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Microstructural investigation of (U,Pu)O_{2-x} with the Pu content and O/M ratio by Raman spectroscopy and imaging

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The best candidate for the nuclear Sodium-cooled Fast Reactor fuel is the uranium-plutonium mixed dioxide (U,Pu)O_{2-x}. The plutonium content is planned to range between 19% to 40% mol. The fuel has to match various physico-chemical specifications: to be chemically homogeneous, oxygen hypostoichiometric (Oxygen/Metal ratio, with M=(U+Pu), between 1.94 and 2.00)... However, for a Pu content higher than 20%, and an O/M ratio lower than 1.98, at room temperature, the phase diagram of U-Pu-O system exhibits a miscibility gap domain [1]. The sample has then two phases of same structure but different O/M ratios. The demixtion of these two phases triggers the creation of cracks into the material. This phenomenon has to be avoided in order to keep a homogeneous microstructure. The behavior of the microstructure has then to be fully investigated.

Recently Talip *et al* [2] and Elorrieta *et al* [3] evidenced that Raman spectroscopy is a promising tool for characterizing (U,Pu)O₂ materials. Coupled with a microscope, this technique can be used to determine several physico-chemical properties at the grain scale. For instance, the cation distribution homogeneity, the grain size, and the crystal defects can be studied.

UO₂, PuO₂ and (U,Pu)O₂ all have the same Fm-3m structure. As the Raman active modes are determined by the symmetry, all these materials have similar Raman spectra. The main band is the T_{2g}, which corresponds to the asymmetrical stretching of the O-O bonds. The position of this band gives information on the chemical composition: O/M ratio, Pu content... Moreover, by studying sintered UO₂ samples by Raman cartographies, Maslova *et al* [4] have evidenced that the T_{2g} intensity is related to the grain orientation. By drawing the T_{2g} intensity map, the grains has been then revealed.

In the present work, this technique has been extended to (U,Pu)O_{2,00} samples. Like in the literature [4], the sample microstructure has been exhibited by performing T_{2g} intensity mapping. Furthermore, the variation in the T_{2g} position, related to the Pu content, has been studied and some heterogeneities in the Pu content were observed at the micrometric scale with the T_{2g} position map.

As this characterization tool was first developed for samples with O/M = 2.00, this method can be now extended to oxygen hypostoichiometric samples. The O/M ratio is fixed by heating the sample under reducing atmosphere (Ar/H₂ 5%). A dedicated device has been specifically designed for *in situ* Raman measurements [5]. The variation in the microstructure according both the Pu content and the O/M ratio values will be then studied.

[1] Sari *et al.* *J. Nucl. Materials*, 35, 267-77 1970

[2] Z. Talip *et al.* *J. Raman Spectrosc.*, 48 (5) 765–772, 2017

[3] J. M. Elorrieta *et al.*, *J. Nucl. Mater.*, 495, 484–491, 2017.

[4] O. A. Maslova *et al.*, *Mater. Charact.*, 129, 260–269, 2017.

[5] G. Montagnac *et al.*, *Carbon*, 54, 68–75, 2013