IAEA-SM 358/15

HIGHLIGHTS ON R&D WORK RELATED TO THE ACHIEVEMENT OF HIGH BURNUP WITH MOX FUEL IN COMMERCIAL REACTORS

M. LIPPENS, TH. MALDAGUE, J. BASSELIER,
D. BOULANGER, L. MERTENS
Belgonucléaire,
Brussels, Belgium

Abstract

Part of the R&D work made at BELGONUCLEAIRE in the field of high burnup achievement with MOX fuel in commercial LWRs is made through International Programmes. Special attention is given to the evolution with burnup of fuel neutronic characteristics and of in-reactor rod thermal-mechanical behaviour

Pu burning in MOX is characterized essentially by a drop of Pu239 content. The other Pu isotopes have an almost unchanged concentration, due to internal breeding. The reactivity drop of MOX versus burnup is consequently much less pronounced than in UO_2 fuel. Concentration of minor actinides Am and Cm becomes significant with burnup increase. These nuclides start to play a role on total reactivity and in the helium production.

The thermal-mechanical behaviour of MOX fuel rod is very similar to that of UO_2. Some specificities are noticed. The better PCI resistance recognized to MOX fuel has recently been confirmed. Three PWR MOX segments pm-irradiated up to 58 GWd/tM were ramped at 100 W/cm.min respectively to 430-450-500 W/cm followed by a hold time of 24 hours. No segment failed.

MOX and UO_2 fuels have different reactivities and operate thus at different powers. Moreover, radial distribution of power in MOX pellet is less depressed at high burnup than in UO_2, leading to higher fuel central temperature for a same rating. The thermal conductivity of MOX fuel decreases with Pu content, typically 4 % for 10 % Pu. The combination of these three elements (power level, power profile, and conductivity) lead to larger FGR at high burnup compared to UO_2.

Helium production remains low compared to fission gas production (ratio < 0.2). As faster diffusing element, the helium fractional release is much higher than that of fission gas, leading to rod pressure increase comparable to the one resulting from fission gas.

1. INTRODUCTION

Plutonium recycling in LWRs has reached today its industrial maturity with MOX fuels. It is fabricated and loaded in reactor in large quantities [1]. It is recognized that the MOX fuel has an in-reactor behaviour almost identical to UO_2 [2]. This similarity is a consequence of the close properties of UO_2 and PuO_2 materials over a broad range of properties (crystallography, physics, neutronics,.), allowing easy replacement of U atoms of UO_2 lattice by Pu atoms, without deep modifications of UO_2 characteristics, and to develop a MOX industry based on UO_2 options regarding fuel fabrication and assembly/rod design, and to irradiate this MOX fuel in reactors mainly conceived to burn UO_2.

However, significant differences exist between the two fuel types. These differences have a neutronic character (mainly the larger fission and capture cross sections and the reduction of the number of delayed neutrons for Pu) or a physical character (lower thermal conductivity and lower creep strength of MOX).

As for UO_2 fuel, a demand for high burnup achievement with MOX fuel exists to reduce the electricity production costs. This demand is even greater for MOX due to a fuel cycle generally estimated to be more expensive than for UO_2. It is also economically meaningful to increase MOX burnup as a high initial **Pu** content is financially not very penalizing compared to UO_2 fuel for which the enrichment cost markedly increases with U235 content.

The present paper aims to highlight some topics on neutronic and in-reactor thermal-mechanical behaviour of MOX fuel needing appropriate consideration for high burnup achievement. These topics are illustrated by experimental results often obtained in the framework of R&D programmes organized by BELGONUCLEAJRE. The programmes are briefly reviewed.

2. BELGONUCLEAIRE R&D PROGRAMMES

As MOX fuel manufacturer, BELGONUCLEAIRE is traditionally concerned by the validation, the performance and the behaviour of MOX fuel in various irradiation conditions.

The major interest in such field is focused on a continuous updating and completeness of MOX fuel data bases through the promotion of R&D work mainly devoted to pending validation and licensing questions of plutonium recycling in commercial reactors.

Since more than 25 years, such work is proposed through a set of international programmes, the unal nf which being an attractive share of the budget and of the resulting data between several organizations directly concerned by MOX fuel and its various aspects within all phases of the fuel cycle.

Such organizations are mainly fuel Designers and Manufacturers, Research Laboratories, Utilities as well as Organizations in charge of wastes, fuel handling and storage activities.

These international programmes which are initiated, negotiated and managed by BELGONUCLEAIRE can be divided in two major classes (Table 1) :

MOX Fuel Core Physics Data	MOX Fuel Behaviour
VIP-PWR	PRIMO (PWR)
VIP-BWR	CALLISTO (PWR)
VIPO	DOMO (BWR)
ARIANE	FIGARO (PWR)
VIPEX	NOK-M109 (PWR)
REBUS	NOK-M308 (PWR)
VIPOX*	GERONIMO (BWR)
	HELISARA *

* under preparation

Table 1. MOX International Programmes

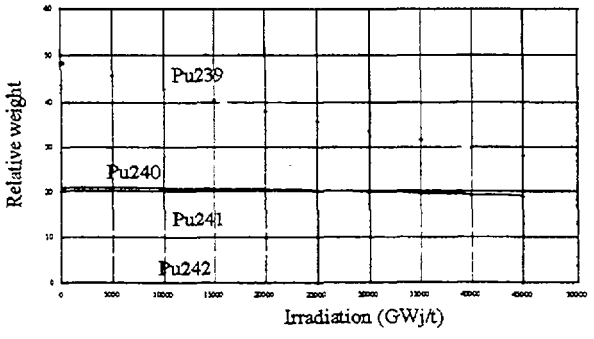

Fig. 1. Evolution of Pu content (8.9% initial) in MOX versus burnup

The first set of programmes focuses on neutronic **investigations** in direct relation to MOX fuel performance, *with two main topics.*

The *first topic,* generally organized jointly by BELGONUCLEAIRE and SCK(CEN is mainly devoted to the determination of reactor physics parameters using the VENUS critical facility located in Mol (Belgium), and simulating recent designs of PWR or BWR assemblies, and mock-ups containing more than 12 % Pu average.

