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# Size and radial origin of fragments formed while heating a 83 GWd/t<sub>U</sub> PWR fuel up to 1200 °C

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## Summary

When submitted to heating tests reaching 1200 °C, pellet long fuel samples generally exhibit cracking after the test, nonetheless, the fuel keeps its original shape and most of the fragments remain inside the cladding section [1].

However, after having been submitted to a 1200 °C, 20 °C/s, heating test with a surrounding pressure close to 0.1 MPa, in the Merarg LECA-STAR CEA device, a UO<sub>2</sub> section, with a local average burn-up of 83 GWd/t<sub>U</sub>, was found fragmented, with most of the fragments outside of the cladding and in the crucible used for the test. These fragments exhibited a wide range of sizes.

Detailed examinations on a sibling sample had been previously conducted. They are partly reported in [2].

Using these previous results as a reference for the fuel microstructure and composition along the pellet radius, a detailed examination of thirty three of these fragments was conducted using the EPMA. The purpose of these detailed examinations was to determine the radial origin of each one of these fragments and to evaluate whether there was, or not, a link between the size of these fragments and their initial location within the pellet.

The result of this study was that all the millimetric fragments visible on the polished surface of the sample came from areas that had shown, at the end of the irradiation, an especially low fission gas bubble formation rate, when compared to the rest of the pellet. On the contrary, very small fragments formed in all areas where many fission gas bubbles had been seen at the surface of a polished cross section.

## Introduction

Out-of-pile heating tests on irradiated fuels are being used at CEA to study of the release of fission products during LOCA type accidents. Within this, the "Gaspard" program, involving EDF and the CEA, was partly reported in [1] and [3]. It has been followed by a series of similar out-of-pile heating of irradiated fuels within EDF-AREVA-CEA collaboration. Some of these test results have been published in [4]. The samples used for these tests are pellet long sections of irradiated fuel inside the corresponding section of cladding, opened at both ends by the sample cutting. During these tests, where the temperatures reach 1200 °C, the fuel generally keeps its original shape and remains inside the cladding section. In order to better understand the origin of the released gases, post-test

examinations have been conducted on some of these samples. Optical microscopy and Scanning Electron Microscopy generally allowed to see that cracks had formed in the fuel but did not lead to any fragment relocation. On a very high burn-up  $\text{UO}_2$  PWR fuel, with a local average burn-up of  $83 \text{ GWd/t}_U$ , two tests have been conducted with and without cutting a longitudinal slit in the cladding, with the purpose to release the stresses induced by the cladding on the fuel itself. After a temperature ramp at  $0.2 \text{ }^\circ\text{C/s}$  up to  $1200 \text{ }^\circ\text{C}$ , the standard sample remained intact, with only a few small fragments missing at both ends, but the sample with the slit was found with all the fuel outside of the cladding with a wide range of fragment sizes. On the same fuel, a ramp up to  $1200 \text{ }^\circ\text{C}$ , but with a ramp rate of  $20 \text{ }^\circ\text{C/s}$  instead of  $0.2 \text{ }^\circ\text{C/s}$  lead to a similar result without any slit in the cladding. In that case, the fragments were embedded and polished for microanalyses. A detailed study, using EPMA, of thirty three fragments, allowed to know their radial origin before the test. For that, the EPMA results were compared to the detailed analyses that had been performed on a sibling sample. The methods used, the results obtained and their consequences are reported hereafter.

## Fuel Data

The fuel samples used for this study had:

- An initial  $^{235}\text{U}$  enrichment of 4.5%.
- Initial porosities:  $\approx 5\%$ .
- Grains sizes:  $\approx 11 \text{ }\mu\text{m}$ .
- Resintering tests lead to 0.4% of densification after one day at  $1700 \text{ }^\circ\text{C}$ , 0.61% after two days, and 0.7% after four days.

The rod was irradiated in the EDF Gravelines 5 reactor for seven one-year cycles. During the last three cycles, the average linear powers of the rod were around  $150 \text{ W/cm}$ . The mean rod burn-up was  $78.4 \text{ GWd/t}_U$ .

The fission gas release from the whole fuel stack was 7.3 % of the production.

