PLUTONIUM RECYCLING IN FRENCH POWER PLANTS: MOX FUEL IRRADIATION EXPERIENCE AND BEHAVIOUR

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Abstract

Plutonium recycling in PWR's started in France in 1987 with the first reload containing MOX fuel in the Saint-Laurent B1 reactor. By the end of 1994, more than 400 MOX fuel assemblies had been delivered by Fragema to 7 different EDF 900 MW power plants. As the number of PWR units recycling Plutonium is increasing, MOX fuel is required to assume the same operational flexibility as UO2 fuel. So, MOX fuel must follow UO2 fuel developments (1/4 core fuel management, load follow) in terms of discharge burnup and maneuverability. The acquisition of new irradiation data of MOX fuel representative of the current product and operation mode allows to optimize the fuel design leading to improved performance. The development programme set up by French partners CEA, EDF and FRAMATOME, analyzes MOX fuel behaviour through: analytical experiments in experimental reactors conducted under normal and transient operation and, surveillance programmes consisting of the irradiation in commercial reactors and the subsequent examination of MOX fuel assemblies and fuel rods. The paper describes our ongoing programme and discusses the main results obtained so far with a particular focus on the surveillance programme that has been set up on the first MOX reload. Several characterized MOX fuel rods have been withdrawn after each of the three irradiation cycles and have been examined in hot cells. Those data are related to a burnup range up to about 43 Gwd/tM. Power ramp tests performed with fuel segments of those rods showed that MOX fuel behaves particularly well from the pellet-cladding interaction standpoint. Recent data related to high burnup fuel rods (4 irradiation cycles, 52 GWd/tM) and to MOX fuel irradiated under load follow conditions are also presented. These examinations show that from both the waterside corrosion and rod dimension aspects, the MOX assemblies behaved similarly to UO2 fuel. However, the rod puncture data indicate a somewhat higher fractional gas release than UO2 rods: this behaviour is first and mainly explained by the linear heat rates of the MOX which are higher than those of UO2 rods at the same burnups, and to a lesser extent by the more heterogeneous microstructure of the MOX fuel. Fuel restructuring during normal or transient conditions as well as the behaviour of fission products have been carefully examined through micrographs and electron probe micro-analysis providing useful information for modelling purposes.

INTRODUCTION

In the early eighties, it was decided in France to implement a closed nuclear fuel cycle policy by recycling the valuable materials contained in the spent fuel. Such a policy was also adopted by other European countries and by Japan.

By the end of 1994, 460 MOX fuel assemblies had been supplied by Fragema to 7 French 900 MWe PWR's and abroad. Table 1 shows the deliveries and the irradiation experience in the French EDF

reactors. 196 assemblies have already been normally discharged after three cycles, with average burnups of 36 to 38.5 Gwd/tM. Four more have completed successfully a fourth cycle up to 44 Gwd/tM. Two of the French reactors loaded with MOX have been operated under load follow conditions.

Reliability of MOX fuel assemblies has proved to be at least as good as for UO2 fuel ($1.5\ 10^{-5}$ leaking rod, per cycle) since only one MOX assembly has shown a leak, which was slight enough to allow reloading according to EDF criteria [1].

Upgrading of MOX product performance is going ahead according to a progressive and pragmatic approach involving the experience feedback and the joint R & D programme of Framatome, EDF and CEA in order to confirm the good behaviour of the current fuel products and to support the licensing of extended burnup and maneuverability.

FUEL DESIGN AND FABRICATION

Currently, the recycle rate is 30 % MOX assemblies per reload and the type of management used is the OUT-IN-IN third-core strategy (UO2 core management mode at the time of plutonium recycle licensing) on an annual cycle basis. However, new MOX assemblies are not loaded immediately near the baffle in order to minimize their contribution to the vessel fluence.

A typical mixed core reload comprises 16 MOX assemblies and 36 UO2 assemblies. The reload assemblies are all of the current 17 x 17 AFA design with a UO2 fuel enrichment of 3.25 % (equilibrium enrichment of third-core UO2 management). The UO2 matrix for the MOX fuel is made of depleted uranium. The average plutonium concentration of the MOX assemblies is adjusted from the isotopic compositions of the plutonium and of the uranium matrix to obtain cycle lengths equivalent to those obtained with an all-uranium core also reloaded on a three-batch basis. The total plutonium average contents for each MOX production run lies within a 4.5 % - 5.3 % range. This content range is due to the wide isotopic diversity of the plutonium. In addition, the same reload may contain MOX from different production runs [2,3].

