Background

In the FUTURE experiment, fuel plates containing U-7wt% Mo atomized powder were irradiated in the BR2 reactor. At a burn-up of approximately 33% 235U (6.5% FIMA or 1.41×10^{21} fissions/cm³ meat), the fuel plates showed an important deformation and the irradiation was stopped. The plates were submitted to detailed PIE at the Laboratory for High and Medium level Activity. The results of these examinations were reported in the scientific report of last year and published in open literature.
Since then, the microstructural aspects of the FUTURE fuel were studied in more detail using transmission electron microscopy (TEM), in an attempt to understand the nature of the interaction phase and the fission gas behavior in the atomized U(Mo) fuel.

Objectives

The FUTURE experiment is regarded as the definitive proof that the classical atomized U(Mo) dispersion fuel is not stable under irradiation, at least in the conditions required for normal operation of plate-type fuel. The main cause for the instability was identified to be the irradiation behavior of the U(Mo)-Al interaction phase which is formed between the U(Mo) particles and the pure aluminum matrix during irradiation. It is assumed to become amorphous under irradiation and as such cannot retain the fission gas in stable bubbles. As a consequence, gas filled voids are generated between the interaction layer and the matrix, resulting in fuel plate pillowing and failure.
The objective of the TEM investigation was the confirmation of this assumption of the amorphisation of the interaction phase. A deeper understanding of the actual nature of this layer and the fission gas behaviour in these fuels in general can allow a more oriented search for a solution to the fuel failures.

Principal results

Analysis of the interaction phase between the atomized particles and the matrix shows that it has a completely featureless character (Figure 1a) and is fully amorphous, as seen by its diffraction pattern (Figure 1b). The amorphous interaction phase is not a good host for the bubble superlattice and the progressing reaction between the pure Al and the U(Mo) will lead to the coalescence of the stored gas into large porosities. The additionally injected fission gas will have a high mobility in the glassy interaction product and will migrate towards the porosities, causing further swelling.

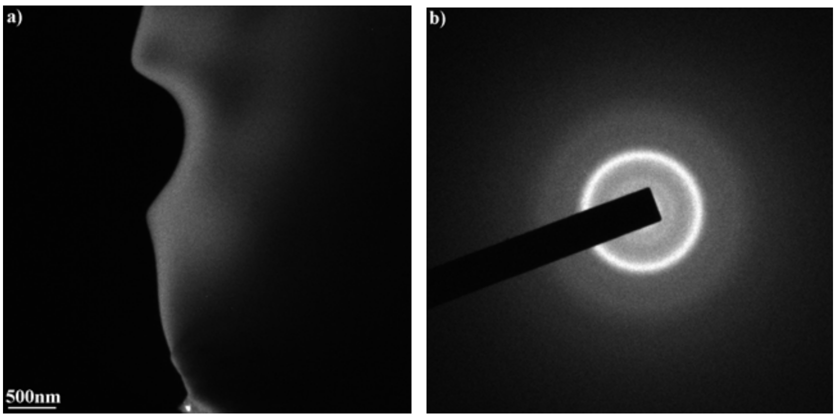

Figure 1: a) Dark field image of the interaction layer with b) the diffraction pattern, demonstrating its fully amorphous character.

A typical dark field image of a U(Mo) grain and its corresponding diffraction pattern are shown in Figure 2. Although the U(Mo) is very clearly crystalline and has a single orientation, as seen from the diffraction pattern, the dark field images contain a lot of grainy contrast. The diffraction pattern (Figure 2c) also shows reflection rings belonging to the UO_2 structure in addition to the sharp reflections of the γ-U structure of the U(Mo) phase. This indicates that the material has oxidized by exposure to the electrolyte and the open atmosphere.

