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Review

Study of the γ-U(Mo) phase decomposition for atomised and ground powders

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ABSTRACT

U(Mo) particles are obtained by centrifugal atomisation or by mechanical grinding fabrication processes and display microstructures which strongly influence the gas retention efficiency and consequently the behaviour of the fuel. Moreover, during fuel plate fabrication, a partial decomposition of the γ-U(Mo) phase toward the α-U and U_2Mo phases can occur and is supposed to influence also the irradiation behaviour of the fuel. In situ X-ray and neutron diffraction experiments have been performed in order to study the γ-U(Mo) phase decomposition for two types of powders produced by the atomisation and the grinding routes. The present study allows assessing differences between these powders, in terms of γ-U(Mo) phase decomposition kinetics and mechanisms.

1. Introduction

The necessity for Material Testing Reactors (MTRs) to develop a Low-Enriched Uranium (LEU) fuel has led to the development of a dense fissile material based on γ-U(Mo) alloys (Glaser et al., 2005). The designed fuel is a composite material, made of dispersed γ-U(Mo) particles embedded in an Al-matrix. This "meat" is then covered by two sheets of aluminium alloy through a hot rolling process to form the final product in a plate shape. In-pile experiments show that the performances of such a fuel are limited under high irradiation conditions. During fuel plate fabrication, a partial decomposition of the γ-U(Mo) phase toward the α-U and U_2Mo phases can occur and is supposed to influence the irradiation behaviour of the fuel (Gan et al., 2012). In fact, depending on the fabrication process of the particles, by centrifugal atomisation (Kim et al., 1997a) or by mechanical milling (Clark et al., 1998), this decomposition is more or less present and influences the fuel gas retention capabilities. In the literature, no exact decomposition process exists according to these metallurgical characteristics and the general composition process can be seen as:

$$\gamma - U(8Mo) \rightarrow \alpha - U + U2Mo$$

Today, no precise links between the microstructure of the fissile material and its capacity to retain the metastable γ-phase have been established. We propose to study the kinetics and modes of transformation of the γ-phase by in-situ diffraction experiments, on the two main types of powders (atomised and ground) tested in MTRs up to now. Isothermal conditions at 450 °C have been chosen as representative of the fabrication process of the plates. The evolution of the phases in terms of relative fraction and composition was evaluated by Rietveld refinements of data collected by:

- high temperature neutron diffraction, on an atomised powder,
- high temperature X-Ray Diffraction (XRD), on atomised and ground powders (neutron diffraction being not authorized, on this last powder, because of its high ^{235}U enrichment and risks of irradiation).

The methodology adopted in this study consists in, a first step, exploiting the results obtained via neutron diffraction on the atomised powder. Indeed, neutron diffraction is the most adapted

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diffraction method to get information from the whole bulk material (in a transmission mode), especially when considering high density materials like the U(Mo) alloy. After neutron diffraction, the atomised powder was studied by X-ray diffraction to check the relevance of this latter diffraction technique in order to apply it to the ground powder (knowing that the ground powder cannot be studied by neutron diffraction).

2. Materials

2.1. Powder obtained via the atomisation process

Atomised U(Mo) particles, made with natural uranium (0.7% in ^{235}U), have been supplied by the Korea Atomic Energy Research Institute (KAERI) and have been produced, more specifically, by a centrifugal atomisation process (Kim et al., 1997b, 1999).

The main characteristics of this powder are a spherical shape of the particles and a size distribution centred on about 50 μm. The microstructure presented in Fig. 1 reveals small sized grains (≈1 μm), generally with a columnar shape at the periphery and an equiaxed one at the centre of the particles, which are typical features of a microstructure obtained via a fast solidification process.

2.2. Powder obtained via the grinding process

The ground U(Mo) powder, made with enriched uranium (50% in ^{235}U), has been supplied by AREVA-CERCA (Romans/Isère – France) and has been obtained from an arc-melted U(Mo) ingot which has been milled in an inert atmosphere. The main characteristics of the ground particles (see Fig. 2) are an irregular shape, a similar main size distribution around 50 μm (obtained after sieving), and an irregular thin oxide layer on top (see yellow (in the web version) array on Fig. 2) or within the particles (see blue array on Fig. 2) (Clark et al., 1998).