The neutronic design of the MOX assembly has to be optimized in order to reduce the power peaking at the UO2 and MOX interface and to obtain the lowest possible shape factor. That is achieved by zoning the assembly using three different plutonium contents in a concentric distribution.

Two processes are currently used for the MOX fuel fabrication:

- the MIMAS (Micronized Masterblend) process adopted by the Belgonucléaire/Dessel plant (35 t/year capacity), consists in the micronizing of UO2 and PuO2 powders to form a master blend with a plutonium content in the range of 20-30 %; this primary blend is then mechanically mixed with a free-flowing UO2 powder (ex. AUC or ex. ADU) to obtain the specified plutonium content

- the COCA (Cobroyage Cadarache) process implemented in the CFCa/Cadarache plant (15 t/year capacity), consists in a single step (ball-milling) mixing of the UO2 (ex. ADU) and PuO2 powders.

  The new MELOX plant (Marcoule) will use the MIMAS process with a UO2 powder obtained via the ADU route. The production is scheduled to start this year, to reach a capacity of 100 tons per year in 1997, with a further increase by the end of the century.

R & D PROGRAMME

It is known that not only the presence of plutonium, but also the fuel materials and the manufacture process used could have an impact on the mixed oxide fuel behaviour compared to the standard Fragema UO2 fuel.

The design codes and models take into account the MOX specific characteristics, nevertheless progressive adjustments are made, based on the development programme set up by the French partners CEA, EDF and FRAMATOME.

It consists of :

(1) analytical experiments in experimental reactors conducted under normal and transient operation in order to assess the kinetics of fission gas release, the fuel temperature evolution, the in-pile densification, the behaviour of pre-irradiated fuel rods under power ramp test in order to determine the failure limit due to PCI, the mechanical behaviour of MOX pellets and the release of fission products from a leaking rod.

(2) global experiments consisting of the irradiation and the examination of MOX fuel assemblies in commercial reactors.

This programme is overviewed hereafter, presenting the main results obtained so far, with a particular focus on the surveillance programmes.

Up to now, most of the data are obtained through the examination of the 3 cycle rods of the first reload in the St Laurent B1 reactor : several characterized MOX fuel rods have been withdrawn after each of the three irradiation cycles and sixteen have been examined in hot cells. These data are related to a burnup range up to about 43 GWd/tM and to the three different plutonium contents of the current MOX assembly.

The examinations of three-cycle fuel rods irradiated under load follow conditions during the last cycle in the St Laurent B2 reactor are almost completed.

On-site measurements have been performed on MOX fuel rods irradiated four cycle in the Gravelines 4 reactor up to a rod burnup of 52 GWd/tM and hot cells examinations are being carried out.

This programme involves also examinations of different fuel types : one, for instance, aimed at providing experience feed-back on the behaviour of MOX pellets fabricated according to the process (MIMAS) implemented in the Melox plant, with a matrix using UO2 powder obtained via the ADU route.

ANALYTICAL EXPERIMENTS

- Physical properties : out-of-pile measurements of lattice parameters, heat capacity and thermal diffusivity as a function of Pu content, Pu homogeneity, stoichiometry of MOX representative of the current PWR fuel fabrication (a significant amount of data on the high Pu content FBR fuel still exists) have been recently carried out in the CEA laboratories of Cadarache. The study of the lattice parameters of the fuel after homogenization confirmed, for example, that the stoichiometric mixed oxide follows the solid solution law (Figure 1). Also, the thermal conductivity of the low Pu content PWR fuel doesn't show significant deviation compared to UO2 fuel (Figure 2).

- Oxide densification : the DENSIMOX experiment was designed to determine the in-pile densification kinetics of MOX fuel from various manufacturing routes in relation with out-of-pile sintering tests. Pellet stack length was monitored through accurate neutron radiographies up to maximum densification. The results concerning two MOX fuels (ADU and AUC powder) are now acquired [4].

