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## Plutonium multi recycling: self-irradiation effects on the structural properties of future (U,Pu)O<sub>2-x</sub> nuclear fuels

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### **I. Objectives**

In France, the UO<sub>2</sub> spent nuclear fuel is reprocessed in order to recover plutonium, which is used to fabricate (U,Pu)O<sub>2</sub> fresh MOx fuels. With the emerging concept of Pu multi-recycling (i.e., recycling Pu from spent MOx fuels), the future MOx fuels will be enriched in Pu isotopes having higher specific activities (such as <sup>238</sup>Pu and <sup>241</sup>Pu) than the main Pu isotope (<sup>239</sup>Pu). Due to the high alpha activity of plutonium (and <sup>241</sup>Am) isotopes, the MOx fuel matrix suffers from alpha self-irradiation (SI), as the two energetic particles (recoil nucleus and He atom) formed by an alpha decay generate defects in the crystal structure. Thus, these future MOx fuels will be subjected to stronger SI effects compared to current ones. At the macroscopic scale, this phenomenon results in lattice swelling due to the damage cascades and/or the accumulation of He atoms over time. Although this swelling and its eventual saturation are well described in the literature, the identification and description of the associated mechanisms such as point defects creation, their stability and consequences on the microstructure are yet to be revealed. The purpose of this thesis is thus to acquire detailed information about the defect formation and their accumulation in the MOx microstructure as a result of the SI damage through investigation of (U,Pu)O<sub>2</sub> samples that have accumulated different levels of SI dose in their microstructure. For a complete description of the defects, we combine several characterization techniques with different probe sizes such as Raman spectroscopy, XRD (X-ray diffraction), EPMA (Electron probe micro analyzer) and XAS (X-ray absorption spectroscopy).

### **II. Experimental**

The methodology is based on starting with an old enough MOx sample that already reached saturation ( $\Delta a/a_0=0.29\%$ ) in terms of lattice parameter increase due to the SI effect. A cut is taken from this old sample and submitted to a thermal treatment, whose conditions (8 hours at 1473 K under Ar/4.3% $H_2$ /1200ppm  $H_2O$ ) ensure a complete defect healing without modification of the sample oxygen stoichiometry. The assumed “defect-free” and defect-saturated samples can thus be compared.

The selected MIMAS type MOx sample (manufactured in 2007) with an average Pu/(U+Pu) content of 10 wt%, allowed us to study three specimens with different doses of SI damage: 1 month after thermal treatment (sample A), 15 months after (sample B) and the historical old sample (sample C). In each sample, three phases are identified using EPMA imaging and Raman microscopy, see Fig 1. They correspond to UO<sub>2</sub> agglomerates containing almost no Pu, Pu-rich

agglomerates with a Pu/(U+Pu) content close to 30 wt%, and a third “pseudo-phase” located between the two others’ with an intermediate, but non constant, Pu content.

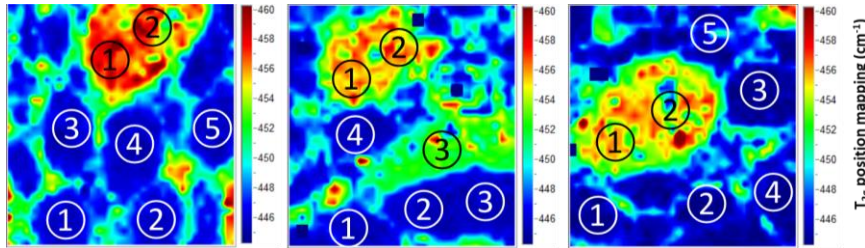


Figure 1 Raman images of the  $T_{2g}$  band position of a) Sample A, b) Sample B, c) Sample C. The size of the images is  $70 \times 70 \mu\text{m}^2$ . The blue zones are considered as the U-rich agglomerates, while the yellow-red zones are the Pu-rich agglomerates.

### III. Results

XRD pattern of the annealed sample is taken periodically and refined to obtain a spatially averaged information and the swelling of the lattice parameter in the three phases. For a rather more local scale information ( $\mu\text{m}$ ), the Raman microscopy is used in the imaging mode. Fig 1. corresponds to the  $T_{2g}$  Raman band position mapping of the selected zones for each of the three samples. As the  $T_{2g}$  represents the symmetric stretching of the O-O bonds, its frequency is highly sensitive to the change in cell parameter. This helps us to locate the U-rich and the Pu-rich zones whose average Raman spectra are further compared. The average spectra collected in these zones allowed to obtain their representative Raman spectra, presented in Fig.2.

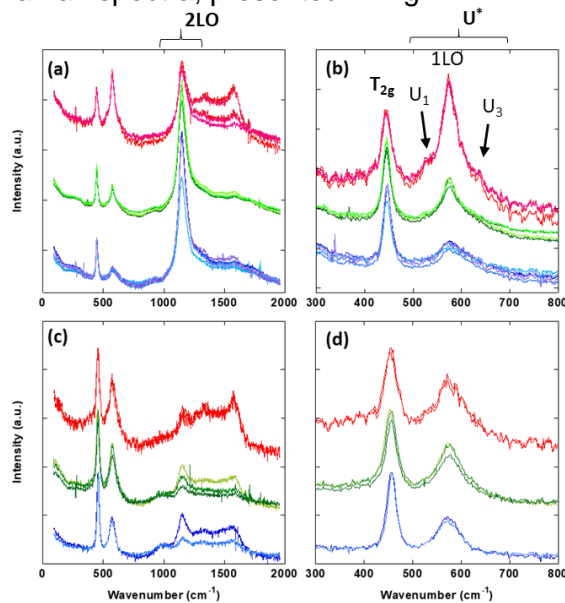


Figure 2 Raman spectra for Pu- (a and b) and U- rich (c and d) agglomerates in the samples. The blue, green and red lines respectively correspond to samples A, B and C. (a) and (c): complete spectra (b) and (d): close-up of the first order region.

The individual characteristics (width, position and intensity) of the different Raman bands ( $T_{2g}$ ,  $U^*$  and  $2LO$ ) can be compared in detail. With increasing SI dose, the  $T_{2g}$  band shows a negative shift, an increase in FWHM and an asymmetry in both U-rich and Pu-rich areas. In the U-rich zones, the gradual decrease in the  $2LO$  intensity, and increase in  $U^*$  intensity gives indirect information about the SI defects. The  $U_1$  and  $U_3$  defect bands observed in damaged U-rich zones gives another indication of structural defects due to SI. The methodology employed in this study thus proved to be adapted to characterize self-irradiation-induced structural defects in MOX. Finally, to have a complete description of the defect nature, the RS outcomes will be coupled with  $\mu$ -XAS results that were collected in the same regions as those analyzed by Raman microscopy.