rods, assemblies – are far from being solved, the resources of physics and technology can provide solutions. The weak link in this chain at present is the cladding, its corrosion, and the chemistry of the primary system. The solution to these problems lies no doubt in changing and also, no doubt, in challenging, a certain conservatism, which will require more active innovative research.

Acknowledgements

Thanks are due to Dr Stefano Baldi for his contribution in this study.

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