All samples, the tested one and the references have been cut from the bottom part of the sixth span of the rod and the local section average burn-up was  $83 \text{ GWd/t}_U$ . Meteor and Alcyone fuel behaviour code calculations showed that the fuel centreline temperatures were in the range  $800\text{-}900 \text{ }^\circ\text{C}$  during the last 3 cycles.

## Experimental techniques used for this study

This work was performed in the LECA-STAR facility of CEA in the Cadarache research centre, in France.

The out of pile high temperature heating test was conducted using the Merarg device. It consists of a high-frequency ( $50 \text{ kHz}$ ) induction furnace, located in a hot cell and coupled to an on-line gas release measurement system. The fuel sample is generally located in a tungsten crucible. Between the crucible and the coil, a quartz tube is used to collect all the released gases. A sweep gas at  $\sim 0.1 \text{ MPa}$  carries the released gases out of the hot cell so they can be analysed by gamma spectrometry of  $^{85}\text{Kr}$  in a counting chamber located in a glove box behind the cell. The sweep gas used in the test was Ar.

After the test, the fuel was embedded in a Sn-Bi metallic alloy. This alloy allows examinations using charged beams, such as that of EPMA, SEM and SIMS (Electron Probe Micro-Analyser, Scanning Electron Microscope and Secondary Ion Mass Spectrometer) without having to coat the surface with

a conductive layer, as it is necessary when using standard epoxy, the fuel electrical conductivity being high enough to avoid local charging problems.

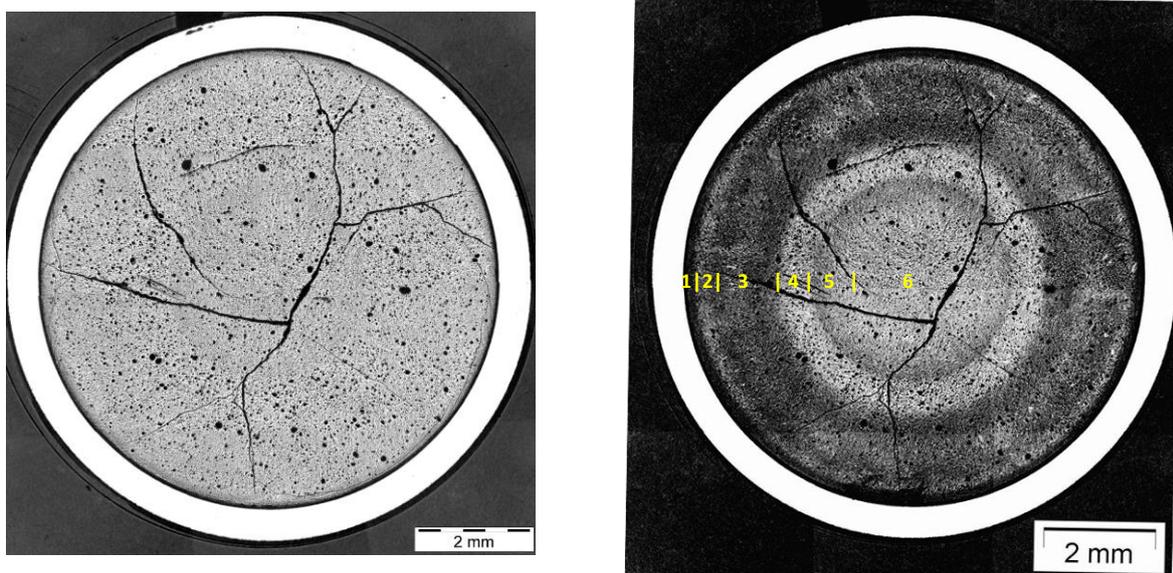
Most of the post-test characterisations as well as the reference characterisations were conducted using a CAMECA shielded EPMA SX 100R.

### **Macrograph and EPMA Post Irradiation Examination of the sibling samples used as references**

Cross sections of this fuel examined by optical macroscopy and by EPMA, situated close to the sample tested, were used as references for identifying the origin of the fragments formed. In Figure 1, the macrograph after chemical etching reveals the presence of at least six concentric areas that we numbered starting with the rim area. In this figure, the dark areas correspond to places where bubble precipitation is high. The very centre of the cut (area 6) is relatively bright though. This corresponds to a lower density of larger bubbles, when compared with area 5, for example.