- Fuel centerline temperature and fission gas release kinetics : the GRIMOX instrumented in-pile experiment was designed to monitor fuel centerline temperature and fission gas release kinetics during irradiation. The irradiation device also contained UO2 pellets allowing "in-situ"

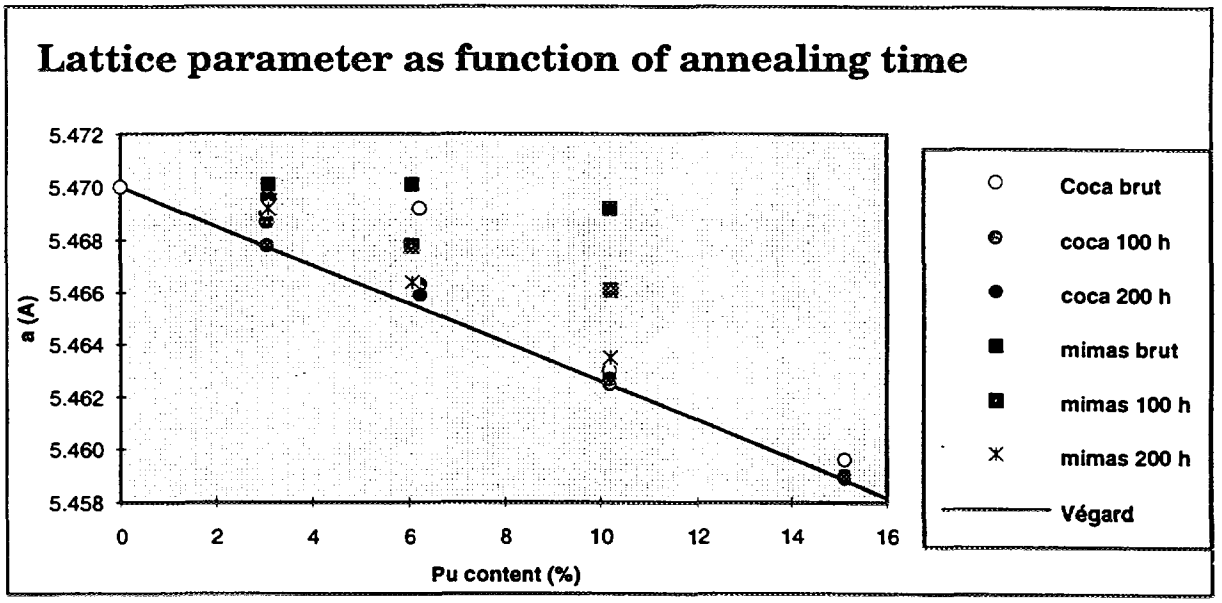


FIG. 1. Lattice parameter as function of annealing time

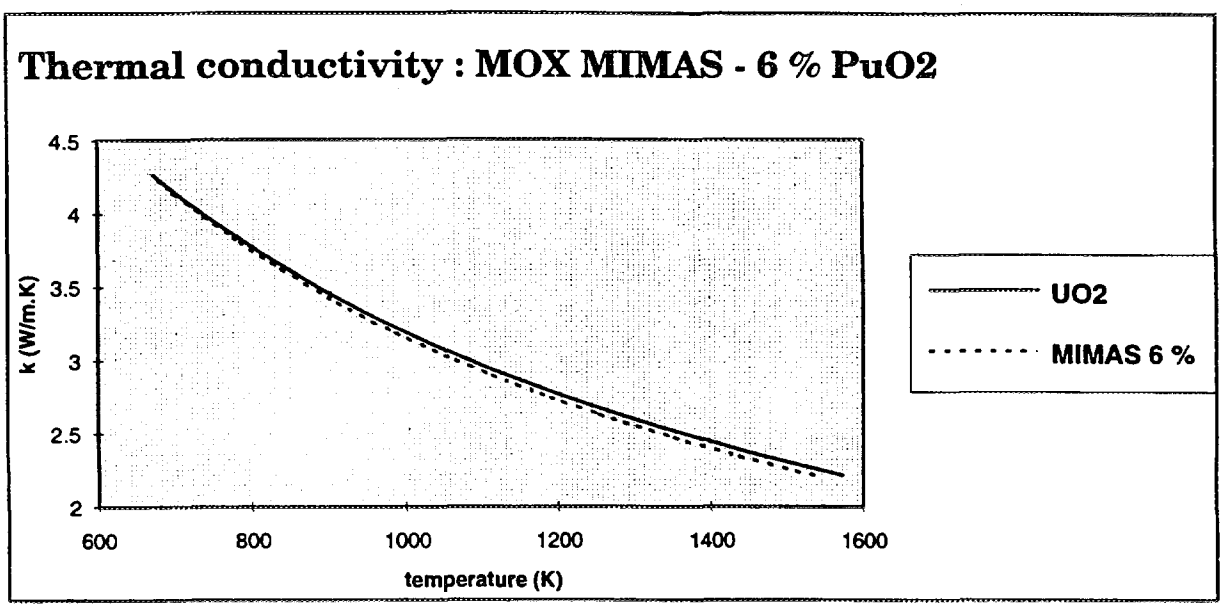


FIG. 2. Thermal conductivity: MOX MIMAS - 6% PuO_2

comparison of changes in parameters. The experiment completed six irradiation cycles in the SILOE experimental reactor (~ 5000 MWd/tM) monitoring moderate as well as high linear heat rates. The results showed that, in the investigated linear heat rate range the MOX temperature was slightly higher than that of the UO2 fuel (Figure 3).

