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Post-irradiation examinations of THERMHET composite fuels for transmutation

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Abstract

The thermal behaviour of composite targets dedicated to minor actinide transmutation was studied using THERMHET (thermal behaviour of heterogeneous fuel) irradiation in the SILOE reactor. Three inert matrix fuel designs were tested (macro-mass, jingle and microdispersion) all with a $MgAl_2O_4$ spinel inert matrix and around 40% weight of UO_2 to simulate minor actinide inclusions. The post-irradiation examinations led to a new interpretation of the temperature measurement by thermocouples located in the central hole of the pellets. A major change in the micro-dispersed structure was detected. The examinations enabled us to understand the behaviour of the spinel during the different stages of irradiation. They revealed an amorphisation at low temperature and then a nano re-crystallisation at high temperature of the spinel in the micro-dispersed case. These results, together with those obtained in the MATINA irradiation of an equivalent structure, show the importance of the irradiation temperature on spinel behaviour.

1. Introduction

In order to optimise the transmutation efficiency and the loaded mass of minor actinides, many fuel concepts have been evaluated. One of them involves recycling with a large amount of minor actinides embedded in an inert matrix target [1].

The THERMHET irradiation [2,3] (thermal behaviour of heterogeneous fuel) was carried out within the framework of the French Atomic Energy Commission (Commissariat à l'Energie Atomique – CEA) programme [4] dedicated to the separation and transmutation of minor actinides. It focused on the in-pile measurement of the thermal behaviour of composite fuels at beginning of life and on the testing of various microstructures.

Three different inert matrix structures were tested and the collected results were used to qualify a thermal modelling of these structures. In addition to this modelling and exploitation of the in-pile temperature measurements, post-irradiation examinations were conducted. The main findings derived from these examinations are presented hereafter. A comparison is made with results taken from another irradiation of a minor actinide separation and transmutation programme, namely the MATINA pin 14 which was irradiated in the Phénix reactor. The comparison was carried out in view of the fact that its microstructure is close to that of the THERMHET micro-dispersion fuel structure [5].

2. The THERMHET and MATINA irradiations

2.1. Fuel design

All the structures tested in the THERMHET irradiation and in the MATINA pin 14 contain $MgAl_2O_4$ as an inert matrix and 19.5% ^{235}U enriched UO_2 to simulate a minor actinide material. The purpose of this is to test the effect of fission and to achieve a representative temperature distribution.

In THERMHET macro-mass fuel, UO_2 macro-masses of about 200 μm in diameter with almost

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spherical particles were dispersed at 41.5 wt% in the $MgAl_2O_4$ spinel matrix.

In THERMHET jingles fuel, there were 41 wt% of UO_2 macro-masses of the same size as in the THERMHET macro-mass fuel, called 'jingles' because of a gap of about 10 μm wide between the inclusions and the spinel matrix.

In THERMHET micro-dispersion fuel, there was a micro-dispersion of 40.2 wt% of UO_2 particles <20 μm in diameter in the spinel matrix.

A micro-dispersion fuel of 40 wt% of UO_2 particles in a $MgAl_2O_4$ spinel matrix was included in the MATINA experiment, pin 14.

Each one of the THERMHET pins was made up of five solid pellets at the bottom of the fissile stack, and of 8 hollow pellets leaving room for a thermocouple. The external diameters of the pellets and the diameters of the cladding were the same as those found on a standard FBR pin in the Phénix reactor.

2.2. Irradiations

Irradiation of the THERMHET pins took place in 1997 in the CEA SILOE reactor inside a TANOX [6] device. Due to the fact that the macro-masses and the jingle concept fabrications were not optimised at that time, the distribution of the inclusions was not homogeneous in the pellets. This caused significant variations in the local linear power. The global power in the TANOX device was adjusted following the measured temperature. As it was impossible to derive precisely the actual maximal temperature in the pins from the thermocouple measurement during irradiation due to the axial heterogeneity in fissile content, a nominal target temperature of 858 K measured by the thermocouple in the first two pins was chosen. Moreover, the irradiation was separated into two cycles with lower power at the end of the first cycle and withdrawal of the first two pins before the second cycle. In order to ensure a higher temperature (more representative of an FBR fuel) for the THERMHET micro-dispersion pin, the nominal power was increased during the second cycle (Fig. 1).