In Figure 2, these six concentric areas can be seen on the Xe measurement profile. This profile is being compared with the Nd radial profile, with selected relative scales chosen so that this Nd profile draws a good estimation of the local Xe creation. In spite of the 7.3 % fission gas release rate in the rod, most of the missing Xe, in the EPMA measurements corresponds actually to gas precipitated in bubbles large enough to induce a significant loss in the EPMA measurement. The important Xe variations along the radius have been used, in this study, for the determination of the fragments origin. Nd and Pu concentrations radial profiles have also been used with that purpose, but they only exhibit a significant gradient along the  $\sim 1000 \mu\text{m}$  on the periphery, so that they cannot be used to make the difference between central fragments.

Figure 3 corresponds to the last part of the EPMA examination that has been used in this study. Electronic images of the polished surface, Xe maps and Mo maps give a good signature of the various parts of this fuel and therefore a good complement to identify the origin of a fragment.



**Figure 1 : Macrograph, as polished and after chemical etching, of a 83 Gwd/t<sub>0</sub> cross section of the UO<sub>2</sub> fuel, naming of the six concentric areas visible on the cut.**

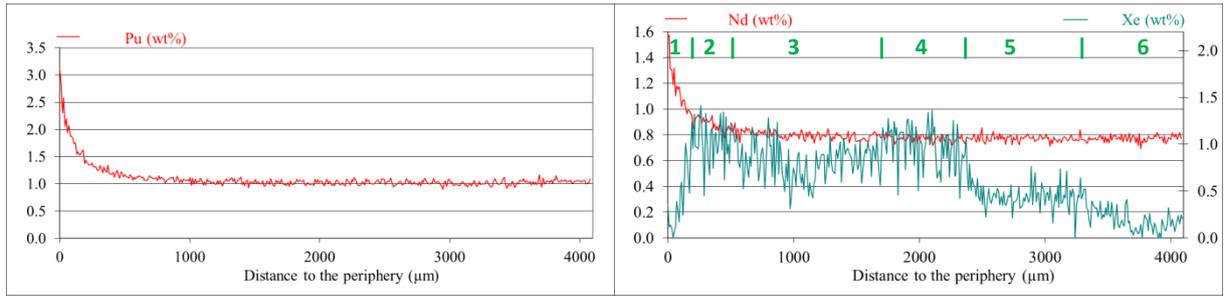


Figure 2 : EPMA radial profiles of Pu, Nd and Xe.