- Pellet-cladding interaction : power ramp tests have been run on 2 and 3 cycle refabricated fuel rods irradiated in the SLB1 reactor. Linear power levels of 480 W/cm have been reached without failure, proving conclusively that MOX fuel behaves particularly well from the pellet-cladding interaction, and therefore from the power plant maneuverability standpoint. Such a favorable

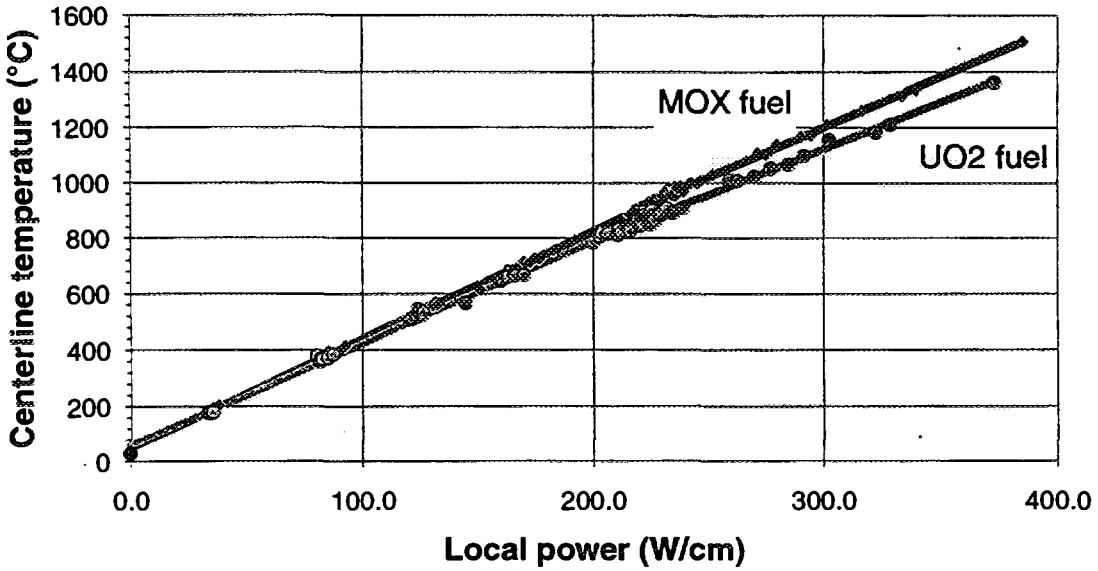


FIG. 3. GRIMOX O2 : centerline temperature versus local power

behaviour is attributed to the higher MOX pellet creep with respect to the UO2 pellet during power transients. This behaviour is in accordance with other published data. Nevertheless, new analytical experiments are being carried out in order to get a better evaluation of this benefit : first, out-of-pile creep tests and also, continuous in-pile measurements of the deformations of MOX and UO2 pellets (of the same geometry) using the DECOR experimental device [5].

