ESRF	**Experiment title:** Study of the UMo/AlSi interaction layer by combining μ-XRD, μ-XRF and μ-XAS	**Experiment number**: MA-651
Beamline: ID22	**Date of experiment**: from: 03/11/2008 to: 07/11/2008	**Date of report**: 31/08/2009
Shifts: 9	**Local contact(s)**: Dr. R. Tucoulou	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

H. Palancher[a]*, E. Welcomme[a]*, J. Allenou[a]*, X. Iltis [a]*

(a) CEA, DEN, DEC, F-13108 Saint Paul lez Durance Cedex- France

Report:

Within the framework of the development of low enriched nuclear fuels for research reactors, U-Mo/Al is the most promising option that has however to be optimised. Indeed at the U-Mo/Al interfaces between U-Mo particles and the Al matrix, an interaction layer grows under irradiation inducing an unacceptable fuel swelling. Adding silicon in limited content into the Al matrix has clearly improved the in-pile fuel behaviour. This breakthrough is attributed to a silicon rich diffusion layer (SrDL) around U-Mo particles grown during fuel manufacturing (hot rolling at about 400-500°C). In this work, the evolution of the microstructure and composition of this SrDL with increasing Si concentrations in the Al matrix has been investigated. The goal of this proposal was to combine three analytical techniques at the micrometer scale (μ-XRD with μ-XRF and μ-XANES at the Mo K edge): on top the crystallographic composition, the Mo behavior could be characterized in this protective layer (SrDL).

<u>Experimental methods</u>:

With that goal, the energy of the incoming X-ray beam had to be set at 28keV. However the beamline staff did not succeed in focusing the X-ray beam down to a micrometer size. For that reason, the experiment was performed at 17keV with a 5×2 μm^2 incoming X-ray beam. This has however made impossible μ-XRF and μ-XANES at the Mo K edge measurements: only μ-XRD data have been collected during this beam-time. Moreover, electronic problems with the CCD FRELON camera have been encountered. **Together, we estimate at 2 shifts (over 9) the beamtime lost. A new proposal will be submitted to perform this characterization.**

Two types of samples have been studied: eight U-Mo7/Al-Si diffusion couples and 2 particles extracted from an as fabricated U-Mo7/Al-Si fuel plate. For the diffusion couples, two sets of four diffusion couples have been characterized:

- U-Mo7/Al-Si2, U-Mo7/Al-Si5, U-Mo7/Al-Si7, U-Mo7/Al-Si10 annealed at 450°C during 2 hours,
- The same samples that underwent an additional thermal treatment at 350°C during 2 months.

On these samples, the μ-XRD measurements were performed in reflection mode. The X-ray beam size on the samples was about 20×2 μm^2. The length of the beam print was positioned parallel to the reaction front and the sample was moved by micrometer step, perpendicularly to this front, between each acquisition.

For the fuel particles, measurements have been carried out in transmission mode. Diffraction data were collected with a 2D FRELON camera and analyzed using the previously detailed methodology [1].

Results:

In this part, results of the characterizations on diffusion couples and fuel plates are successively presented.

In diffusion couples, two types of SrDL have been evidenced depending on the Si content in the Al-Si alloy: the threshold value is found at about 5 wt% but obviously evolves with diffusion couples annealing temperature. It has been shown that for Si concentrations ranging from 2 to 10 wt%, the SrDL is bi-layered and the Si-rich part is located close to the Al-Si for low Si concentrations (below 5 wt%) and close to the U-Mo for higher Si concentrations. For Si weight fraction in the Al alloy lower than 5 wt%, the Si-richest sub-layer (close to Al-Si) consists of $U(Al,Si)_3$ + UMo_2Al_{20}, when the other sub-layer (close to U-Mo) is silicon free and made of UAl_3 and $U_6Mo_4Al_{43}$. For Si weight concentrations above 5 wt%, the Si-rich part becomes $U_3(Si,Al)_5$ + $U(Al,Si)_3$ (close to U-Mo) and the other sub-layer (close to Al-Si) consists of $U(Al,Si)_3$+UMo_2Al_{20}. Figures 1 and 2 present the compositions of the SrDL in the U-Mo7/Al-Si2, U-Mo7/Al-Si7 diffusion couples respectively. Except a phase transformation in the UMo7 alloy (destabilization of the γ phase to form α-U), the 2 months thermal annealing at 350°C did not modify the composition of the diffusion couples, showing the stability of the SrDLs. On the basis of these results and of a literature survey, a general scheme is proposed to explain the formation of different types of SrDLs U-Mo and Al-Si alloys. **Main results of this work have been submitted for publication in Journal of Nuclear Materials in June 2009**.