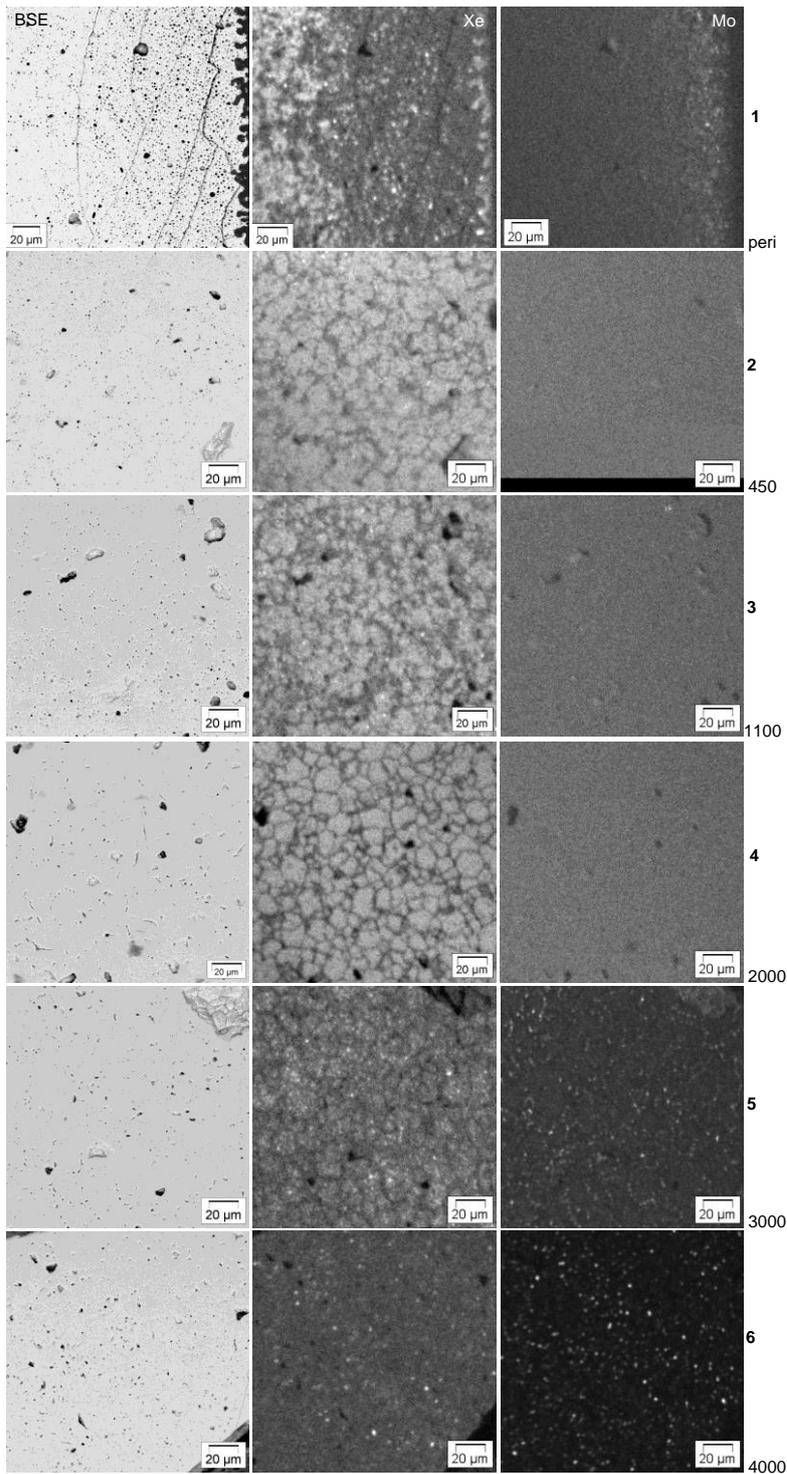


Figure 3 : EPMA, electronic image, Xe and Mo maps, examples in each concentric area.

## The sample prepared for micro-analyses

As stated in the introduction, after taking a pellet long section of this fuel, with its original irradiated cladding cut at both ends, up to 1200 °C, with a ramp rate of 20 °C/s, with an argon circulation around 0.1 MPa, a large part of the fuel was found, in the crucible, but outside the cladding.

During this test, 24.7 % of the fission gas produced at this level was released.

Figure 4 shows a macrograph of the sample prepared for microanalyses. Most of the surface is the Sn-Bi embedding alloy. The cladding on the left side of the image is the cladding of the irradiated sample studied here. Some fuel, still bounded to it, is visible. The piece of cladding on the right side is another cladding sample (without fuel pellets inside) not studied here. The numbers on the image on the right correspond either to the number given to a fragment, or to the number given to an area from which several fragments have been examined. These images clearly show that a wide range of fragment size was found, some of the fragments being even too small to be visible with this low magnification. Thirty three fragments have been examined, in particular, all the large ones.

Figure 5 shows two examples of images along the inner surface of the cladding. It shows that, at the polished level, fuel was still bonded with the cladding, though the HBS area is highly cracked. It may not have been the case all along the length of the cladding.