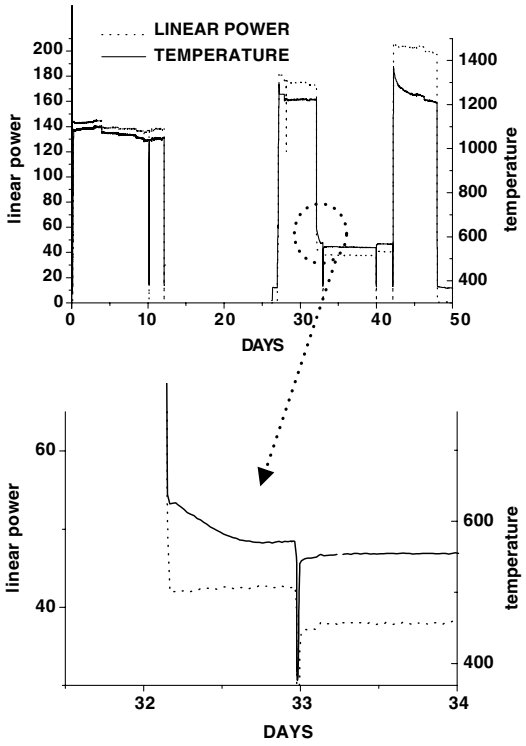

Fig. 1. Linear power and thermocouple measurement for the micro-dispersion THERMHET fuel.

The irradiation parameters are summarised in Table 1.

2.3. Temperature measurements

In the macro-mass and jingle structure cases, no peculiarities are observed in the temperatures measured during irradiation [2] and for the same linear power, the temperatures measured on the composite fuels are all different and lower than the temperature measured in the UO_2 reference fuel. The thermal calculations [2,3] matched the measurements for the jingle structure. For

Table 1
Irradiation parameters of the THERMHET and MATINA 06 pins

	THERMHET				MATINA 06
	Cycle 1			Cycle 2	
	Macro-masses	Jingles	Microdispersion	Microdispersion	Microdispersion
Fluence ($E > 0.1$ MeV)	2.8×10^{23} n/m^2	2.5×10^{23} n/m^2	4.1×10^{23} n/m^2	14×10^{23} n/m^2	1.95×10^{26} n/m^2
Burnup	0.38% FIMA	0.34% FIMA	0.50% FIMA	1.3% FIMA	1.27% FIMA
Irradiation duration	21 EFPD	21 EFPD		42 EFPD	61 EFPD
Maximum linear power	<110 W/cm	<95 W/cm	<150 W/cm	<200 W/cm	48 W/cm
Central temperature range (calculated in solid pellets)	<1073 K	<1073 K	1073 K	Between 573 and 1473 K	~1673 K

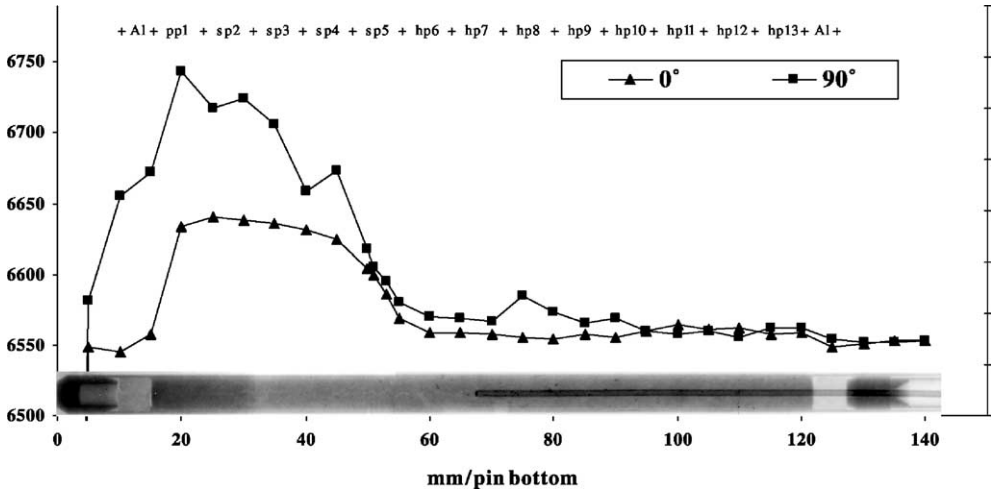

Fig. 2. Diameter measurements along two generatrix on THERMHET micro-dispersion pin and neutronograph after irradiation. Position of the alumina pellets (Al) the solid pellets (sp) and the hollow pellets (hp) before irradiation.

the macro-masses, the calculations overestimated the temperature by about 12%. This certainly comes from the heterogeneous axial and radial distribution of macro-masses and a poor evaluation of the thermocouple's exact axial position during irradiation.