- Fission products release : the EDITHMOX experiment consisted of the irradiation of a leaking fresh MOX fuel rod in an experimental loop. The measured gaseous and airborne fission products release rates were comparable with the findings of the uranium oxide experiments. Metallographic examinations showed an appreciable evolution of microstructure with gas precipitation bubbles due to fuel oxidation under water vapor conditions [6].

THE SURVEILLANCE PROGRAMMES

The overall dimensions and the cladding characteristics of the examined fuel rods were similar to those of the standard AFA UO_2 17x17 fuel rod. However, so far as higher fission gas release was expected in design calculations, the helium fill pressure has been lowered for the MOX fuel rods. The total plutonium content of the pellets ranged from 2.88 % (low content) to 5.57 % (high content).

Two characterized MOX fuel assemblies were loaded in 1987 in the 900 MW St Laurent B1 reactor and irradiated during three cycles. A total of sixteen fuel rods have been extracted after one, two or three irradiation cycles for hot-cell examination. Moreover, poolside examinations were also performed in order to verify the geometric behaviour of the assembly components.

The highest burnup rod reached 43 GWd/tM at a nearly constant linear power of about 220 W/cm.

The cladding of the examined fuel rods showed a similar dimensional and corrosion behaviour compared to those of UO_2 rods [7]. Pellet-clad interaction was observed at the end of the second irradiation cycle and a maximum ridge height of 20 μm was measured at the end of three cycles. After three cycles the maximum corrosion layer of 43 μm lies among the low values of the standard UO_2 data

range. Nevertheless, higher fuel densification than standard FRAGEMA UO_2 fuel was observed, in agreement with the expected behaviour of the ex-AUC pellets. Fuel swelling, however follows the same trend as standard UO_2.

The rod puncture data shown in Figure 4 indicates a somewhat higher fractional gas release after two irradiation cycles (about 0.7 %) than UO_2 rods irradiated in similar conditions (about 0.2 %). After three cycles, the fractional release ranges from 1 to 7 %.

This behaviour mainly results from the quite high power levels of some MOX rods (ranging from about 150 W/cm to 220 W/cm) during the last irradiation cycle, as can be seen on Figure 5.

The data obtained from the St Laurent B2 (SLB2) surveillance programme are coherent with the SLB1 results and don't show an effect of the load follow operation mode during the third irradiation cycle.