- **The VIP Programme**

From 1990 to 1992, the Pu recycling in LWR's was investigated in the VENUS critical facility at SCK(CEN. The programmes called VIP (VENUS International Programme), used fuel with high Pu and Gd content. The aim of the VIP programmes was the validation of reactor codes with respect to MOX-fuel for both PWR's and BWR's. They were focused on the criticality and fission rate distribution calculations inside the MOX fuel assembly, especially at the UO_2 interface.

These programmes were divided in two parts :

VIP-PWR

mock-up 1 : All MOX 17X17 subassembly
mock-up 2 : MOX-Gd 17X17 subassembly.

VIP-BWR

mock-up 1 : All U02 : 8X8 subassembly
mock-up 2 : All MOX : 8X8 subassembly
mock-up 3 : Island MOX : 8X8 subassembly

- **The VIP0 Programme**

Since 1993, the void coefficient in LWR's has been investigated. Calculations have shown the possibility that at high plutonium contents the void coefficient could become positive. The VIPO Programme (Void Coefficient Measurement in Plutonium Mixed Oxide Lattice) was devoted to the measurements of the perturbation caused by void bubbles in a LWR reactor using high Pu enrichment (i.e. from 10 % to 15 %) and the validation of the related computer codes. A special experimental device has been developed and constructed in order to simulate a void in the reactor's core, the so-called void box.

- **The VIPEX Programme**

As a complement to the VIP PWR Programme, the VIPEX Programme provides basic MOX physics data derived from specific investigations based on critical MOX mock-up measurements in VENUS.

The parameters investigated are the effect of Am241, the Beta-effective, the flux tilt inside a MOX comer rod, the effect of simulated water density (by introduction and removal of Al microrods in the MOX lattice), the control rods worth in MOX fuel assembly and, as supplementary investigation, the detector (fission chamber) response in MOX fuel assemblies.

The second ***topic*** of these MOX neutronic evaluations includes fuel irradiation studies at high burnup with experimental determination of actinides and fission products inventory.

- [illegible]

The ARIANE Programme investigates the irradiated MOX fuel source term and will serve to validate licensing computer code of the ORIGEN type by means of an extensive programme of radiochemistry.

Attention is focused on the accuracy in determining actinides, minor actinides and fission products contents through analyzes in three laboratories (SCK(CEN, TUI and PSI) on fuels unloaded after extended irradiation in commercial reactors, such as BEZNAU-1 (MOX samples), GÖSGEN (UO_2 samples) and DODEWAARD (MOX and UO_2 samples).

The highly fissile minor actinides (Am242m, Cm243, Cm245) are investigated as well as strong alpha emitters (Pu238, Cm242). Fission products investigations focus on neutron absorbing (Sm149, Sm151, ..) or long-lived (1129, Tc99,...) nuclides. More that 20 elements and 50 nuclide contents are determined in most of the selected fuel samples.

The second set of programmes concerns complete investigations of MOX thermal-mechanical fuel behaviour under normal conditions obtained through large scale irradiations in various reactors like BR3, DODEWAARD, BEZNAU-1 and GUNDREMMINGEN and under off-normal conditions which are simulated in BR2, OSIRIS, HALDEN and HFR testing reactors.

Such programmes focus on the determination of MOX fuel rods characteristics for what concerns fission gas release and inner pressure, fuel central temperature, fuel microstructure, rod integrity and pellet-cladding interaction, measured for a wide range of irradiation conditions.

. PRIM0 and DOMO

These programmes, completed in 1994 and 1996 respectively, contributed to provide base information on fission gas release and microstructure at extended burnups in PWR and BWR respectively, and to verify that rod power failure threshold is similar or better than for UO_2 fuel. Detailed results of both programmes were published elsewhere [3,4].

. **FIGARO**

The objective of that programme, now completed, was to evaluate the thermal behaviour of MOX fuel at burnup of about 50 GWd/tM and to determine whether fission gas release threshold was different or not from UO_2. The results show essentially that the fuel conductivity degradation with burnup follows the same rule as for UO_2. About fission gas release, on-line pressure measurements indicate that the temperature threshold for gas release is close to that of UO_2 [5]. These items are detailed in § 4.1 and 5 4.4.

. NOK-M109

Taking the opportunity of the extraction of the 2 FIGARO rods from a BEZNAU-1 MOX assembly M109 irradiated during five cycles, 8 additional MOX rods were extracted as well. The PIE programme, focusing mainly on fission gas release, is now completed.

Results allow to study MIMAS MOX behaviour at high burnup, considering various power histories and fuel fabrication parameters.

. NOK-M308 (PWR) and GERONIMO (BWR)

These two programmes aim at extending the MOX performance database at still increasing burnup (58 GWd/tM peak pellet for NOK-M308, 65 GWd/tM for GERONIMO). Post irradiation examinations and ramp testing are performed on MIMAS fuel segments.

Another objective for the NOK-M308 Programme is a comparison with an alternative MOX fabrication process. These alternative MOX segments have been fabricated by PSI from the gelation process.