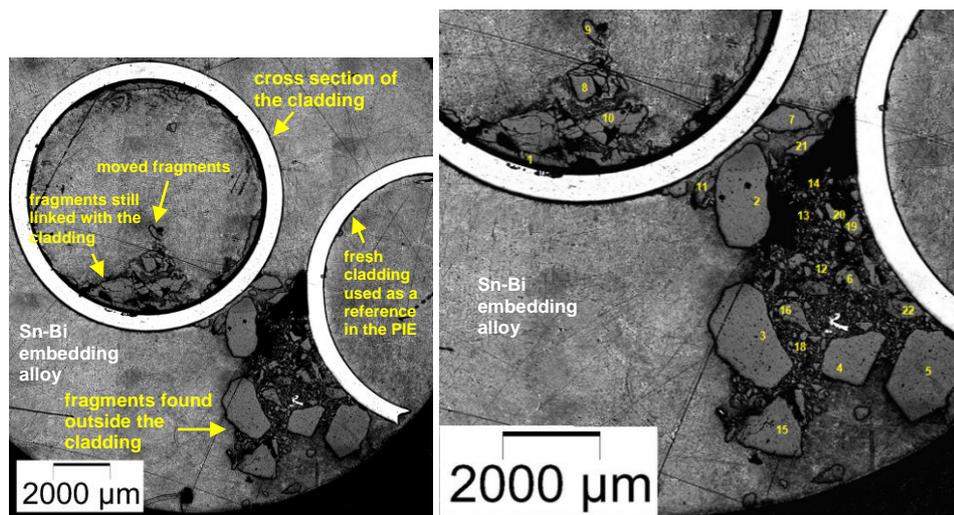


Figure 4 : Macrograph of the sample prepared for micro-analyses, naming of the fragments examined or sample areas in which several samples were examined.

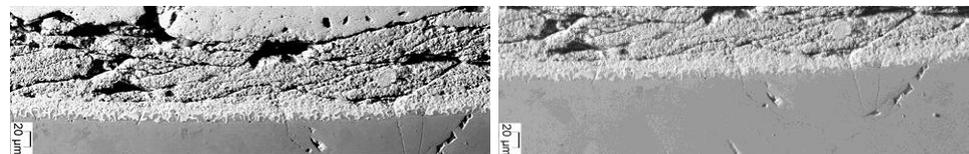
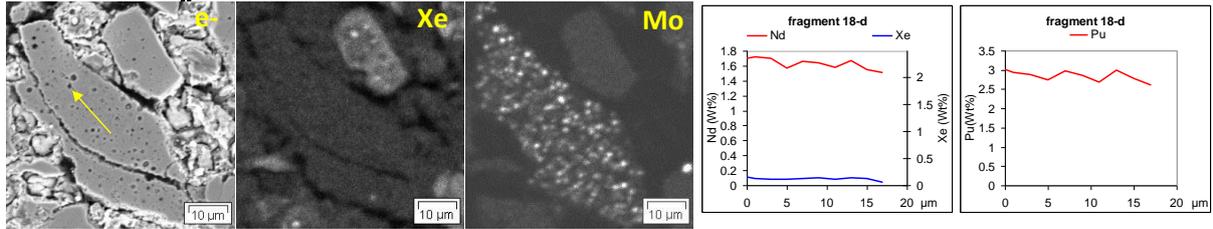


Figure 5 : SEM images along the inner surface of the cladding. Presence of cracked HBS.

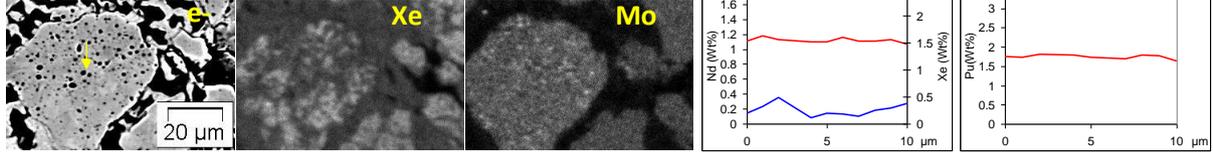
## Detailed EPMA fragment examinations

This chapter gathers, in Figure 6 and Figure 7, examples of the detailed examinations that have been conducted in order to evaluate, with some confidence, the radial origin of the fragments. For each fragment, these figures show an electronic image, a Xe map, a Mo map and quantitative measurements of Nd, Xe and Pu along a line. The location of the line within the fragment is given by an arrow drawn on the electronic image. A global view of the large fragment is also given, a square locating the detail elementary maps.