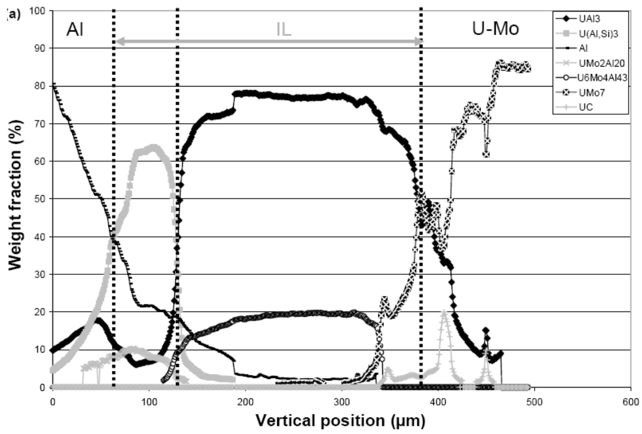

Figure 1: Major phases distribution throughout the SrDL: result of the µ-XRD analysis of an U-Mo7/Al-Si2 diffusion couple.

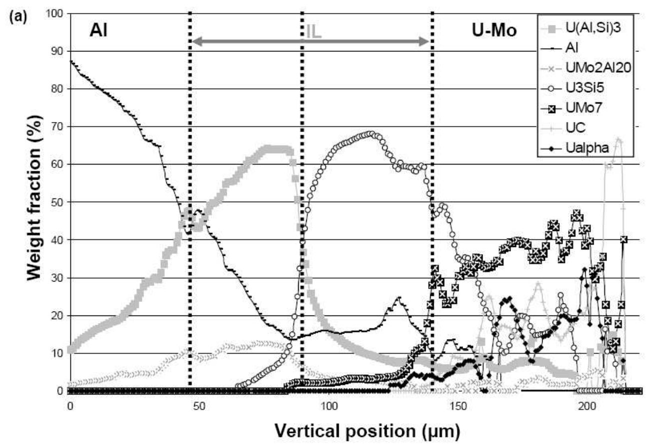

Figure 2: Major phases distribution throughout the SrDL : result of the µ-XRD analysis of an U-Mo7/Al-Si7 diffusion couple.

The µ-XRD characterization of the U-Mo7 particle taken from a full-sized IRIS3 fuel plate (U-Mo7/Al-Si2) has shown the presence of $U(Al, Si)_3$ and $U_3(Si, Al)_5$ i.e. the SrDL composition in "real U-Mo7 samples" differs from that foreseen on the basis of a diffusion couple study. Indeed in U-Mo7/Al-Si2 diffusion couples, the phases $U(Al, Si)_3$ and UMo_2Al_{20} and or $U_6Mo_4Al_{43}$ have been found.

However since the availability of fuel plates manufactured with different Si concentrations and different annealing temperatures is limited, research activities on diffusion couples must go on and the discrepancies induced by the geometry of the samples corrected afterwards.

References:

[1] H. Palancher et al., *J. Appl. Cryst.* (2007) 40, 1064-1075 ; H. Palancher et al., *J. Nucl. Mater.* (2009) 385, 449-455.