The NOK-M308 Programme started in 1997 with the extraction of segmented rods from the mother assembly irradiated in BEZNAU-1. The PIE programme is underway. The ramp programme has been recently completed, with no cladding failure of pelletized MOX fuel rods tested up to 500 W/cm after a power ramp at 100 W/cm min.

The GERONIMO Programme, just officially started, focuses on irradiation of full-length rods and segmented rods in GUNDREMMINGEN BWR. Extractions are foreseen in 1999 and 200 l,

- EIELISARA

Release of helium produced during irradiation significantly contributes to rod inner pressure increase. Conservative prediction of such increase includes uncertainty margins which may penalize rod operation. Uncertainties result from source term calculation, kinetics of release and helium solubility in fuel. The programme aims to perform experimental work focusing on these questions.

3. NEUTRONICS AT HIGH BURN-UP

3.1. Actinides inventory

0 Initial Pu content

Initial fissile Pu content in MOX fuel is progressively increased to achieve high burnup objective. The use of Pu from highly burned UO_2 leads also to an additional increase of the Pu content in MOX to compensate for the reduction of the initial Pu_{fiss}/Pu ratio. Typical initial Pufiss/Pu ratios range from 70 % when using Pu from low burned UO_2 (- 33 GWd/tM, 3.25 % U235) to 65 % for highly burned UO_2 (- 53 GWd/tM, 4.4 % U235). This ratio decreases very slowly for higher burnup.

Maximum achievable burnup is very depending on thermal-mechanical rod performance and maximum authorized Pu content. With a 8 % Pu content, an assembly average discharge burnup of about 45 GWd/tM can be achieved in PWR (this is the present situation of the Belgian reactors loaded with MOX). To reach 60 GWd/tM an initial 8.8 % Pu content (with 0.2 % U235) is needed. This burnup is reached also with UO_2 fuel having an initial U235 enrichment of 4.5 %.

The non linearity of initial Pu content versus burnup allows us to contemplate burnups as high as 70-80 GWd/tM with initial Pu content around - 10 % Pu. This content is well below the (conservative) 12 % upper limit beyond which the reactivity coefficient becomes positive under emptying conditions,

- Evolution with burnup

Pu burning in MOX fuel continues to reduce the fissile Pu content to a ratio of 50 % at 50 GWd/tM, starting from 70 %. At the same time, the Pu240 content increases in the isotopic composition from about 24 % to 35 %.

The evolution of the compositions with burnup is due to the marked drop of Pu239 (Figure 1) consumed by fission and capture. The concentration of isotopes of mass 240 and above is remarkably stable with burnup. This stability creates a drop of reactivity with burnup markedly less pronounced than in UO_2 fuel (Figure 2).

The fissile minor actinides Am242m, Cm243 and Cm245 originate from the evolution of actinides towards isotopes of higher mass. At high burnup they contribute significantly to the reactivity. Good experimental data and well calibrated neutronic codes on actinide chain are therefore imperative when achieving high burnup with MOX fuel.

A second consequence of the internal breeding is the rather slow drop of total Pu content with burnup. A typical relative decrease of about 30 % only is noticed, resulting essentially from the reduction of the Pu239 content.

3.2. Radial distribution

The presence of Pu in the pellet increases the thermal flux depression towards the centre, due to large absorption of thermal neutrons on fissile and fertile Pu isotopes. The radial power profile is therefore, at least at beginning of life (BOL), more depressed than in UO_2.

With burnup increase, the consumption of the initial Pu239 is therefore more pronounced at pellet periphery, leading to a higher residual Pu239 content at centre, as clearly demonstrated by SIMS measurements (Figure 3). The other isotopes show radial distributions in agreement with neutronic properties, e.g. Pu240 shows a strong depression at periphery, resulting in an increase of Pu241. The depression of Pu240 at centre results from the isotopic composition normalisation and the relative increasing abundance of Pu239. Absolute Pu240 content measurements in fact confirm that residual Pu240 content increase towards pellet centre as expected.

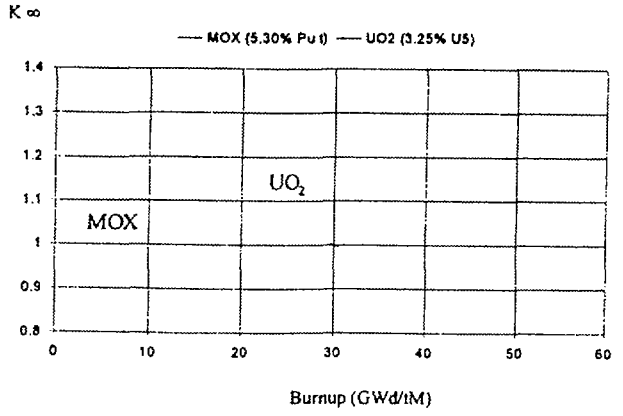

Fig. 2. Drop of reactivity as a Function of Burnup for MOX & UO_2 Fuel

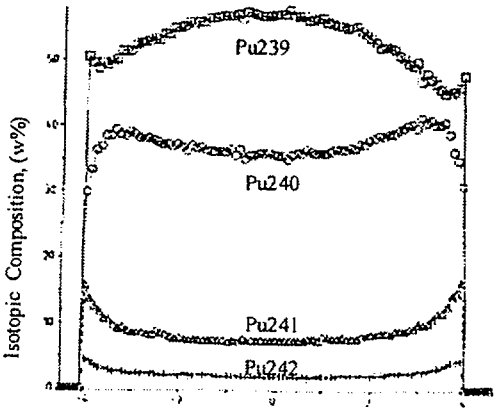

Fig. 3. Radial distribution of Pu isotopes across pellet measured by SIMS on irradiated MOX