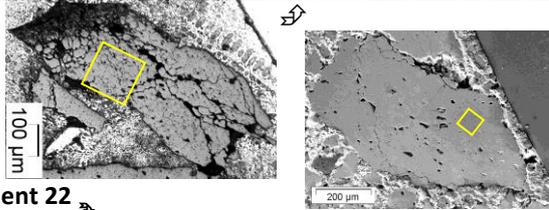
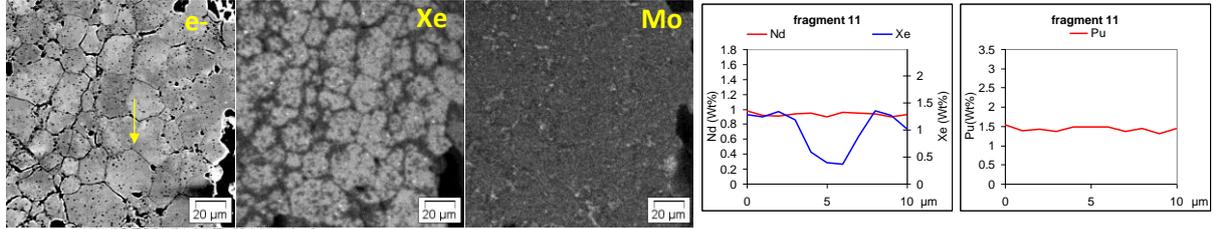
**Fragment 18-d Area 1, HBS fragment, extreme periphery**



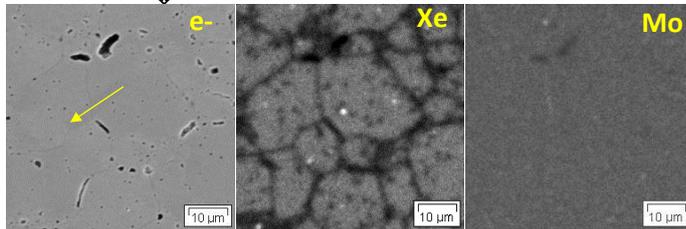
**Fragment 13-3 Area 1, ~96 μm from periphery**



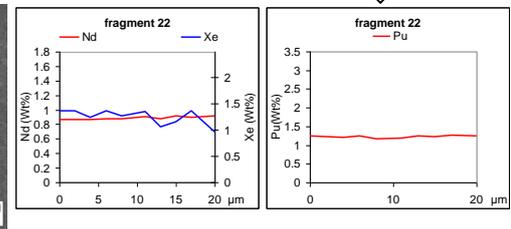
**Fragment 11 Area 2, ~200 μm from periphery**



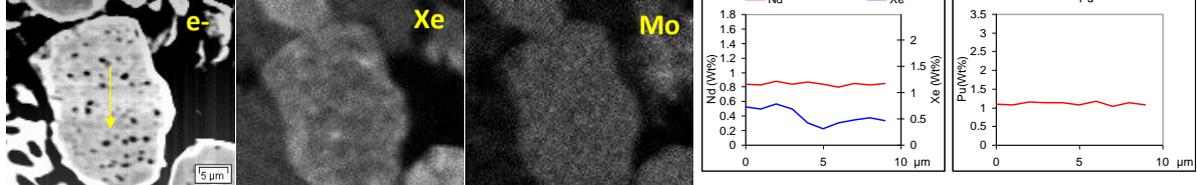
**Fragment 22**



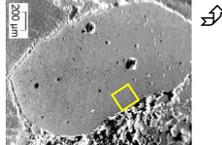
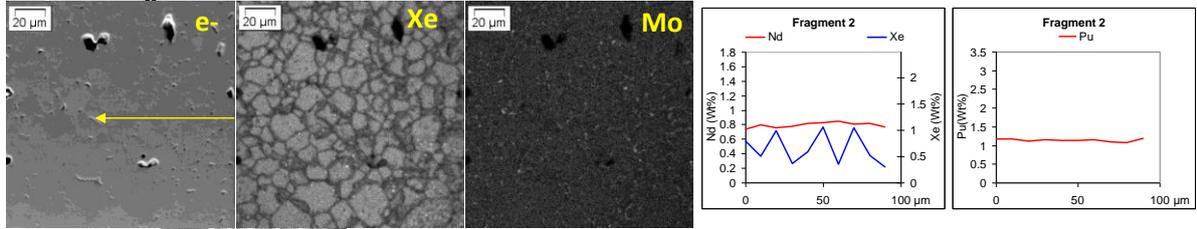
**Area 2, ~350 μm from periphery**