The macro-homogeneity of the micro-dispersed structure implies a smooth axial power profile. It is one of the reasons why the calculation of the temperature during the first cycle was in a very good agreement with the measurements. Discrepancies are observed during the second cycle, especially at its end (Fig. 1). Indeed, during this last part of the irradiation, the measured temperature decreased much more than the local power variation would imply. Another difference between the expected temperature and the measured one is observed during the second cycles at the beginning of a low linear power period. During this period, the measured temperature decreases while the linear power is stabilised at a low level, with the stabilisation of the temperature occurring about 14 h after the linear power one.

3. PIE results

3.1. Non-destructive examinations

The neutron radiographs did not detect any variations in the case of the macro-masses and pins. The gamma scanning of these pins showed the presence of the measured isotopes proportionally to the axial heterogeneous distribution of the UO_2 particles. No evolution in the diameters of these pins was detected.

In the case of the microdispersion, all the non-destructive examinations showed a major swelling in the fuel:

- There was a clear diameter increase (Fig. 2). This increase is higher at the bottom of the solid pellet stack than in front of the hollow pellet stack. Between those two parts, at the top of the solid pellet stack, a smooth transition zone is observed. This difference between solid pellets and hollow pellets in front of the thermocouple wire must be attributed to the additional gap that has to be filled around the thermocouple for the hollow pellets and to the additional material at the centre of the solid pellet. This diameter increase actually shows that there was a fuel volume expansion during irradiation.
- The neutron radiographs before and after irradiation (Fig. 3) show that there was an axial expansion of $\Delta L/L = +1.1\%$ of the fissile stack. These neutron radiographs also show that the central hole located under the thermocouple seems closed apart from an axial crack. The fuel to cladding gap seems closed all along the pin. This has been confirmed by the metallographs.

If we consider the whole volumes before irradiation and after irradiation, taking into account the axial expansion and a clad thermal expansion, the mean expansion was about 15% of the initial fuel volume when all the free volumes of the stack were filled with fuel.

If we now consider the shape of the pin at the top of the solid pellet stack, we can deduce that there was a special axial expansion from these solid pellets towards the bottom of the hollow pellets owing to the great volume to be filled under the thermocouple. This axial transfer occurred during irradiation.

- Axial gamma scanning confirms this axial movement as the step in the axial gamma profiles are in front of

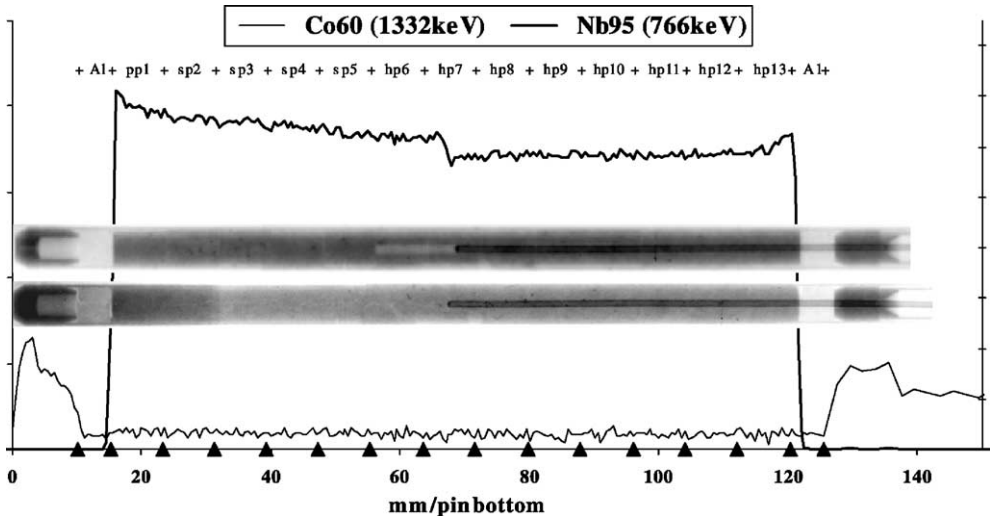

Fig. 3. Gamma scanning of Nb95 and Co60 on THERMHET micro-dispersion pin and neutronograph before and after irradiation. Position of the alumina pellets (Al) the solid pellets (sp) and the hollow pellets (hp) before irradiation.

the bottom of the thermocouple and not in front of the initial interface between solid and hollow pellets (Fig. 3). The lack of material under the thermocouple has been reduced.

