	Experiment title: Grain Orientation and Full Strain/Stress Tensors Mapping in He Implanted UO_2 Nuclear Fuel Using µ-Laue Diffraction	**Experiment number**: MA-452
Beamline: BM32/IF	**Date of experiment**: from:18/02/2008 to: 23/02/2008	**Date of report**: 25/09/2008
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Report:

This work is part of the PRECCI project (CEA, EDF) dedicated to the study of nuclear fuel (UO_2) microstructure evolutions in conditions of long term and interim storage. In that framework, one of the main issue is related to the behaviour of the significant quantity of the helium produced by the actinides alpha decay in the UO_2 pellets. The main aim of this work was to derive mechanical data from He implanted UO_2 polycristalline disks.

After anneling of He implanted UO_2 polycristalline disks (9 µm in diameter), it has been recently shown by nuclear reaction analysis at the micrometer scale (µ-NRA) that [1]:

- grain boundaries act as short cuts for He diffusion,
- He diffusion is furthermore significantly enhanced in the vicinity of grain boundaries from a temperature of 900°C.

The main aim of this work was to characterize the full strain/stress tensors in the UO_2 in the centre of the grain and at its periphery. In that view, 2D mapping using micro-focused Laue X-ray diffraction measurements were required since this technique enables the localization of grain boundaries and also the determination of the strain/stress tensor.

The samples of interest were UO_2 sintered polycristalline disks, the average grain size being around 9 µm. He (500 keV) implantations and annealings were performed at the CEMHTI laboratory. Three kinds of samples have been analyzed during this experiment: (1) non-implanted (fresh) samples (mono and polycristals), (2) as He implanted samples, (3) He implanted and subsequently annealed samples. Based on µ-NRA results, two kinds of annealing conditions were selected:

- in the first no He depleted zone had been observed at grain boundaries (typically 1 to 4 hours at 800°C),
- in the second thick depleted zone were evidenced: this is the case for temperatures higher than 900°C and short durations.

The influence of sample preparation on the stress/strain in these three kinds of samples has also been evaluated. 12 samples have thus been characterized. They were conditioned under kapton tape for safety reasons.

The conventional set-up of BM32 set-up for µ- focused X-ray diffraction in Laue mode has been used. The size of the pink X-ray beam has been reduced down to about 1×2 µm² (H×V). To be more sensitive to the He implanted volume in the sample which is located at 1.0±0.5 µm under the sample surface, the energy range of the X-ray pink beam has been restricted: at the beginning the natural range was used (5-28 keV) and successive limitations were performed first at 16 keV and then at 13 keV. This energy range reduction does not affect significantly the accuracy of the calculated mechanical data: the number of Bragg peaks measured on the CCD camera remains large enough for good statistic. On the UO_2 single crystal used as reference, 21

Bragg peaks have been found with the 5-13keV energy range (cf. Fig.1). At 13keV, X-ray penetration depth into UO_2 was about 7 µm (I_R/I_0=10%).

A- B- C-

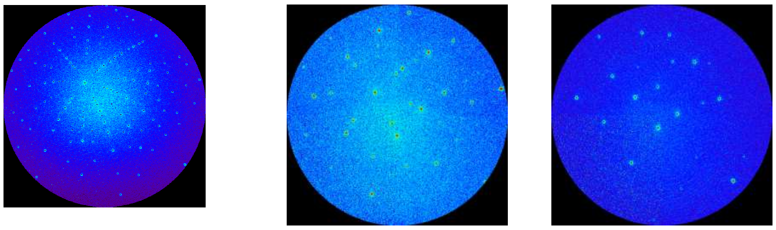

Fig. 1: Influence of the energy range of the X-ray beam on the number of peaks measured on the same UO_2 single crystal.
- A- 5-28keV (129 peaks found),
- B- 5-16keV (43 peaks found),
- C- 5-13keV (21 peaks found).