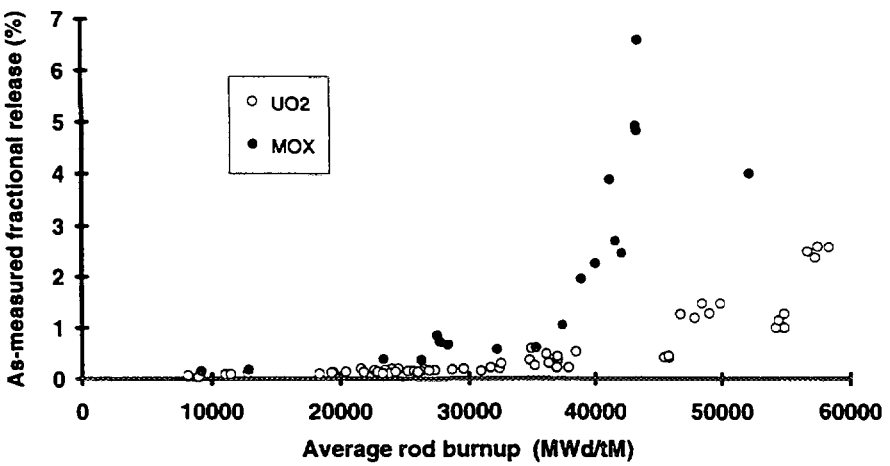


FIG. 4. Rod puncture data

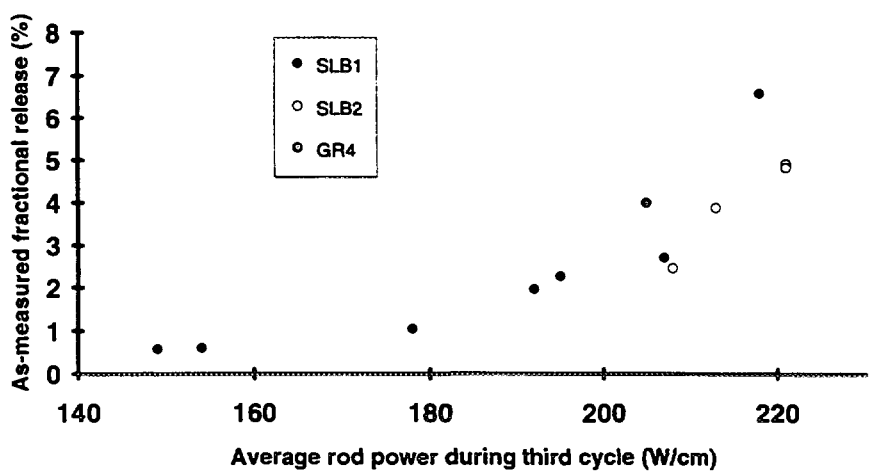


FIG. 5. MOX fractional gas release as function of linear heat rate during the third irradiation cycle

Only a part of this higher fractional release is attributed to the specific MOX fuel properties :

- somewhat higher fuel temperature during irradiation (a little bit lower thermal conductivity than UO_2 fuel and a different radial power profile),
- somewhat higher initial open porosity compared to the standard Fragema UO_2 fuel,
- plutonium agglomerates (up to 30 % Pu rich zones) resulting from the MIMAS fabrication process in which the local burnup is very high (more than 100 GWd/tM) producing high local fission gas concentration. Some fission gas atoms can be dissolved in the UO2 matrix and, to a lesser extent, migrate to the rod void volume through cracks or porosities.

Fuel microstructure as well as the behaviour of fission products were carefully examined through micrographs and electron probe micro-analysis (EPMA) providing useful information for modelling purposes [8].

The typical microstructure of the 3 cycle MOX fuel is shown in Figure 6.

The specific features relevant only to the MOX fuel behaviour and to fission gas release are described hereafter.

At the pellet edge, the Pu agglomerates are well characterized by a dense pore population resulting from fission gas generation and bubble coalescence. Fine metallic precipitates are also observable.

Towards the pellet center, the fine porosities coalesce to yield larger pores in the plutonium rich zones ; the surrounding UO_2 matrix preserving its as-fabricated microstructure.

At the pellet center, pore coalescence in the Pu agglomerates leads to the formation of big cavities surrounded by large metallic precipitates (4-5 μm). Grain boundary porosity is also observed in the UO_2 matrix of the fuel which yielded significant fractional gas release.

It is evident that this fuel restructuring is correlated to the burnup and the linear power the fuel experienced, nevertheless it is difficult to find a correlation with the measured fractional release of the fuel rods. In this respect, EPMA provided interesting information on the fission gas release mechanism in MOX fuel by means of the analysis of 1, 2 and 3 cycle fuel irradiated at various power levels.

In the low temperature regions, high xenon concentration peaks are measured in, or in the close vicinity of the plutonium rich zones, whereas in the high temperature zones or in the high burnup fuel, xenon is no longer detected in the Pu agglomerates and exhibits a quite flat distribution in the surrounding UO_2 matrix. The fission gases which are present in the UQ matrix due to the migration from the Pu agglomerates, recoil or in-situ formation follow the expected mechanism of the thermally-activated fission gas release. This is illustrated in Figure 7 which compares the diametral profile of Xenon concentration in 1, 2 and 3 cycle fuel.

In one-cycle rod, xenon is mainly localized in the plutonium agglomerates, very little is found in the UO_2 matrix. With increasing burnup, the amount of xenon in the matrix increases significantly and for the high rated three-cycle fuel, release of xenon at pellet center is clearly observed.

EPMA data showed also that no measurable plutonium diffusion in the UO_2 matrix occurred in this fuel operated under normal condition. Conversely, UO2 and PuO2 homogenization has been observed at the pellet centerline of the ramped fuel rods.

