ESRF	**Experiment title:** **µ-XRD study of the interaction layer between UMo alloys and aluminum**	**Experiment number**: **ME-1285**
Beamline: ID18f/ID22	**Date of experiment**: from: 05 December 2005 to: 10 December 2005	**Date of report**: 15/04/2006
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Report

A worldwide program supports the use of Low Enriched Uranium (LEU, $^{235}U<20$ at.%) in fuels for research reactors such as the future Réacteur Jules Horowitz (RJH) in Cadarache or the FRM2 in Munich (Germany). But due to the decrease in ^{235}U enrichment for the conversion to LEU, new nuclear fuels have to be designed. The current choice for RJH fuel is uranium-molybdenum, in γ-phase, with 8 weight% Mo content (18 at.% UMo8). The fuel, which consists of UMo8 spherical particles (20-30 µm radius) surrounded by an Al alloy matrix, is encased in between two aluminium cover sheets. Post-irradiation examinations of UMo8 [1] demonstrated its strong potentialities as fuel but they also point out its great reactivity with aluminium. This reactivity leads to the formation of an interaction layer between fuel and matrix that is suspected to cause the strong fuel plate swelling and an unacceptable mechanical behaviour. The main aim of these XRD studies was to determine using a micro-focused beam (µ-XRD) the crystallographic composition of this interaction layer UMo/Al.

12 shifts have been allocated to this project on **ID18f beamline**.

The diffraction measurements were performed at 16keV in reflexion and/or transmission mode with a bidemensional MAR130 detector. The size of the X-ray beam was usually 4.1*3µm². Semi-quantitative phase analysis has been carried out with the FullProf software package [3] after conversion with the Fit2d software of the pictures into conventional 1D patterns [4].

Two kinds of samples have been analysed during this session.

In the first set of specimen (three fuel plates and on two diffusion couples(cf. figure 1)), the interaction layer UMo/Al has been grown by thermal annealing. These studies were needed to:

- understand the metallurgy of the U, Mo, Al system and to precise previous analyses carried out with large X-ray beams available on laboratory diffractometers (see for example [2]),
- determine the phase composition of the intercation layer when limited concentration of other elements (in particular silicon) were added to the alumium alloy.

The second set of analysed samples consists of fuel plates irradiated by heavy ions. This methodology enables to simulate the fission fragment damages and so to grow an interaction layer under non-equilibrium conditions (closer to the in-pile one). Very recently, our group (collaboration between CEA, FRM2 and CERCA) succeeded in the irradiation of UMo nuclear fuel plates using a 80MeV iodine beam at the Maier Leibnitz laboratory (Garching, Germany): an interaction layer surrounded each γ-UMo7 particles after heavy ion irradiation (cf. figure 2) [3]. Five irradiated and non-irradiated specimens were characterised by µ-XRD during this session.

Two examples of results are detailled in the following, when the synthesis of these analyses is still ongoing.

1. Study of the γ-UMo7/Al diffusion couple prepared by thermal annealing at 600°C 10 hours:

A large area on this sample (15*700µm²) has been characterised in reflection mode with a fixed X-ray beam incidence angle. The size of the X-ray beam remain limited at the sample surface (15*3µm²). So the probed part ranges from the aluminium side up to the UMo alloy part and so includes the totality of the UMo/Al interaction layer (cf. figure 1-a). First results of the semi-quantitative analysis of the XRD patterns show that at least three regions must be considered in the interaction layer (cf. figures 1-b and 1-c):

- in the first (≈40µm thick), the ternary UMo_2Al_{20} and binary UAl_4 phases have been identified with fluctuating concentrations. Note that in few first microns of the UMo-Al interaction layer some peaks remain non-attributed but they come very probably from impurities present in the aluminium alloy before the thermal treatment,
- in the second (≈70µm thick), the binary UMo_2Al_{20} is still present but associated with the UAl_3 phase,
- in the last (≈300µm thick), the phases $U_6Mo_4Al_{43}$ and UAl_3 have been found with constant relative proportions.

This µ-XRD study confirm our previous µ-EXAFS measurements at the Mo-K edge on this sample [5]: ternary phases ($U_6Mo_4Al_{43}$ and UMo_2Al_{20}) have been identified on each part of the interaction layer with a non negligeable concentration (more than 20%wt).