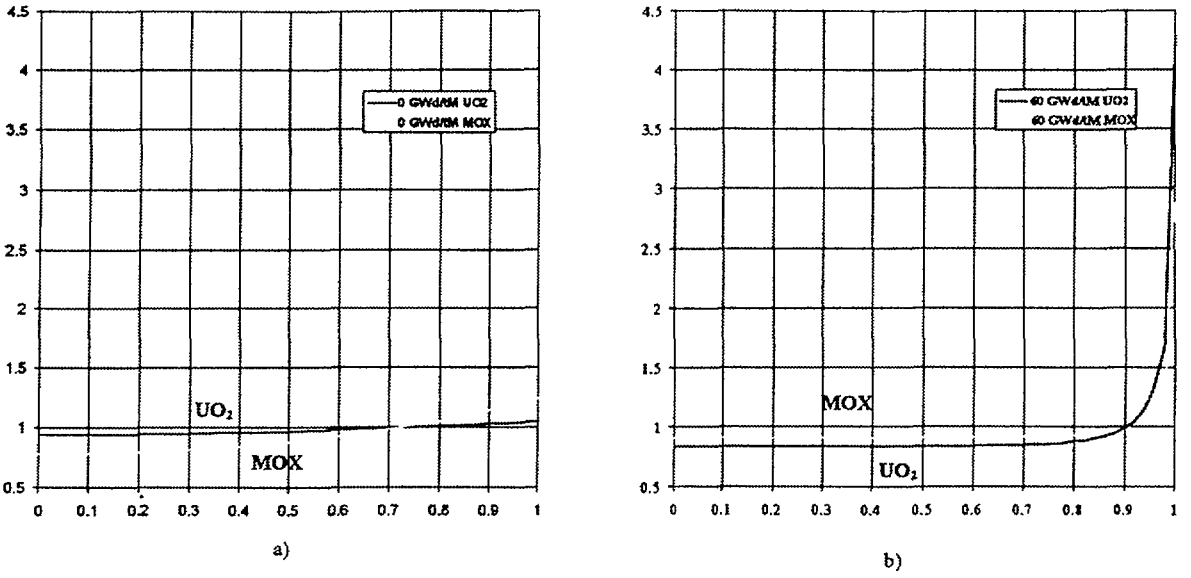

Fig. 4. Radial distribution of power in UO_2 (4.5% U5) and MOX (0.2% U5, 8.8% Pu) fuel pellet at a) 0 and b) 60 GWd/tM in PWR

At pellet rim, an increase in Pu239 content is observed. This build-up exists in MOX as well as in UO_2 fuel. Calculations indicate that the breeding of U238 in Pu239 is almost identical in MOX and UO_2. However, the net Pu239 production in MOX is influenced by the balance between the enhancement of the number of epithermal neutrons due to higher neutron production per fission, and the presence of Pu242 which absorbs neutrons on resonances overlapping with resonances of U238. Calculations suggested that breeding at rim is slightly less important in MOX than in UO_2.

Figure 4 illustrates the calculated evolution of radial power profiles versus burnup for UO_2 and MOX fuels. At BOL, the power is more depressed in MOX compared to UO_2, whereas at end of life (EOL), the higher residual Pu content at centre leads to higher local power in MOX fuel. The reduced breeding at MOX pellet periphery at high burnup contributes also to slightly increase the relative power at pellet centre.

3.3. The fission products

Fission yields for Pu239 and Pu241 are generally close to that of U235. Significant differences are however noticed for the light fission products, the peak of which is shifted towards higher masses. Practically, this means a decrease of Kr (leading to a Xe/Kr ~15 compared to 7 for UO_2), the stability of Mo, and an increase of Ru. The total fuel solid swelling versus burnup is therefore expected to be almost identical to UO_2, with enhanced Ru content in metallic inclusions created during irradiation.

3.4. Helium production

Helium is produced in MOX fuel in greater quantity than in UO_2. The main contribution to helium production in MOX is Cm242 (half life 0.45 years). The quantity produced is very depending on initial Pu content and quality, and on burnup (Figure 5). The total amount of produced helium remains always small compared to Xe and Kr contents, with a He / (Xe + Kr) ratio of 0.07 at 40 GWd/tM and 0.18 at 62 GWd/tM (according to assumptions of Figure 5).

