EXPERIMENTAL REPORT
RAPPORT D'EXPERIENCE

Programme Committee Proposal Number
N° Projet Comité de Programme

02-01-60

PROJECT TITLE : *TITRE DU PROJET :*

Densification mechanisms in UO_2 and $(U,Ce)O_{2+x}$

LIGNE : **D2AM**

INSTRUMENT : PETITS ANGLES

NUMBER OF RUNS USED
NOMBRE DE SESSIONS EFFECTUEES : 12

STARTING DATE
DATE DE DEMARRAGE : 05 may 99

AUTHORS : *AUTEURS :M. RIPERT, A. BOULORE, P. GARCIA, L. DESGRANGES, P. GOEURIOT*

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The dimensional stability of $(U,Pu)O_2$ fuel pellets under irradiation is of crucial importance for power reactors. This stability is the consequence of a competition between fission product induced fuel swelling and the disappearance of as manufactured porosity under irradiation [1]. The study of this phenomenon for uranium dioxide has resulted in a modelling which brings in the size distribution of initial porosity [2-5] and which generally assumes a rapid disappearance of the small size porosity. The experimental characterisation of this submicronic porosity is therefore a first step towards improving fuel modelling.
Small angle X-rays scattering (SAXS) allows one to obtain structural information such as dimension, pressure and density in very small bubbles (below 0.2 microns in diameter) [6].

The main aim of this study was to apply this technique to nuclear fuels (i. e. for safety reasons, $(U,Ce)O_{2+x}$, a non radioactive analogue of $(U,Pu)O_{2+x}$), and characterise the pore size distribution. We thus used the 12 shifts allocated by the Review Committee to perform the SAXS measurements on different samples :
* a UO_2 reference pellet
* 4 $(U,Ce)O_{2+x}$ pellets (with Ce concentrations of 5, 10, 25 and 50 %)
Same samples with 2 different thermal treatments (10 and 24 hours at 1700°C under controlled atmosphere) were also analysed to determine the submicronic porosity evolution during annealing.

This experiment required experimental conditions [7] which had not been used before. The local contact, J.P. Simon, has had to involve himself fully in the experiment and has been working full time with the Cadarache team :
- the density of UO_2 together with the difficulty to prepare thin slabs of this brittle material impose to work at the highest photon energy possible, underneath the U L3 edge (17168eV). The transmission of

a 0.1mm thick sample is about 0.005 at 17000eV. Secondly, due to the wide range of expected porosity, two configurations where chosen, with a sample-detector distance of 2165mm and 660mm respectively. The first one allows us to detect down to 0.0025 angström^{-1}, but has too weak a signal above 0,015 angström^{-1} to be able to make a significant subtraction of the dark noise of the CCD camera. The second one gives an overlap with the previous scattering range and leads to a significant signal up to 0.05 angström^{-1}.

- the use of the anomalous scattering at such a high energy. We had the good surprise that there is no variation of the counting efficiency within 2% between 15463eV and 17155eV. This was checked by using an AlZnMg alloy in an aged hardening stage (with precipitation of Guinier-Preston zones).
- the methodology of data reduction and of data analysis was developed by J.P. Simon and checked on the reference sample :

* The scattering pattern only presents a q^{-4} power law, the signature of abrupt interfaces. Since it extends down to the lowest measured q values, most of the signal comes from the porosity in this sample elaborated during the sintering stage (fig.1).
* Anomalous data recorded at four energies 17100, 17130, 17145 and 17155eV have the same pattern shape. The relative intensities are constant within only +/-10%, due to the too low transmission. The calculated relative variation of the intensity is of -8.5% from 17100eV to 17155eV assuming pores, i.e. with a contrast equal to the square of the mean electronic density (fig.2).

The analysis of the other samples is in progress at Cadarache.

Figure 1

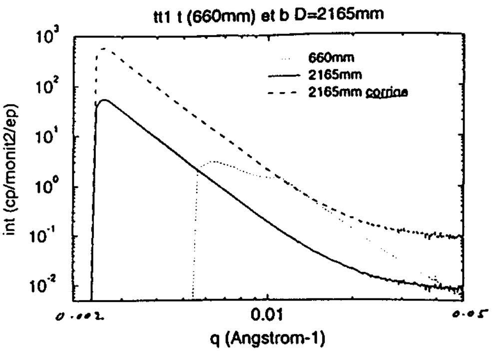
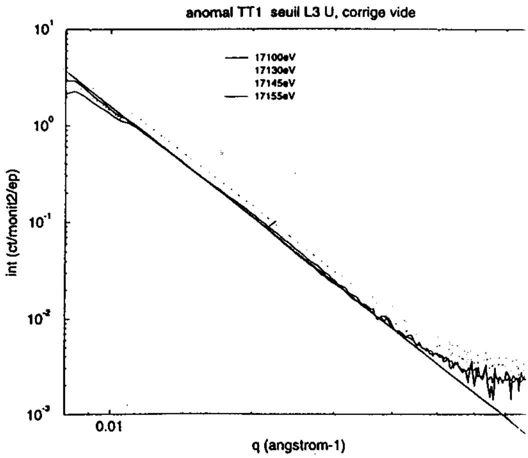


Figure 2



References:

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