Globally this programme as well as previously published data show that MOX fuel, in spite of its heterogeneous microstructure, has a similar fission gas release mechanism in the thermal regime as UO_2 fuel. Nevertheless, it seems that a small fraction of the fission gases coalesced in bubbles in the plutonium rich zones can leave the fuel for the rod void volume via cracks and open porosities of the pellet. That

could explain the slightly higher fission gas release observed for the MOX fuel which has experienced a significant local burnup in MOX agglomerates (typically after two irradiation cycles).

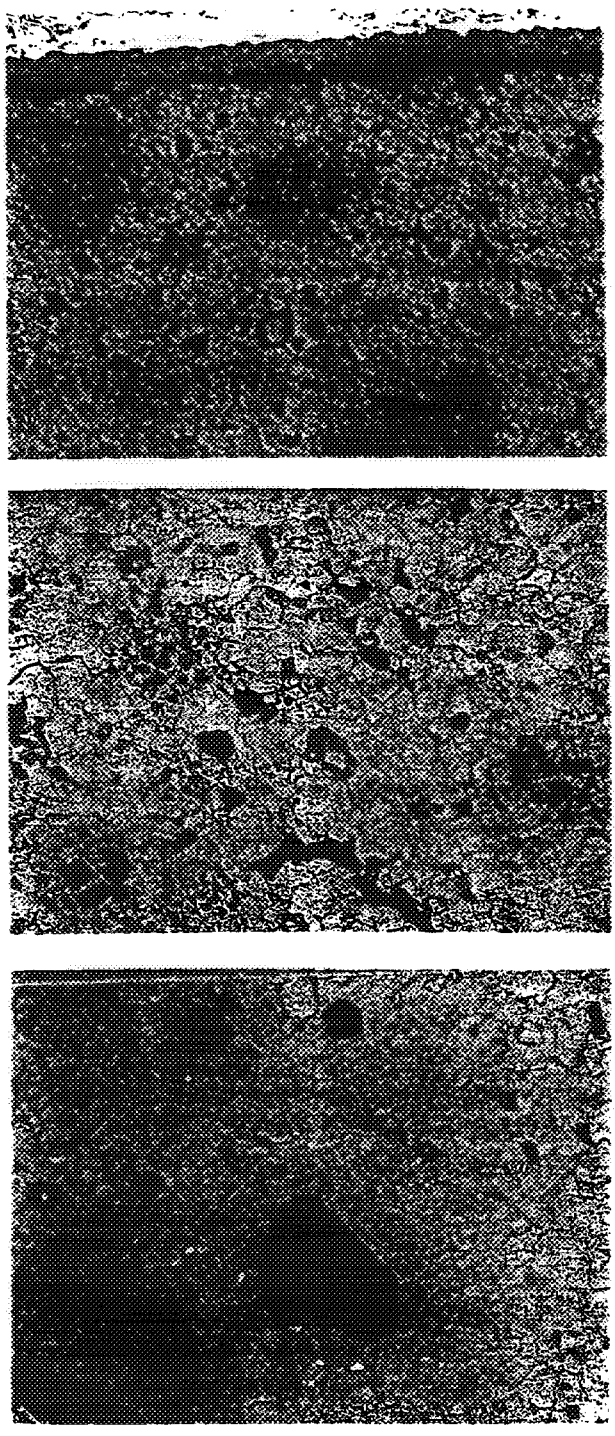
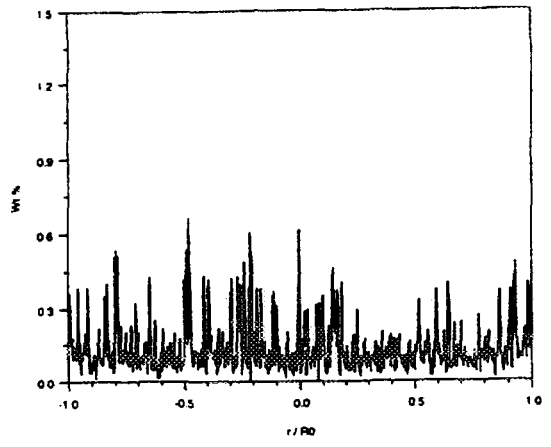
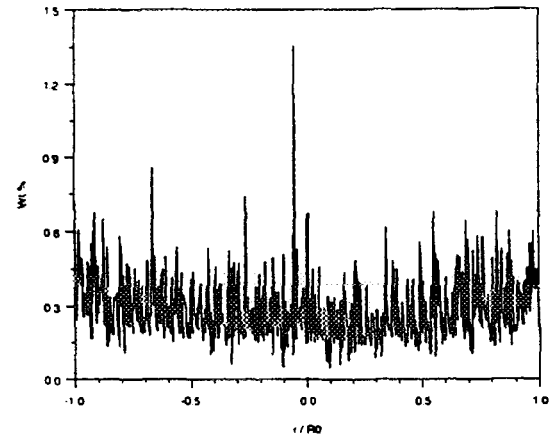