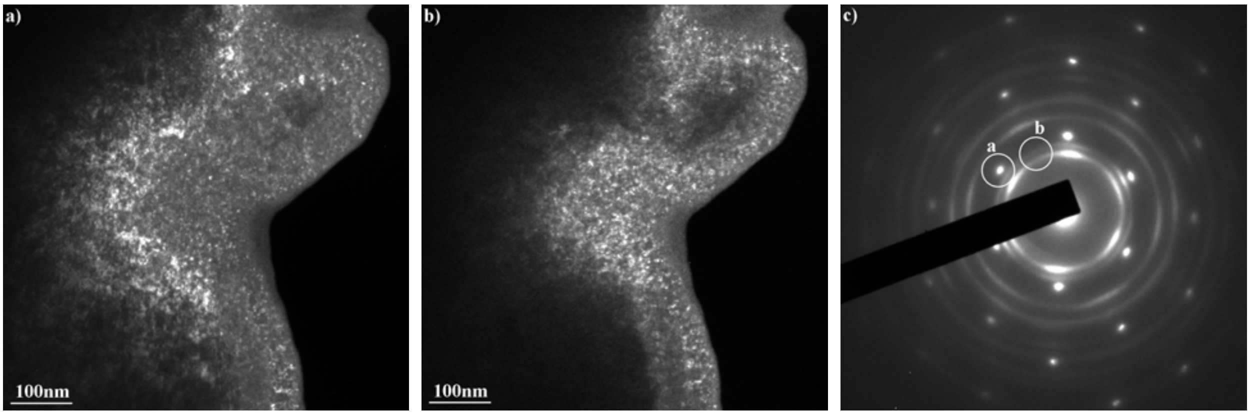

Figure 2 : Dark field images and the corresponding diffraction pattern of a transparent region in the U(Mo) fuel, taken using a) a (110) U(Mo) reflection and b) with part of the UO_2 diffraction ring as indicated by the white circles in the diffraction pattern in c).

When observed more in detail and at higher magnification in bright field, the U(Mo) cells show a lattice of regularly spaced bubbles or voids, which is particularly well visible at slightly under-focus conditions (Figure 3a). The bubbles or voids have a size of around 2 nm and a spacing of roughly 6-7 nm. The orientation of the superlattice is coherent with the U(Mo) lattice orientation, which was checked by observation of the superstructure reflection satellites close to the U(Mo) reflections in the diffraction pattern (see inset of Figure 3b).

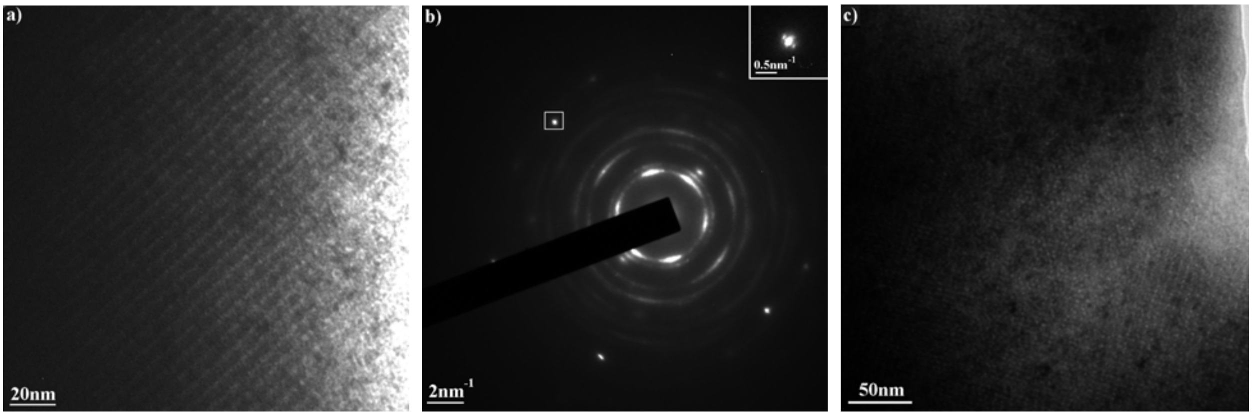

Figure 3 : a) Bright field image of the bubble superlattice found in the U(Mo) particles. b) Corresponding diffraction pattern, where the inset shows an enlarged image of the (222) reflection from the [-211] zone indicated by the square. c) Observation of the bubble superlattice in an adjacent grain at the same position. Here, the orientation is such that individual bubbles can be more clearly observed.

Formation of such gas bubble superlattices has been observed in many metals after implantation with He gas (see publications by P.B. Johnson) at temperatures below 0.2 T_m where T_m is the melting temperature of the metal. In view of the irradiation temperature of ~150°C, the conditions for formation of these lattices are fulfilled.

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Main reference

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