ESRF	**Experiment title:** Irradiation effects on rare gas behaviour in UO_2	**Experiment number**: 30-02-714
Beamline: BM30B	**Date of experiment**: from: 7 September 2005 to: 13 September 2005	**Date of report**:
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REPORT:

As described in the our proposal, the experimental result presented here are part of a much wider modelling and experimental program aimed at improving our understanding and modelling of the in-pile behaviour of UO_2[1]. The project is essentially geared towards deriving fundamental properties relative to the behaviour of rare gas atoms (one of the most abundant class of fission products) in UO_2 fuels. The program is based on the study of ion implanted polycrystalline UO_2 samples because it is one way of circumventing the many scientific and technical difficulties inherent to the study of in-pile irradiated UO_2. The use of ion implantation techniques also enables experimentalists to focus on a particular physical or chemical effect, whereas in-pile irradiated materials or fission experiments always involve a myriad of coupled phenomena. **The first aim of the experiment was to the study the interaction of energetic ions with fission gases.**

Results obtained with our previous experiment 30 02 658 [1] on beam-line BM30B have given us an insight into the effect of annealing temperature on the pressure inside xenon and krypton inclusions. The experiments also provided information as to the physical state of the gas inside the inclusions and further revealed the far greater sensitivity of XAS to Kr rather than Xe. It has thus been proved that experiments can be performed on Kr implanted samples for which Kr concentrations are closer to those encountered in irradiated fuels**. In the second part of the measurements, thermal behaviour of Kr aggregates in UO_2 was studied.**

Experimental

The samples are depleted UO_2 pellets, 8 mm in diameter and approximately 1 mm thick implanted with 400 keV Kr ions. Concerning the first part of measurements we used implantation conditions leading to a Kr local concentration of 2 at.%. Samples were submitted to 10^{14} 800 MeV $Kr.cm^{-2}$ and/or annealed at 600°C for 18 min. Concerning the second part, in order to examine the effect of Kr concentrations on bubble precipitation 2 doses were used leading to local Kr concentration of 0.8 and 0.4 at%. Samples were submitted to thermal treatment at temperature ranging between 600 and 1500°C. Measurements were realized at 12K at the krypton K edge.

During our experiment we observed a major problem: diffraction peaks due to the highly crystalline UO_2 matrix. To resume during each scan, intense Bragg peaks were observed in every fluorescence channel of the 30 elements detector. Clearly extracting EXAFS spectra was impossible whatever the analyzed sample. Moreover, resulting XANES spectra suffer from a poor signal to noise ratio: for this reason, the characterisation of samples with 0.4 at.% could not be performed.

Results

As in 8 at.% sample presented in the previous experimental report [1] we obtained on UO_2 sample doped with 2 at.% without any thermal treatment a XANES spectrum with a strong white line and a resonance located at ~15 eV (*Cf* Figure 1). Such a spectrum indicates that Kr atoms have precipitated into pressurized bubbles [2]. Irradiation with 10^{14} krypton ions at 800 MeV induced a modification on rare gas aggregates as we can observed on Figure 1. But EXAFS characterization is needed to interpret such evolution. As observed in Figure 2, thermal annealing of 600°C 20 minutes and an irradiation give the same XANES spectra. Thus a short annealing at 600°C and a heavy high energy ion irradiation leads to the same result on Kr rare gas aggregates. Furthermore, annealing of irradiated sample induces no variation.
The initial implanted Kr atoms are located on the first 300 nm from the surface, the energy loss of 800 MeV Kr ions in this area is only by electronic interaction. Thus, such interaction induced modification on rare gas aggregates. This could be an experimental evidence of the re-solution of rare gas atoms induced by interaction of fission products.

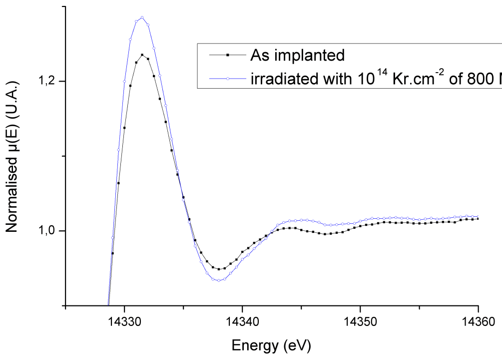

Figure 1: XANES spectra collected at 11K

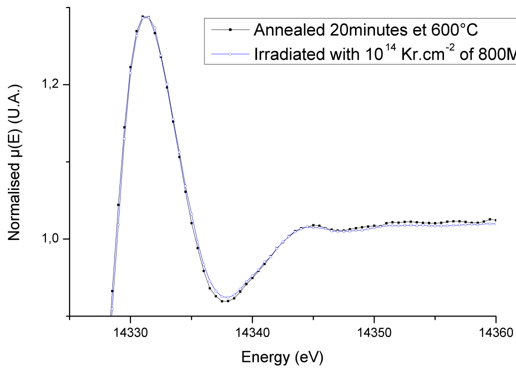

Figure 2: XANES spectra collected at 11K

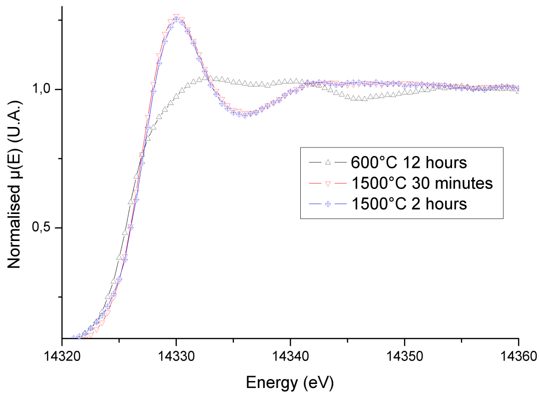

Figure 3: XANES spectra collected at 11 K on samples implanted with 0.8 at% of Kr.

As observed in Figure 3, contrary to 8 at.% sample[1], no evidence of Kr aggregates is present after annealing at 600°C. Thermal treatments above 1400°C and 1500°C induce precipitation but accurate information on temperature of precipitation is still lacking. Furthermore, since no difference between XANES spectra collected after annealing at 1500°C 0.5 hours and 2 hours is observed, study of the pressure evolution at high temperature could not be performed.
On top of instrumental development to ride of Bragg peaks, the next step of this project will be the determination of the influence of krypton concentration on bubble precipitation. We will also try to study thermal condition needed to induce precipitation.

References
[1] P. Martin et al, ESRF BM30B experimental report 30-02-658 (2004).
[2] A. Di Cicco *et al*, Phys. Rev. B 54(1996)9086-9098.