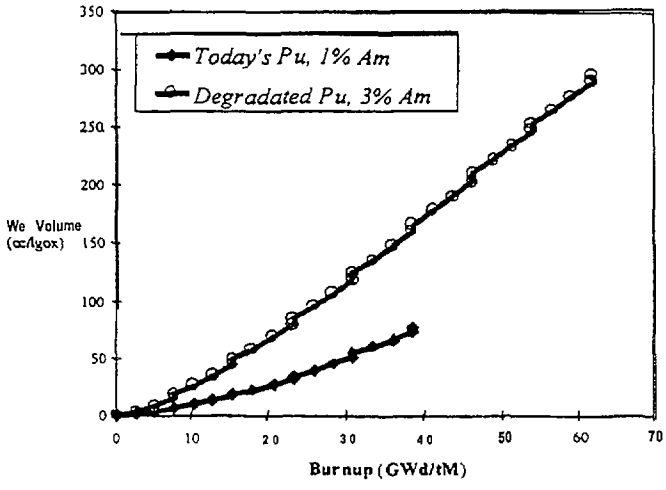

Fig. 5. Present & future production of helium in LWR MOX fuel

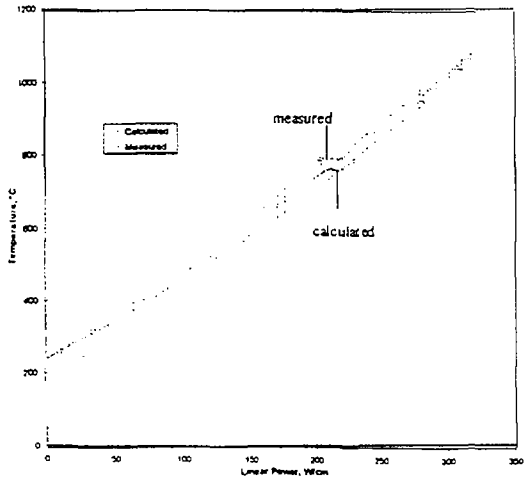

Fig. 6. FIGARO Experiment (IFA - 606) Temperature in the annular region during phase 2 of HALDEN irradiation

4. THERMAL MECHANICAL BEHAVIOUR

4.1. Thermal conductivity

A brief review of the thermal conductivity of MOX fuel has recently been published elsewhere [6]. This review concludes that the presence of small quantities of PuO_2 in UO_2 slightly decreases the thermal conductivity. Assuming that the thermal resistivity can be represented below 1500°C by the expression $1 / k = A + BT$, the A and BT terms representing phonon scattering and phonon-phonon scattering respectively, then the presence of Pu increases the A and B terms due to reduction of atomic radius and of melting temperature respectively (Table 2). The overall conductivity decrease is small, typically 4 % for 10 % Pu/U+Pu.

With burnup, the conductivity of MOX fuel degrades due to irradiation damage and accumulation of fission products in stable or dynamic solution. The degradation is expected to follow quantitatively that of UO_2 as MOX is composed by UO_2 for more than 90 % and as the fission yields are close for Pu239, Pu241 and U235.

Measurements of fuel central temperature on MOX irradiated up to 50 GWd/tM were made in the framework of the FIGARO Programme [5]. Temperature calculations performed with the COMETHE-4D Code show that measured temperatures can be well reproduced assuming that the conductivity degradation with burnup in MOX is the same as in UO_2 (Figure 6).

4.2. Fuel central **temperature**

Calculations of fuel central temperature versus burnup are summarized in Table 3. At BOL, the MOX fuel operates with central temperature lower than in UO_2 (the difference reaches about 30°C at 250 W/cm), due to higher depression of radial power at centre. At 80 GWd/tM, the reverse situation is noticed, MOX operates about 85°C above UO_2 fuel at 250 W/cm, due to the increasing residual Pu239 towards centre.

Material	UO_2	PuO_2
crystallographic structure	Fluorite SC : O^{2-} ; FCC : M^{4+}	
lattice parameter (Å)	5.470	5.396
melting temperature (°C)	2840	2290
cation atomic radius (Å)	0.97	0.93
density (g/cm³)	10.96	11.45
molecular mass	~ 270	~ 271

Table 2. Comparison of UO, and PuO_2 properties having an influence on thermal conductivity

k type	k (UO_2, Bu = 0)	k* (UO_2, Bu)	k* (MOX, Bu)
$\delta(T_c - T_s)$(°C) (2)			
BOL			
150 W/cm	-25	-25	-16
250 W/cm	-51	-51	-33
EOL			
150 W/cm	11	17	27
250 W/cm	24	65	85
T_c(°C) (3) EOL			
150 W/cm	570	840	850
250 W/cm	770	1250	1260

(1) UO_2 (4.5 % U5) and MOX (0.2 % U5 ; 8.8% Pu) up to 60 GWd/tM
(2) $\delta(T_c - T_s) = (T_c - T_s)$ MOX $- (T_c - T_s)$ UO_2
$T_s \sim 500$° C BOL - open initial gap
350° C closed gap (400° C at EOL/250W/cm for k = k*)
(3) $T_c = (T_c(MOX) + T_c(UO_2))/2$

Table 3. Comparison between MOX and UO, fuel central temperatures for different types of MOX thermal conductivities (1)

4.3. Pellet cladding mechanical interaction

Post-irradiation examinations on several hundreds MOX and UO_2 rods indicate that both fuels have the same level of mechanical interaction with the cladding. This result can be explained once again by the fact that MOX is made for more than 90 % of UO_2 and generates nearly the same fission products.

In ramp conditions, the superiority of MOX over UO_2 has been demonstrated [7]. This superiority has recently been confirmed in the M308 International Programme with tests on MOX fuel fabricated following the MIMAS process. Three PWR segments pre-irradiated up to 58 GWd/tM were ramped at 100 W/cm min., followed by a hold of 24 hours respectively at 430, 450 and 500 W/cm. None of the segments failed.

4.4. Fission gas release (FGR)

Fission gas released from fuel is extensively investigated, as being a key parameter in high burnup achievement. A topic largely debated is the possible enhancement of FGR in MOX fuel, especially in case of Pu distributed in Pu-rich zones across pellet. Early results (accumulated in the years 1970-80) suggested an acceleration of FGR in MOX fuels compared to UO_2. These observations were made on unpressurized fuel rods loaded with unstable fuel. Moreover, the concerned fuel was prepared by mixing UO_2 granules with PuO_2 powder, giving a highly heterogeneous fuel, with large open and flat porosity and preferential path for gaseous fission products. Such fuel promoted FGR and thermal feedback.