Therefore, non-destructive examinations on the microdispersion pin show that during irradiation there was a major volume increase (~15%) of the fuel, with some axial transfers. It filled all the free volumes and led to an axial lengthening of the stack and pin diameter increase.

3.2. X-ray diffraction on THERMHET macro-mass fuel and micro-dispersion

The X-ray diffraction acquisitions on the micro-dispersion structure and on the macro-mass structure, that were not performed for [3] are compared in Figs. 4 and 5.

A shielded Bragg–Brentano (Θ–2Θ) X-ray diffractometer equipped with a germanium back monochromator was used. This removes the copper Kα2 peaks. The samples were axial cross section solid samples.

The comparison between the diffraction patterns of the macro-mass structure and those of the micro-dispersed one, both after irradiation, first show that the peaks of the spinel almost disappeared in the micro-dispersed case. It may be amorphisation or re-crystallisation in nanostructures. On the diffraction patterns, the usual background evolution due to amorphisation is not detectable but the already high background level of these diffraction patterns may hide it. Some spinel peaks can still be detected with very low intensity. The deduced lattice parameter is very close to that of the starting material.

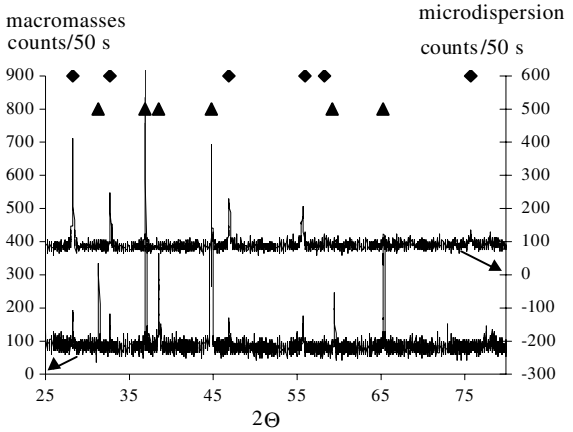

Fig. 4. X-ray diffraction patterns on irradiated micro-dispersion and macro-mass fuels (axial cuts): (▲) position of the spinel peaks, (◆) position of the UO_2 peaks. In order to separate the backgrounds, two ordinates have been used.

On the X-ray diffraction pattern of the macro-mass structure, the spinel lattice parameter is also very close to that of the starting material. Not far below the (4 0 0) peak of the spinel, an extra diffraction line is found (Fig. 3). It cannot come from the spinel, nor from the UO_2. This peak was also detected on the XRD pattern of Kr 412.2 MeV 10^{14} ions/cm^2 irradiated spinel [7]. It shows that there was a partial order to disorder phase transition. The peak broadening was understood as the sign of a local strain field. This new phase was also reported in [8].

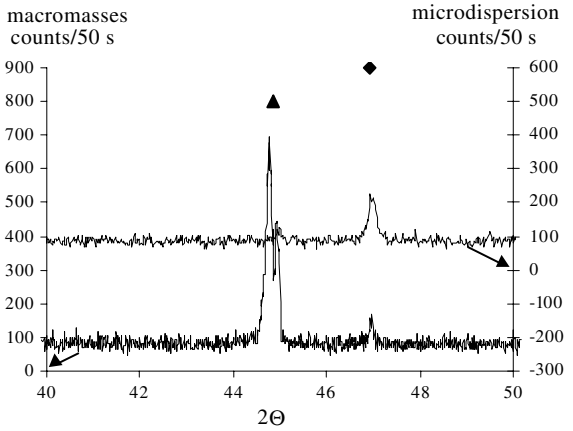

Fig. 5. X-ray diffraction patterns on irradiated micro-dispersion and macro-mass fuels around (4 0 0): (▲) position of the spinel peaks, (◆) position of the UO_2 peaks. In order to separate the backgrounds, two ordinates have been used.