**Fragment 14-4 Area 3, ~700 μm from periphery**



**Fragment 2 Area 4**



**Figure 6 : Examples of EPMA fragment examinations, e- image, Xe and Mo map, Nd+Xe and Pu analyses, periphery.**

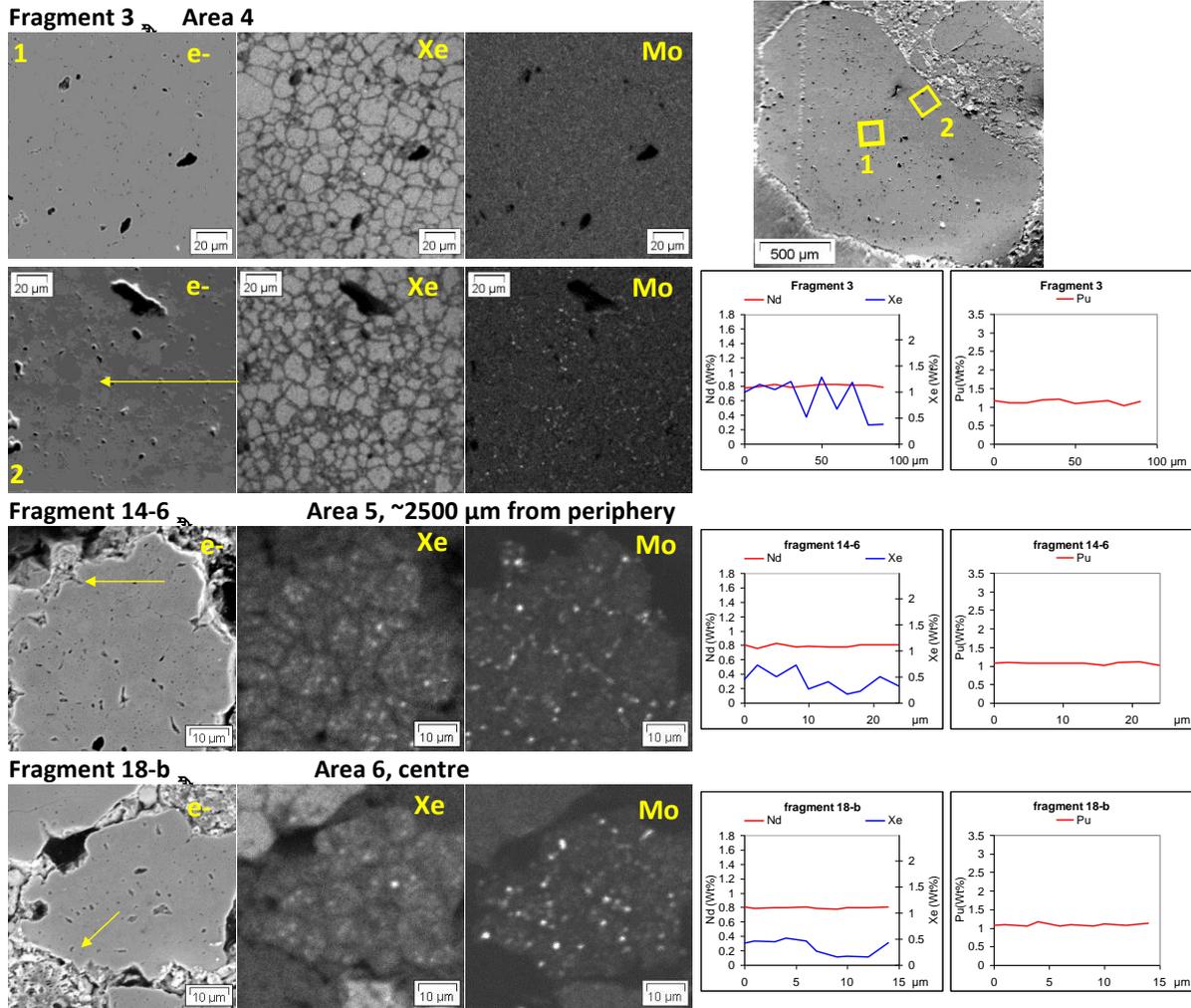


Figure 7 : Examples of EPMA fragment examinations, e- image, Xe and Mo map, Nd+Xe and Pu analyses, centre.