On each of the 12 polycristalline samples, a 58×30 µm² µ-XRD mapping (H×V) has been carried out using this last energy range (5-13keV). XRD Laue images were analyzed with the Xmas software package. If only the most intense Bragg peaks are considered, the automatic indexation procedure leads to grain orientation maps in excellent agreement with optical micrographs (cf. Fig. 2). However, it must stressed that despite the band pass reduction of the X-ray beam energy, a very large amount of Bragg peaks can be found on all images collected on polycristalline samples: about 200 peaks are identified that correspond to an unexpected high number of grains (10 grains defined by more than 10 Bragg peaks). It is very difficult to interpret those results by just comparing the measured size of the X-ray beam (2×1µm²), the crystallite size (6 to 9 µm diameter) and the X-ray penetration depth into the matrix (7µm). Most probable hypothesis at this step is that the conventional method for defining the X-ray beam size which is based on the full width at half maximum, is not relevant when dealing with the characterization of a matrix containing heavy scatterers like U (Z=92e^-). In this case, the X-ray beam appears to be larger.

During this expriment, 2D µ-XRD mapping has only been performed using a pink X-ray beam; for this reason only the deviatoric part of the strain tensor could be determined. Preliminary analysis of these data shows that whatever the sample preparation methodology, only as-implanted samples present a streaking of Bragg peaks (espacially those measured at high 2θ angles) (cf. Fig.3-A). This streaking is directed towards the center of the camera. For annealed samples this streaking has not been observed demonstrating that only elastic deformations remain. To illustrate the strong deformations in UO_2 caused by He implantation, it must be added that average von Misès stress in the as implanted samples is close to 1000 MPa whereas this value reaches only 100-200 MPa in fresh polycristalline UO_2 disks. This stress in the as-implanted samples has been obtained by taking into account only the Bragg peak part marked with yellow squares (cf. Fig 3-A).

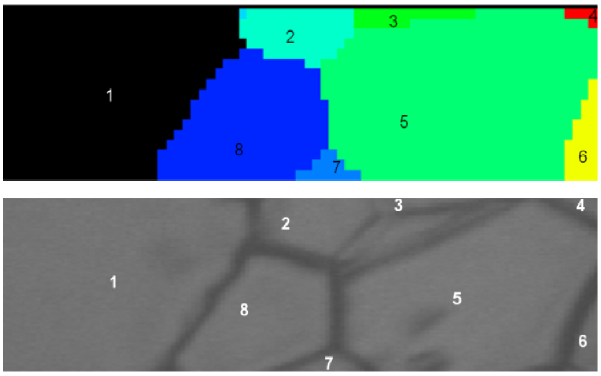

Fig. 2: Grain orientation map compared with an optical micrograph of the same UO_2 polycristalline sample (He implantation followed by a 1 hour annealing at 1000°C). Numbers written on the grains help for comparison.

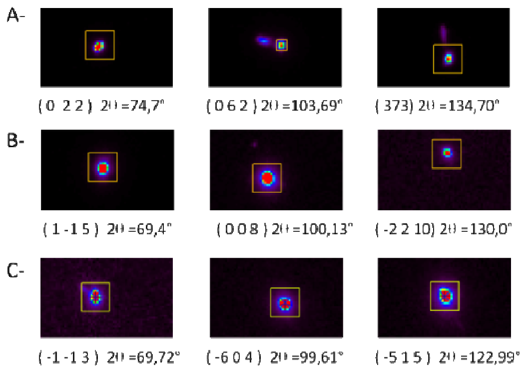

Fig. 3: Comparison of the shape of the Bragg peaks with their 2θ angle of occurrence for three He implanted UO_2 polycristalline samples. (as implanted (A-), 1 hour annealing at 800°C (B-) and at 1000°C (C-) respectevely after He implantation).

Conclusions:

In this first experiment, the interest of Laue diffraction using micro-focused X-ray beams for strain/stress study in polycristalline He implanted UO_2 has been demonstrated. Specificities of the UO_2 matrix have been overcome. Further calculations are ongoing to characterize quantitatevely the stress gradients along the grains after the different annealing treatments to establish definetly whether this variation may be correalated with the He depletion. However it is now clear that these measurements will have to be combined with µ-XRD using monochromatic X-ray beams to adress this issue.

References:

[1] G.Martin et al, NIM B249, (2006) 509-512.