Since these early observations, the fabrication process has been changed ; numerous experimental results concerning more recent fuel fabrications were obtained and detailed neutronic studies were performed.

The experimental results of the PRIM0 and DOMO international programmes in which various fuel types were irradiated and examined, conclude that FGR in MOX and UO_2 are similar and governed mainly by power history [3, 41.

French results indicate [8] that MOX fuel irradiated three cycles show an acceleration of FGR compared to UO_2 fuel. Analysis of that situation reveals that FGR increase can be attributed to a large extent to the higher linear power sustained by MOX fuel in these fuel assemblies. This assumption is confirmed in the same paper by examination results of a MOX assembly loaded at core periphery for a fourth cycle and showing no additional FGR.

Study of fuel operating conditions conclude that MOX and UO_2 fuels are indeed never strictly irradiated in the same power conditions, due to smaller drop of reactivity versus burnup for MOX fuel. This promotes – depending on core reshuffling policy – higher linear power in MOX late in life compared to UO_2. On the other hand, with the same rating, MOX fuel sustains larger central temperature (§ 4.1) with increasing burnup, favouring also FGR.

The conclusions drawn from these studies and our programmes are that FGR in MOX and UO_2 fuels are governed by the same parameters, i.e. rod linear power, fuel surface temperature, central temperature and burnup, and fuel microstructural parameters such as open porosity and grain size. The same conclusion was reached elsewhere too [9].

The COMETHE code includes model of FGR developed and calibrated for UO_2 fuels. It has been used to simulate the pressure increase in MOX fuel segments irradiated in Halden reactor in the framework of the FIGARO Programme [5] (Figure 8). Onset of FGR is observed around 1200°C, which is close to the Halden threshold for FGR at a burnup of 48 GWd/tM. Total FGR observed at end of test reached 12 %, which is also well reproduced by the code. The inner pressure however is overestimated, due to overprediction of helium release during test.

One major conclusion of our studies on FGR is that the MOX fuel releases more than UO_2 because it sustains generally larger central temperatures. For this reason, achievement of burnup with MOX (and UO_2) well above the present limits has required and will continue to require optimization and changes in

fuel fabrication, fuel design and design criteria, and operating conditions. A non exhaustive list of actions needing particular attention is :

increase of rod free volume at fabrication
diminish excessive free volume reduction during irradiation (cladding creep-down, fuel accommodation,)
optimize core reshuffling strategy (reduction of maximum power late in life,)
develop fuels more resistant to FGR (large gram,).
accept new design criteria (non lift-off, .).

4.5. Microstructure

Microstructure of MOX fuel is governed to a large extent by the UO_2 matrix in which PuO_2 is finely dispersed at fabrication. The MOX fuel will therefore sustain during irradiation the common phenomena of densification, swelling accommodation, and intergranular and intragranular porosity development associated to fission gas production and accumulation.

MOX fuels obtained by mechanical process show a fine dispersion of Pu across pellet, with the presence of some microscopic Pu-rich zones of Pu content larger than pellet average. The detailed study of such fuels provides useful information on evolution of fuel at high burnup in the Pu-rich zones and in the UO_2 matrix. These observations can to some extent be applied to the rim structure occurring in UO_2 when local burnup reaches 60-80 GWd/tM.

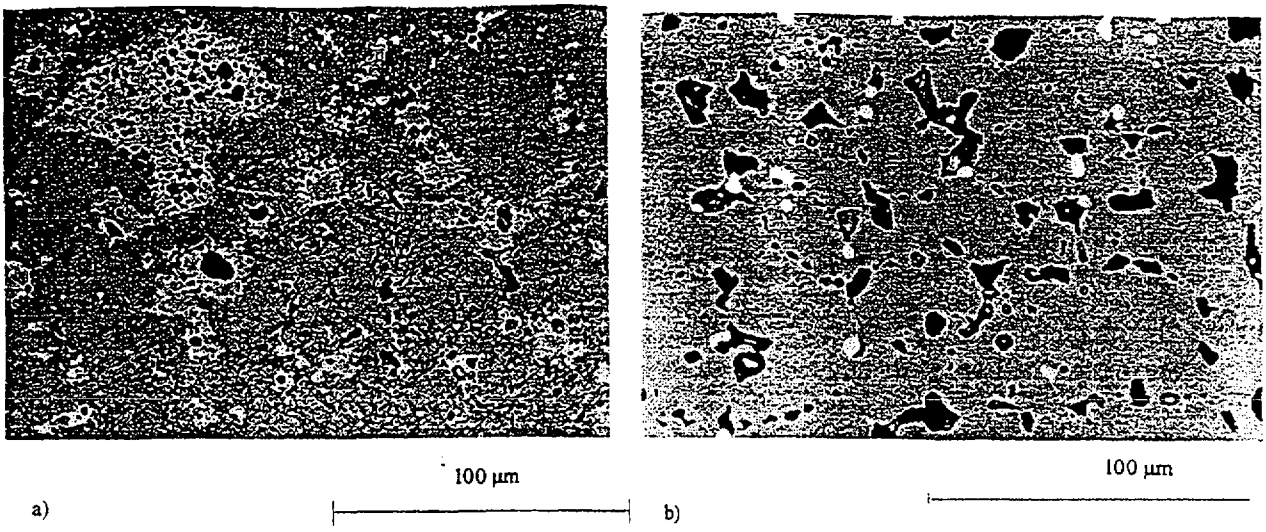

Fig. 7. Microstructure evolution of Pu-rich zone irradiated at (a) low and (b) high temperature

The negligible solubility of fission gas leads, in the Pu-rich zones irradiated at low temperature, to the formation of intragranular nanometer-sized bubbles. With burnup increase, they reach a micron size and become visible by optical microscopy (Figure 7.a). When irradiation proceeds, these bubbles continue to grow whereas their density number decreases. This process is pursuing as long as the temperature remains low (< 1000°C), and for burnup much beyond 200 GWd/tM.