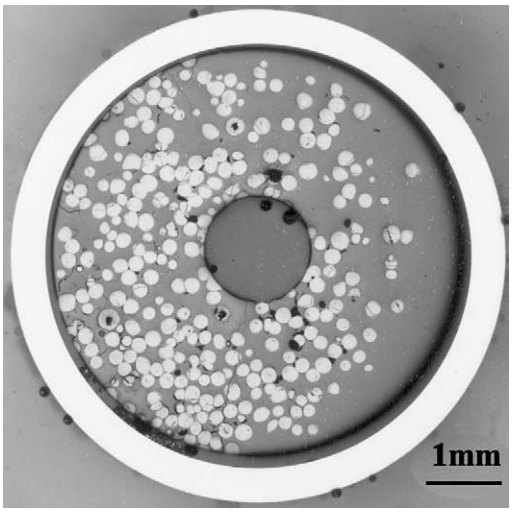

Fig. 6. THERMHET macro-mass structure cross section at the thermocouple's level.

This phase transition was probably even greater on the micro-dispersion structure at the end of the first cycle of irradiation, due to the much higher fission product implantation rate outside the UO_2.

3.3. Optical microscopy and EPMA

The metallographic examinations of the macro-mass and the jingle structures showed that these fuel microstructures were not much modified by the irradiation. Some cracks are the only modification detected. In spite of these cracks, the pellets could be removed easily from the pins without fragmentation and geometrical measurements could be performed. These geometrical measurements showed a very small decrease in the length of the pellets (<0.6%) whereas their diameter slightly increased (<0.15%).

The observations of the cracks in the pellets showed that the restructuring is greater in the macro-mass case (Figs. 6 and 7) than in that of the jingles (Figs. 8 and 9). Most of the macro-masses exhibit a fracture due to a differential thermal expansion between UO_2 and spinel. The cracks probably occurred during cooling, around the macro-masses or through it when they were not directly a result of the fabrication. The other cracks in the spinel led to a network between a few macro-masses. This should have an effect on the fission gas release at higher burnup. In the jingle case, the pre-existence of gaps around the jingles avoids most of the cracks through UO_2. Nevertheless some cracks developed in the spinel. Their path is modified by the jingle gaps and again, some networks between a few jingles appeared during irradiation.

No dissociation of the spinel was detected by EPMA, that is to say that even at the interface between the UO_2 and the spinel, the composition measured by EPMA is still that of the spinel.

Optical micro and macrographs of the micro-dispersion structure (Figs. 10–13) confirm that the fuel to clad gap is completely closed at all the examined levels. Almost no crack is visible in the peripheral part of the fuel and no sign of the previous pellet limits is detected on the axial cut. The centre of the pellets broke into pieces with great radial and transverse cracks stopping around 450 μm from the edge of the pellets. The breadth between the edges of the cracks is wider at the very centre than at the periphery of the cracked zone. Some azimuthal cracks are also completely separating fuel blocks, but others are found all around the fuel pieces. At the centre of the pellet, even in the previous central hole area, the corresponding shapes of the fuel pieces show that the parts were filling this free volume. A measurement of the fuel surface was conducted by image analysis. It is possible that a few fuel pieces were removed during sample preparation but the comparison of the radial and axial cut shows that this must be limited.

The fuel cross section surface at the top of the solid pellet stack, at the bottom of the hollow pellet stack measured on the axial cut and at the thermocouple level measured on the cross section are presented in Table 2, compared with the equivalent values before irradiation.

Furthermore, during the irradiation, most of the already very low fabrication porosity (1.7%) disappeared as it is lower than 0.2% in the bulk of the fuel.

One of the main results of this examination is that the fuel, at room temperature has a surface on the cuts, very close to what it was before irradiation. No major swelling is evidenced. Of course, this does not mean that there was no swelling at all due to some uncertainty in the image analysis and the possible loss of fuel