2. Characterisation of the interaction layer grown by a I-127 beam on a UMo7 dispersed nuclear fuel:

Considering the limited I-127 penetration depth into the UMo particles (a few micrometers), µ-XRD measurements were performed in reflexion mode with a low (15°) and fixed angle of incidence. The size of X-ray beam at the sample surface was 15*3µm². The comparison between XRD patterns collected on fresh and irradiated fuels reveals the unique presence of the UAl_3 binary phase in this UMo-Al interaction layer (cf. figure 2) underlining the differences with fuel plates after thermal annealing. However using this technique, it was thus impossible to determine the location of the molybdenum elements inside the UMo-Al interaction layer. The allocation of beamtine on BM-30B to study these irradiated samples by EXAFS at the Mo K edge should enable us to elucidate this problem [5].

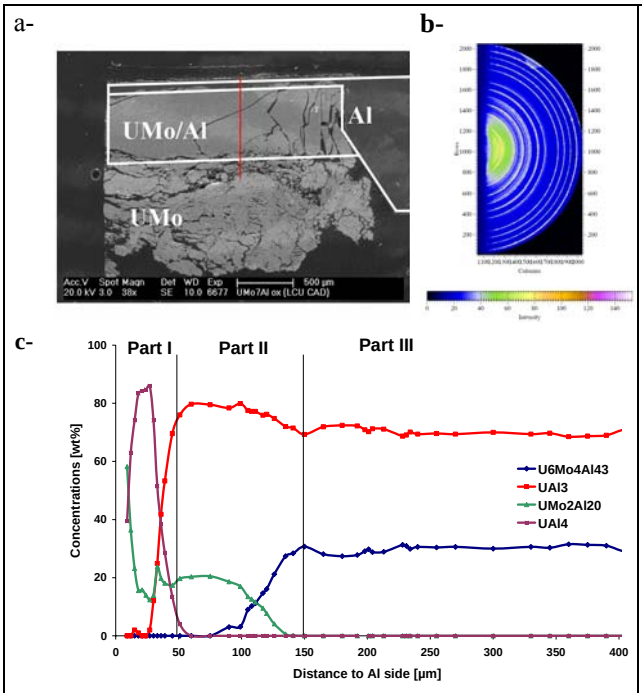

Figure 1:
Morphology (a-) and crystallographic composition (b- and c-) of the γUMo7-Al interaction layer obtained by SEM and µ-XRD respectively. Red line on the diffusion couple indicated the area probed by µ-XRD.

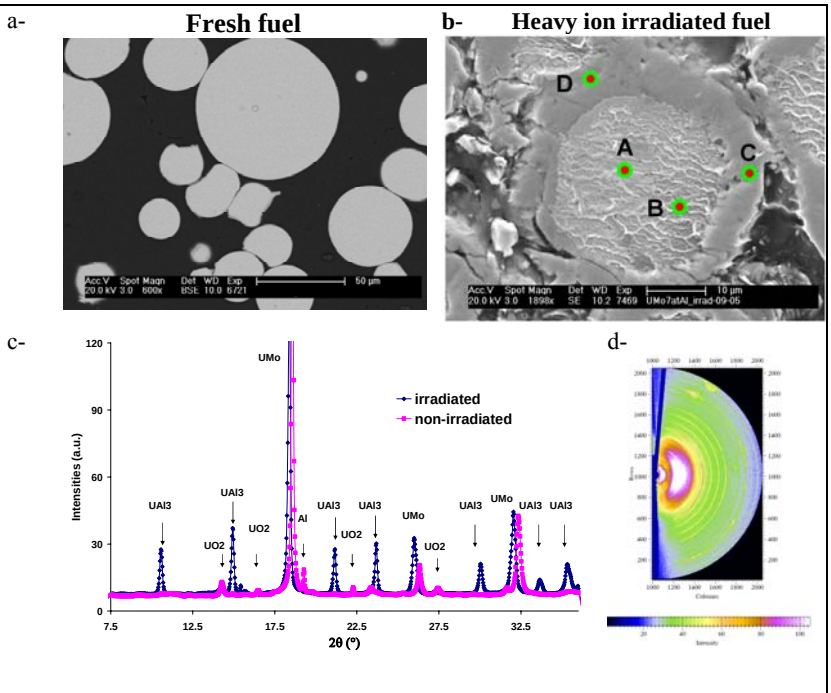

Figure 2:
Influence of heavy ion irradiation on a dispersed UMo7 nuclear fuel studied by SEM (a- and b-) and µ-XRD (c- and d- on irradiated fuel). Energy dispersive analyses confirm the presence of U, Mo, Al elements in spots C and D when only U and Mo were identified in A and B (cf. fig b).

References

[1] M. K. Meyer *et al.*, *Journal of Nuclear Materials* (2002) 304, 221-236.
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