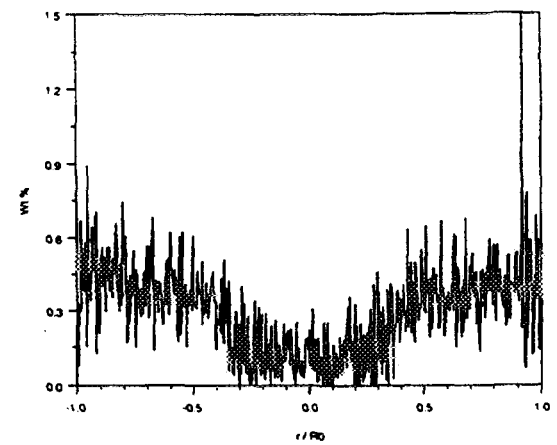
FIG. 6. Microstructure of a 3 cycle MOX fuel (local burnup : 46 Gwd/tM)



1-Cycle fuel



2-Cycle fuel



3-Cycle fuel

FIG. 7. EPMA diametral scan of the Xe element across 1, 2 and 3 cycle fuel pellets

The first data obtained from the High Burnup Programme (GRA4) showed no burnup enhancement of the fractional release. The fission gas release after 4 cycles is similar to that measured after 3 cycles due to the lower heat rate experienced by the rods during the last irradiation cycle.

Modelling of these phenomena in fuel rod design codes leads to calculations with satisfactory predictive quality.

However, investigations are still underway to take the most advantage of the large amount of data obtained in these programmes.

Table 1 : MOX fuel assembly deliveries and irradiation experience in FRANCE up to end 94

YEAR	87	88	89	90	91	92	93	94
Number of assemblies delivered	16	32	48	56	64	64	56	88
Number of reactors with Mox fuel	1	2	4	5	5	5	6	7
Irradiation Experience (cumulated number of assemblies x cycles)	0	16	64	128	256	448	624	820

CONCLUSION AND PROSPECTS

These results obtained so far confirm the good behaviour (dimensions, corrosion, internal pressure, PCI) of MOX fuel up to the typical burnup reached with 1/3 annual core management.

- Nevertheless, with the applied fuel managements, the fission gas release measured in MOX fuel was larger than in UO_2 fuel. This is mainly due to higher power histories, particularly a higher power level during the last irradiation cycle.

However, some fission gas release data obtained at higher burnup by reloading MOX fuel assemblies for a fourth cycle don't show any unexpected burnup enhancement.

The results of analytical experiments provided useful information for the confirmation or the validation of the main models simulating the behaviour of the fuel rod.

The acquired experience feed-back, together with the very satisfactory operating results, has led to advances in MOX assembly design [9]. The resulting product - AFA 2G MOX - presents an optimized zoning and includes the improvements made on the current AFA 2G UO2 product : mainly an optimized Zircaloy-4 cladding with an enhanced corrosion resistance, mixing grids with higher performance and an anti-debris device. AFA 2G MOX is designed for an assembly burn-up of 43-45 GWd/t and for an intensive grid-follow operation in order to satisfy the needs of EDF reactors.

AFA 2G MOX batches will be delivered this year to a 900 MWe EDF unit. They will be used with the newly implemented "hybrid" fuel management scheme where 3.7 % enriched UO2 assemblies are irradiated for four annual cycles and MOX assemblies remain three cycles.

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