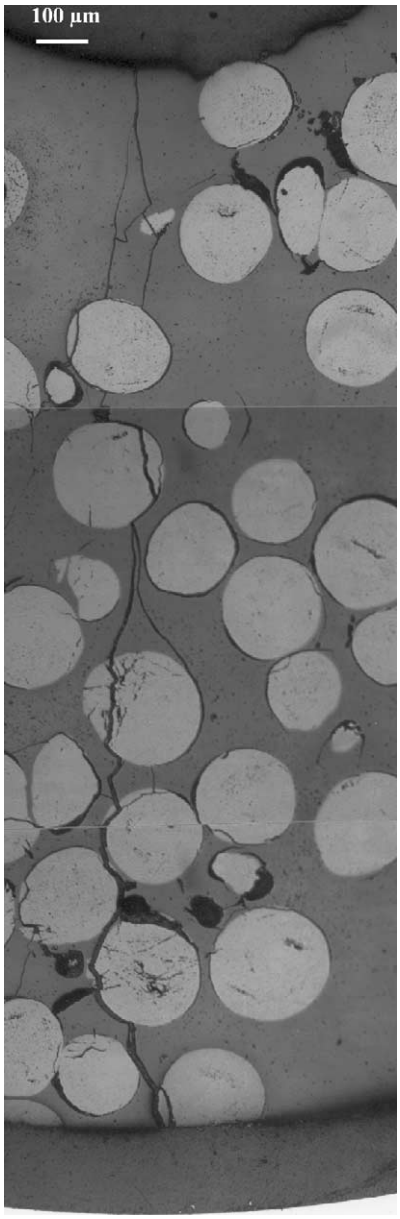

Fig. 7. THERMHET macro-mass structure, detail of the main crack observed on the cross section at the thermocouple's level.

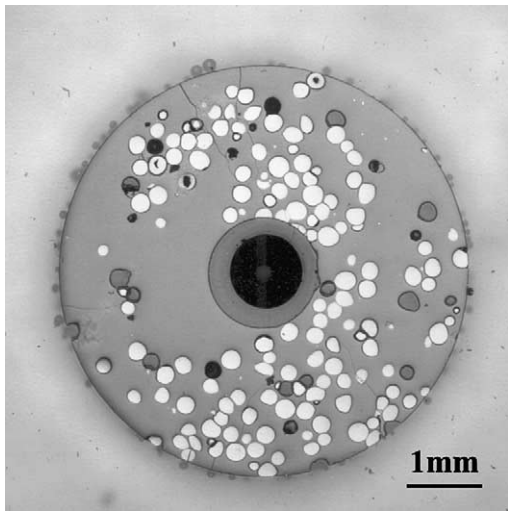

Fig. 8. THERMHET jingle structure cross section at the thermocouple's level.

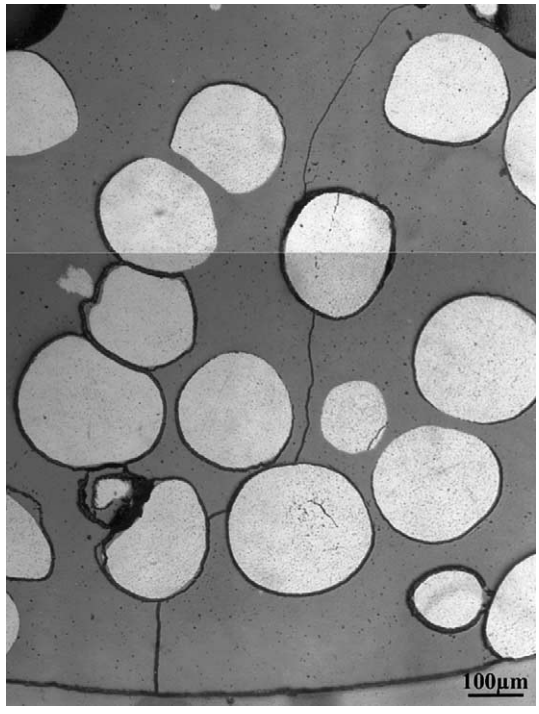

Fig. 9. THERMHET jingle structure detail of a crack on the cross section at the thermocouple's level.

pieces. Nevertheless, it shows that most of the volume expansion occurring during the irradiation vanished. We know from the observations of spinel irradiated with heavy ions that swelling occurs even without amorphisation [9,10].

The surface after irradiation at the top of the axial cut is very close to what it is at its bottom. This is consistent with the gamma scanning and the X-ray radiography observations and confirms that there was some movement of the fuel of the solid pellets towards the bottom of the hollow pellets. The lower value after irradiation at the thermocouple level is probably also a sign of axial departure from this level towards the bottom of the hollow pellet stack. This movement implies plastic deformation or creep. It may be promoted by the structure modification, major mechanical evolutions of