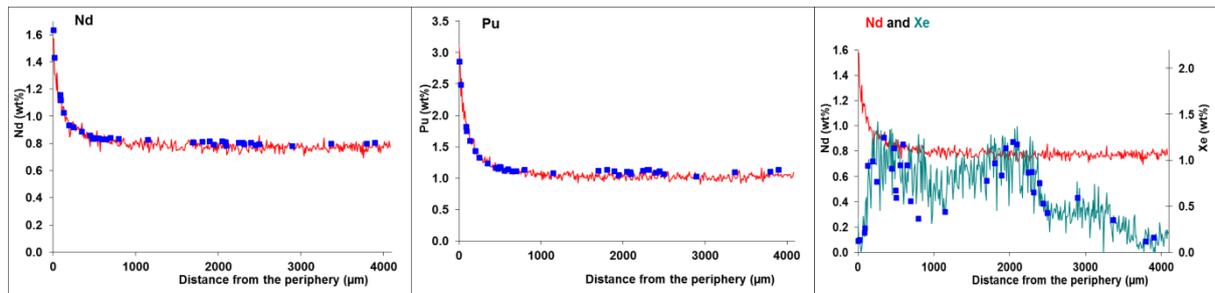
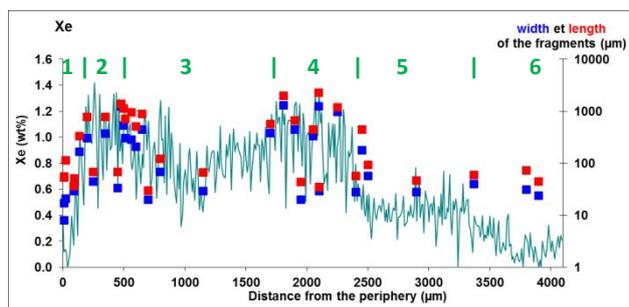


Figure 8 : Average Nd, Pu and Xe quantitative analyses along the detail lines in each fragment, as a function of the estimated radial origin of the fragment. Comparison with the quantitative measurements in the reference sample

The electronic images give the bubble distribution. The Xe maps show characteristic concentration distribution that change with the radial position. The Mo maps show the metallic fission product sizes, densities and distribution relatively to the grain boundaries, also characteristic of the initial radial position. The quantitative analyses of Pu and Nd give a precise information on the distance to the periphery in the first 1000  $\mu\text{m}$  and for the lowest values, the indication that the fragment comes from beyond that point. On the periphery, the gradients observed in the Nd and Pu profiles also give some information on the initial orientation of the fragment. Xe quantitative analyses are mainly used through the average measurement along the line, but the variations are a good complement to the Xe maps.

Using all this information together, and comparing it to the equivalent measurements on the sibling sample, it was possible to establish the initial location of the thirty three fragments. Figure 8 shows the average Nd, Pu and Xe quantitative analyses along the detail lines in each fragment, as a function of the estimated radial origin of the fragment, compared with the quantitative measurements in the reference sibling sample.

The apparent sizes of the fragments have been measured, extracting the maximum and minimum length measured, named here the "width" and "length" of the fragment. Figure 9 gives these widths and lengths as a function of the initial location. It appears that all the large fragments came from area 2 and 4 where gas bubble formation was the lowest, and EPMA Xe measurement close to local production.



**Figure 9 : Width and Length of each fragment, as they appear on the polished surface, as a function of the estimated radial origin of the fragment.**

## Conclusion

The fragments formed during a 1200 °C, 20 °C/s heating test, with a surrounding Ar pressure around 0.1 MPa, on a 83 GWd/t<sub>U</sub> fuel, have been examined with EPMA. Using the initial EPMA examination of a sibling sample, as-irradiated, the radial origin of the fragments were determined. These fragments exhibited a wide range of sizes. It was shown that all the large, millimetric, fragments came from areas where, before the test, gas precipitation into large bubbles was the lowest.

Other observations have been made on fuels submitted to temperature transients in out of pile heating tests [1], LOCA type tests or in-pile LOCA tests [5], where, in sections where the fuel did not move much during the test, the highest densities of cracks were observed in particular areas. Moreover ref. [6] showed that the smallest fragments formed after a ~70 GWd/tU out of pile LOCA type test were not only coming from the rim of the pellets.

This new result shows that the parts of very high burn-up fuels that are prone to fragmentation in case of high temperature transients, with no high pressure surrounding, are the parts where a high density of large bubbles had formed, under irradiation, prior to the transient.

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