3. Experimental techniques

3.1. High-temperature neutron diffraction

Neutron powder diffraction (NPD) measurements have been performed on the natural U atomised powder at ILL (Laue-Langevin Institute in Grenoble – France) on the D1B line (http://www.ill.eu) equipped with a high resolution powder diffractometer working at a wavelength of 2.52 Å, a position sensitive detector (PSD) of 128° and a furnace able to warm up to 600 °C. A vanadium can filled under vacuum with 10 g of U(Mo) powder was used as the sample holder. The diffraction patterns have been registered over two ranges [$0° \leq 2\theta \leq 128°$]. The collection strategy was to collect a first acquisition at room temperature, to perform acquisitions every 15 min at 450 °C for 40 h and to acquire a final diffractometer at room temperature, after cooling.

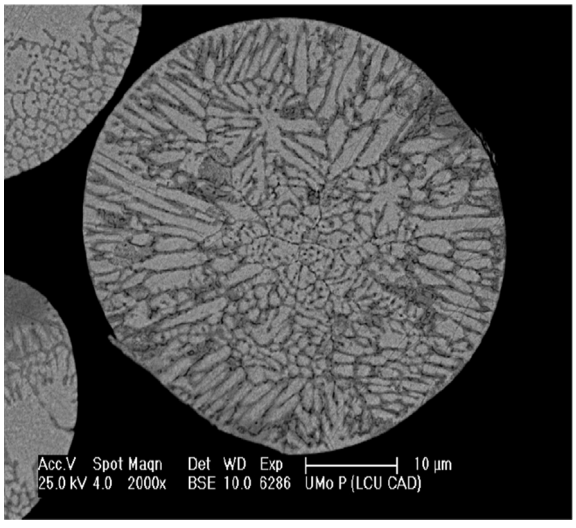

Fig. 1. Scanning electron micrograph (backscattered electron mode) of an atomised U(Mo) particle (cross section).

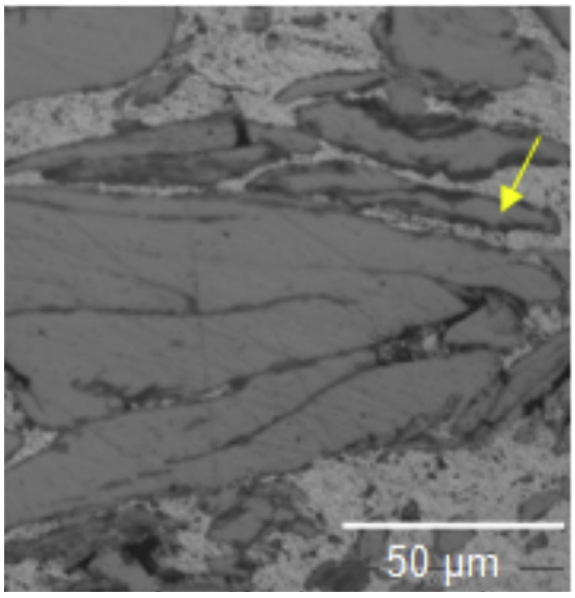

Fig. 2. Optical micrograph of ground U(Mo) powder (cross section).

3.2. High-temperature X-Ray diffraction

High-temperature X-ray diffraction (HT-XRD) measurements were performed at CEA Cadarache – France, under secondary vacuum ($6.0 \cdot 10^{-6}$ mbar) in the angular range $25° \leq \theta \leq 120°$ (0.1° steps and 0.2 s/step counting time), using a BRUKER D8 Advance diffractometer equipped with a θ–θ Bragg-Brentano goniometer and a copper anticathode (Kα1 + Kα2, $\lambda_1 = 1.5406$ Å and $\lambda_2 = 1.5444$ Å). A 1D LynX'Eye detector, with a viewing angle of 3° (2θ), was used. The instrument is placed in a N_2 atmosphere glove box designed for handling nuclear materials. The temperature was ramped up with a MRI Physikalische Gerate GmbH TC-Radiation heater plate (0.5 L volume) which was used to heat the sample up to 450 °C in an alumina crucible.

The temperature was calibrated using a tungsten powder (Aldrich, purity of 99.999%) which was heated in 100 °C stages from ambient temperature to 1700 °C, by comparing the refined lattice parameter as a function of temperature to those given in references Dubrovinsky et al. (1997) and Wang et al. (1998). This operation was performed several times which made it possible to determine an uncertainty of ±15 °C on the temperature that was actually reached, compared with that given by the heater thermocouple.

The measurements strategy was similar to that used for the NPD experiment with: a first scan at ambient temperature, followed by scans every 30 min (for 20 h for the atomised powder and 50 h for the ground powder) during the isothermal annealing at 450 °C, and a last scan after cooling down to ambient temperature.

3.3. Diffraction data refinement

The diffraction data were processed using the Rietveld method, with the FullProf software (Rietveld, 1967; Rodiguez-Carvajal,