Grains subdivide in Pu-rich zones reaching a submicron size, typical of the "rim structure" [10]. This structure is observed for temperature as high as 1000°C. Formation of rim structure at such a temperature is noticed also for UO_2 [11].

The extrapolation of this result to UO_2 fuel indicates that the existence in UO_2 of rim structure over a depth of about 300 μm and its rapid disappearance towards pellet centre must be attributed to local burnup drop and not to the temperature increase towards centre as long as thermally assisted diffusion processes remain negligible.

The structure of the Pu-rich zones changes when fuel operates above some temperature, with a restructuring similar to UO_2 fuel operating in the same conditions [10]. At sufficiently high temperature, grain growth and development of intergranular porosity typical of UO_2 are noticed for MOX as well (Figure 7.b).

About the rim structure development in MOX fuel, the neutronic studies conclude to accumulation at pellet periphery of burnup similar to UO_2. In the MIMAS fuel, the Pu is distributed at fabrication between the UO_2 matrix where it is finely dispersed, and few Pu-rich zones. The average burnup reached in UO_2 matrix is therefore lower than pellet average, which has for effect to postpone the development of a continuous rim structure at pellet periphery.

4.6. Helium release

Helium is produced in MOX fuel in significant quantity, but lower than that of noble fission gas (§ 3.4). Experimental inventory of helium content in free volume of irradiated MOX and UO_2 - pre-pressurized and unpressurized - rods has been made, with the following conclusions :

- excess of helium compared to as fabricated content is found in low-pressurized MOX rods, increasing with rating and burnup,
- increase in helium partial pressure can be of the same order as pressure increase due to noble fission gas,
- helium balance is often negative at low burnup,
- no excess of helium is found in rods pressurized at 20 bars,
- UO_2 rods show a negative balance of helium,
- dispersion in obtained results is large.

The absence of helium content increase in free volume in pre-pressurized rods over a broad range of burnup has been observed for MOX rods as well [12].

Study of helium behaviour made elsewhere shows that helium diffusion coefficient in UO_2 is several orders of magnitude larger than that of noble fission gas [13] and that helium is highly soluble in UO_2 [14]. In view of that, the above experimental observations were interpreted :

- large dispersion on helium balance comes from uncertainty on initial rod free volume and pressure,
- negative helium balance for UO_2 could be attributed to infusion of helium in fuel during irradiation,
- helium is not released from fuel as long as its content does not exceed the solubility limit, which - depends on helium partial pressure in rod,
- fraction of released helium is depending on operating conditions, and can be correlated to fraction of FGR.

Modelling of helium release has been made on base of experimental results and an empirical correlation set up between helium and FGR release [15]. Simulation of helium release during a BWR MOX irradiation is shown at Figure 9. Conservative results indicate an helium partial pressure increase comparable to fission gas partial pressure. The conservative curve corresponds to use of the envelop correlation inferred from experience. Best estimate result is unaccurate as many uncertainties exist in helium production term, solubility effects and extent of diffusion.

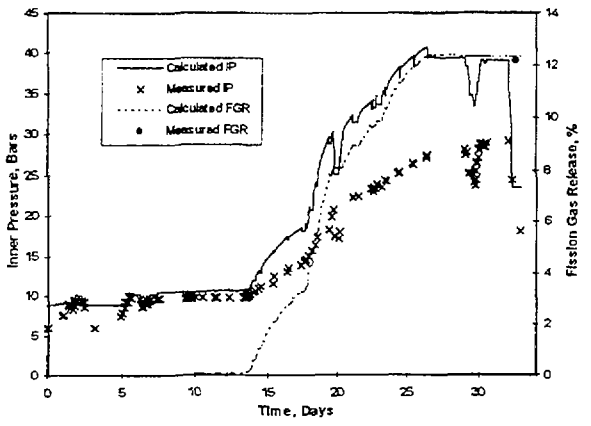

Fig. 8. Calculated and measured FGR and inner pressure in FIGARO experiment (IFA 606/phase 2)

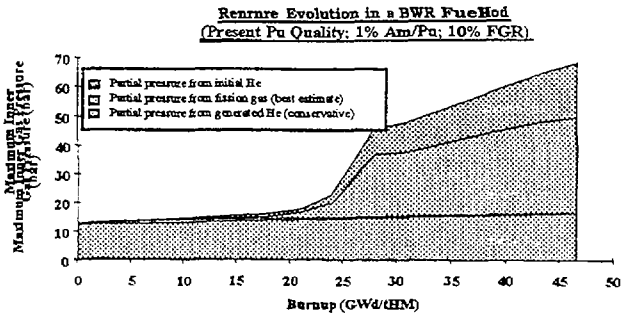

Fig. 9. Pressure Evolution in a BWR Fuel Rod (Present Pu BOL 1% Am/Pu; 10% FGR)

5. Conclusions

UO_2 and PuO_2 materials have properties relevant to in reactor performance sufficiently close to each other to fabricate (UO_2-PuO_2) fuel and to burn it according to scenarios largely conceived to burn UO_2. The R&D efforts made to achieve high burnup with MOX fuel are therefore largely similar to UO_2 activities, with special emphasis however on the evolution of the actinide chain and its consequences, and FGR. The conclusions of our observations on MOX behaviour at high burnup are :