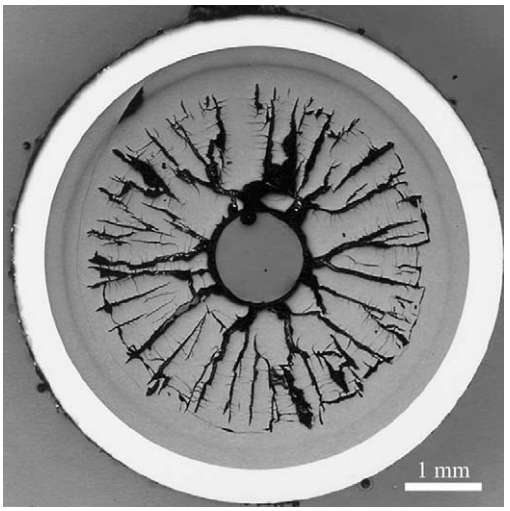

Fig. 10. THERMHET micro-dispersion structure cross section at the thermocouple's level.

the spinel having been evidenced after heavy ion irradiation [11] even before amorphisation.

Hence, the optical metallographs confirm that there was a great volume expansion in the fuel but nevertheless, this volume expansion was reversible and the final volume is very similar to that of the fresh fuel.

4. The MATINA pin 14 case

The MATINA pin 14 did not exhibit such swelling. In this irradiation up to the same burnup but with a higher temperature and a higher fast neutron flux, on the contrary, a slight decrease in the length of the fissile stack was detected. The pellets remained intact so that individual measurements were possible and showed a volume decrease in all the directions. The metallographs showed a probable disappearance of the slightest porosity.

This difference with the equivalent THERMHET pin behaviour, apparently due to the temperature difference, is consistent with what is observed during ion implantation in spinel [12,13]. In this case, the temperature seems to be one of the most sensitive parameters.

5. Interpretation of the temperature measurements and of the PIE for the micro-dispersion fuel

We know from the heavy ion irradiation results that the spinel may exhibit dramatic volume expansion with an order to disorder phase transition and eventually an amorphisation. We also know that high temperatures have an influence on this phenomenon during heavy ion irradiation. It can prevent it as in the MATINA case. At

Fig. 11. THERMHET jingle structure detail of the cracks on the cross section at the thermocouple's level.

last nano re-crystallisation may occur in the amorphous phase, under the TEM beams, as in [9].

The post-irradiation examinations showed that there was a major volume expansion during irradiation. That volume expansion is certainly due to amorphisation of

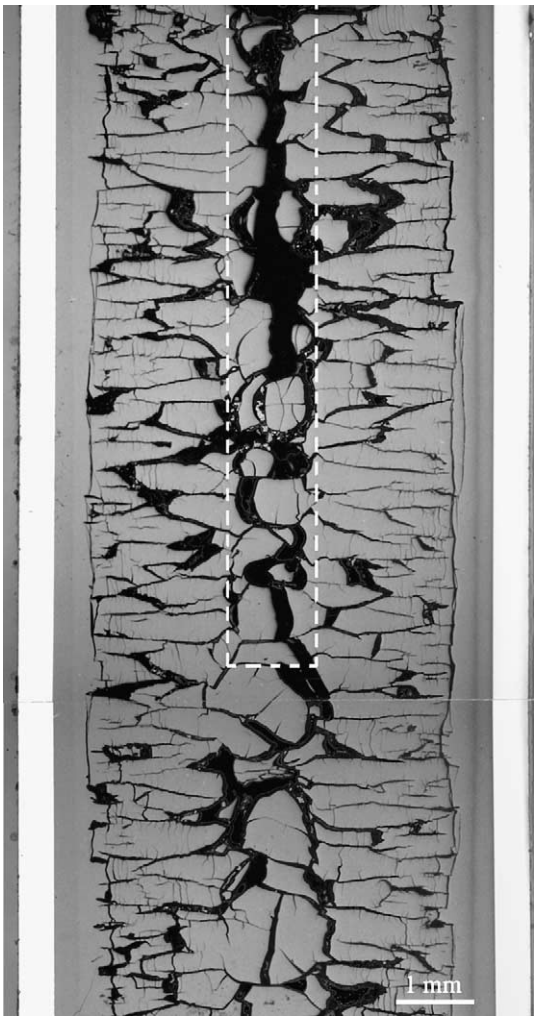

Fig. 12. THERMHET micro-dispersion axial cut at the position of the previous interface between the solid and the hollow pellets. The initial central hole limit is the white line.