1□ With burnup, the Pu content is modified due to a drop of Pu239. The other isotopes remain almost constant,
2□ The reactivity drop with burnup is much less pronounced than in UO_2,
3□ The minor actinides Am242m and Cm243/245 start to play a role in total reactivity with a contribution of several %,
4□ Difference in radial distribution of Pu burning across pellet results in high residual Pu at pellet centre, and thus to higher fuel central temperature with burnup increase,
5□ The presence of Pu slightly reduces the fuel thermal conductivity, about 4 % for 10 % Pu,
6□ The burnup degrades the fuel thermal conductivity as in UO_2,
7□ MOX fuel operates under different temperature conditions than UO_2 due to conclusions 2, 5 and 6. This increases FGR in MOX and must be taken into account for high burnup achievement,
8□ PCMI in steady state conditions is similar to UO_2. The behaviour in ramp condition is favourable to MOX fuel,
9□ The microstructure evolution during irradiation follows that of UO_2 which represents 90 % of the fuel. Investigations of microscopic Pu-rich zones included in pellet indicate a rim type structure. With burnup increase, such zones show pore size increase and density number decrease. Such a structure is observed up to about 1 000°C,
10□ The decay of Cm242 leads to observable helium production and release, especially for lowly pressurized high rated fuel.

In view of these conclusions, the achievement with MOX fuel of burnups higher than achieved in routine today in LWRs is feasible. Recommendations for further R&D work are :

- accumulate experimentally burnups of at least 80 GWd/tM at low power
- obtain accurate data on actinide chain
- confirm fuel central temperature evaluation
- obtain data on helium production, solubility and release
- develop fuels having improved gas retention
- study rods in non lift-off conditions.

References

[1] DERAMAIX, P., et al., "MOX Fuel Fabrication and In-Reactor Performance : Realizations and Prospects", RECOD'98 (Proc. Int. Conf. Nice, France, October 1998).-(in press).

[2] LIPPENS, M., et al., "MOX Fuel Performance – BELGONUCLEAIRE Views", Paper presented at the OECD/NEA Workshop on the Physics and Fuel Performance of Reactor-Based Plutonium Disposition - Paris, France, September 1998.

[3] BASSELIER, J., et al., "Validation of MOX fuel through recent BELGONUCLEAIRE international programmes", Recycling of Plutonium and Uranium in Water Reactor Fuel" (Proc. IAEA TCM Windermere, UK, July 1995), IAEA, Vienna, IAEA-TECDOC-941 (1997) 277.

[4] ASAHI, K., et al., "Irradiation and post-irradiation testing program of BWR MOX fuel rods", LWR Fuel Performance (Proc. Int. Topical Meeting West Palm Beach, Florida, USA, April 1994), ANS (1994) 726.

[5] MERTENS, L., et al., "The FIGARO programme : the behaviour of irradiated MOX fuel tested in IFA-606 experiment, description of results and comparison with COMETHE calculation", Paper presented at the Enlarged HRP Meeting - Lillehammer, Norway, March 1998.

[6] LIPPENS, M., et al., "Comparative thermal behaviour of MOX and UO_2 fuels", Thermal Performance of High Burnup LWR Fuel (Proc. Int Seminar Cadarache, France, March 1998) OECD/NEA (1998) 243.

[7] ROEPENACK, H., et al., "Development of thermal Plutonium recycling", Nuclear Technology, 77 (1987) 175.

[8] BLANPAIN, P., et al., "Plutonium recycling in French power plants : MOX fuel irradiation experience and behaviour", Recycling of Plutonium and Uranium in Water Reactor Fuel (Proc. IAEA TCM Windermere, UK, July 1995), IAEA, Vienna, IAEA-TECDOC-941 289.

[9] WALKER, C.T., et al., "MOX fuel irradiation behaviour : Results from X-ray microbeam analysis", Recycling of Plutonium and Uranium in Water Reactor Fuel (Proc. IAEA TCM Windermere, UK, July 1995), IAEA, Vienna, IAEA-TECDOC-941 (1997) 301.

[10] VERWERFT, M., et al., "On the thermal evolution of Pu-rich agglomerates in MOX", MOX Fuel Cycle Technologies for Medium and Long Term Deployment (Proc. Int. Symposium Vienna, 17-21 May 1999) (these Proceedings).

[11] KINOSHITA, M.; et al., "Temperature and Fission Rate Effects on the Rim Structure Formation in a UO_2 Fuel with a Burnup of 7.9 % FIMA", J. Nucl. Mat. **252** (1998) 71-78.

[12] BLANPAIN, P., et al., "Recent results from the in-reactor MOX fuel performance in France and improvement program", LWR Fuel Performance (Proc. Int. Topical Meeting Portland, Oregon, USA, March 1997), ANS (1997) 39.

[13] STARK, W.A., Jr., "Helium release from $Pu^{238}O_2$ microspheres", Nuclear Metallurgy, Vol. 17, Part 2 (1970) 554.

[14] GRIMES, R.W., "Simulating the behaviour of inert gases in UO_2. Fundamental Aspects of Inert Gases", Plenum Press, New York (1991).

[15] BILLAUX, M., et al., "Production of helium in UO_2-PuO_2 mixed oxide fuel", Water Reactor Element Computer Modelling in Steady State and Accident Conditions (Proc. IAEA TCM Preston, UK, September 1988), Rep. IWGFPT No. 32, IAEA, Vienna (1989) 182.