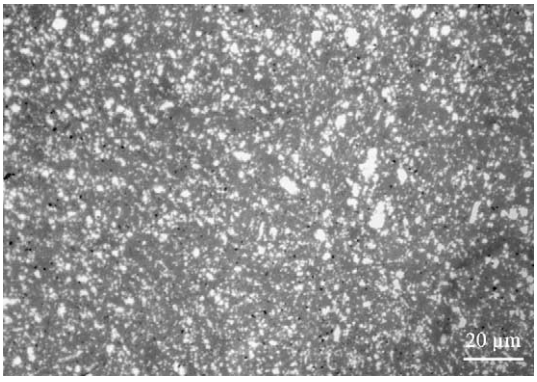

Fig. 13. THERMHET detail of the structure of the micro-dispersion after irradiation.

the spinel. It also showed that after this volume expansion there was an almost equivalent volume decrease. And finally, it showed that after the irradiation the spinel is mostly amorphous or nano-crystallised.

The temperature measurements exhibit two special periods: the first one at the beginning of the low power period during the second cycle is a decrease in temperature at a constant linear power, the second one during the last part of the second cycles is a decrease in temperature during a high power period.

The first interpretation, proposed in [3], explains this last temperature decrease as the effect of the amorphisation of the spinel, the volume increase leading to a gap closure. The temperature obtained at the very end of irradiation is then the result of the complete gap closure moderated by a significant decrease in the overall conductivity of the fuel due to the amorphisation of the spinel. If this is so, apart from the first temperature event that still must be explained, it implies that the volume reduction observed on the metallographs occurred during the cooling of the fuel. This volume reduction cannot be due to thermal expansion of the amorphous spinel, as it would imply a volume thermal expansion coefficient 9–10 times higher than the initial spinel one. Moreover, it would not be consistent with the implantation volume increases observed at room temperature. No known phenomena can explain this volume reduction at cooling.

The progressive decrease of temperature during the last part of the second cycle may be understood not as the progress of amorphisation, but on the contrary as recrystallisation by nucleation at high temperatures in the amorphous material created mainly at the beginning of the middle part of the second cycle of irradiation, at low temperatures. It would then be during this last high temperature period that the cracks would appear with volume reduction between the amorphous phase and the nano-crystalline material.

Consequently, we consider that the best description of this irradiation is:

- After a relatively high irradiation temperature during the first cycle and the first period of the second cycle, the irradiation at a lower temperature, in the middle of the second cycle, induces in about 14 h to a dramatic amorphisation. The volume expansion evidenced on the PIE occurs during this period. By the same time there is probably a huge thermal conductivity decrease. No real evaluation of this decrease was performed.
- The power increase at the end of the second cycle allows a nano-crystal structure formation by nucleation. This re-crystallisation induces a volume reduction. This volume reduction equivalent to the previous expansion is a radial movement of the central part towards the periphery, with crack formation. By the same time there is a new increase in

Table 2
Fuel cross section surfaces after irradiation obtained through image analysis of the axial cut and radial cut of the micro-dispersion structure, comparison with the initial state

	Top 4.5 mm solid pellets stack cross section fuel surface (mm^2)	Bottom 8.5 mm hollow pellets stack cross section fuel surface (mm^2)	Thermocouple's level cross section fuel surface (mm^2)
After irradiation	21.7	21.7	20.6
Before irradiation	23.0	21.6	21.6

the thermal conductivity limited by the cracks and also by the periphery of the fuel, where cracks do not appear and that may have remained amorphous because of the lower temperatures to which it was submitted.

6. Conclusion

A new evaluation of the PIE and of the irradiation data of the THERMHET pins was conducted. It showed that:

- An extra diffraction line is detected on the X-ray diffraction pattern of the macro-mass structure at a low burnup, in spite of an apparent absence of modification (pellet volume, micrographs, EPMA measurements). This peak is also observed in heavy ion irradiated spinel X-ray diffraction patterns and is the sign of a partial order to disorder phase transition in the spinel.
- There was a dramatic amorphisation of the spinel of the micro-dispersed pin, probably at the beginning of the low temperature irradiation period in the middle of the second cycle. A major volume expansion estimated around $\Delta V/V = 15\%$ occurred with this amorphisation together with a major decrease in the thermal conductivity.
- The increase of the temperature probably led to a nano-crystallisation of the amorphous phase of the micro-dispersion fuel with a volume reduction and a new augmentation of the thermal conductivity.

The comparison with the MATINA pin 14 irradiation shows that with higher temperatures during irradiation such phenomena do not occur. In the case of irradiation of macro-masses at low temperatures and higher burnups, the great volume changes during amorphisation around the macro-masses can also become a problem.

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