CHARACTRIZATION OF AN IRRADIATED RERTR-7 FUEL PLATE USING TRANSMISSION ELECTRON MICROSCOPY

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ABSTRACT

Transmission electron microscopy (TEM) has been used to characterize an irradiated fuel plate with Al-2Si matrix from the Reduced Enrichment Research and Test Reactor RERTR-7 experiment that was irradiated under moderate reactor conditions. The results of this work showed the presence of a bubble superlattice within the U-7Mo grains that accommodated fission gases (e.g., Xe). The presence of this structure helps the U-7Mo exhibit a stable swelling behaviour during irradiation. Furthermore, TEM analysis showed that the Si-rich interaction layers that develop around the fuel particles at the U-7Mo/matrix interface during fuel plate fabrication and irradiation become amorphous during irradiation. An important question that remains to be answered about the irradiation behaviour of U-Mo dispersion fuels is how do more aggressive irradiation conditions affect the behaviour of fission gases within the U-7Mo fuel particles and in the amorphous interaction layers on the microstructural scale that can be characterized using TEM?

This paper will discuss the results of TEM analysis that was performed on a sample taken from an irradiated RERTR-7 fuel plate with Al-2Si matrix. This plate was exposed to more aggressive irradiation conditions than the RERTR-6 plate. The microstructural features present within the U-7Mo and the amorphous interaction layers will be discussed. The results of this analysis will be compared to what was observed in the earlier RERTR-6 fuel plate characterization.

1. Introduction

Some fuel materials contained in research reactor dispersion fuels have been demonstrated to go amorphous at relatively low fission densities. Both U_3Si and U_3Si_2 fuels have been tested using ion or neutron irradiation techniques and both materials were shown to go amorphous at relatively low exposures [1,2]. Amorphization of interaction layers in U-Mo dispersion fuels has also been assessed [3]. It has been suggested that at low temperatures the phases that comprise the interaction layers in U-Mo/Al and U-Mo/Al-Si alloy matrix dispersion fuels can go amorphous during irradiation, and this has been confirmed with TEM analysis [4,5]. The parameters that have been identified for influencing whether or not a phase will be amorphous are composition, fission rate, and temperature. Due to the fact that at low temperatures most materials will go amorphous if they are exposed to enough irradiation damage, it is of interest to determine if there is a point where U-Mo alloys will go amorphous as well. If amorphization does occur, then the fuel behavior of the U-Mo alloy could change at that point. For example, the fission gas bubble superlattice that has been present in the U7Mo alloy (at least up to a fission density of around 4.5×10^{21} f/cm^3 and a fission rate of around 3.8×10^{14} f/cm^3 s) would disappear, thereby impacting the mobility of the fission gases such that they would be organized differently in the U-7Mo alloy. In fact, in fuels irradiated at higher fission rates a different behavior of the fission gases has been noted when characterizing irradiated U-Mo dispersion fuel plates using optical metallography [6,7]. During this characterization, relatively large bubbles have been observed at the U-7Mo/interaction layer interface. In irradiated U-7Mo dispersion fuels with pure Al, relatively large fission gas bubbles have also been observed, but they were mainly at the interaction layer/Al matrix interface [8]. If in fact the U-7Mo goes amorphous at the higher fission rates, then the fission gases could have been released from the fission gas bubble superlattice, at which time they could migrate to the U-7Mo/interaction layer interface and agglomerate into relatively large bubbles. This would suggest that the Si-rich interaction layer was acting as a barrier to further migration of the fission gas bubbles, allowing the fission gases to possibly migrate through any areas in the interaction layers that were not enriched in Si.

To test the concept that relatively high fission rates could cause U-7Mo alloy to go amorphous, a sample from the low-flux side of an irradiated RERTR-7 fuel plate with Al-2Si matrix has been characterized using TEM. Choosing a sample at this location on the fuel plate allowed for the characterization of a sample with the same matrix composition and a very similar fission density compared to an RERTR-6 sample with Al-2Si matrix that has been characterized using TEM [5], but with the major difference being the fission rate. This would allow for comparison of the RERTR-7 characterization results to those for the RERTR-6 sample to determine the effect of an increased fission rate. For the RERTR-6 plate, the U-7Mo was observed to be crystalline and to contain a fission gas bubble superlattice, and the interaction layer was observed to be Si-rich and to be amorphous. For the RERTR-7 sample, focus was given to crystallinity of the U-7Mo, the identification of the fission gas superlattice, and the attributes of the interaction layer, particularly with regard to the Si content and morphology of any contained fission gas bubbles.

2. Experimental

The RERTR-7 dispersion fuel plate (U-7Mo/Al-2Si), labeled R2R040, was irradiated in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL). A small cylindrical punching of 1.0 mm diameter and roughly 1.4 mm in length was produced from the low flux side of the fuel plate at the Hot Fuel Examination Facility (HFEF) at INL. The estimated local fission density and time-averaged fission rate are 3.3×10^{27} f m^{-3} and 4.3×10^{20} f m^{-3} s^{-1}, respectively. The fuel plate centerline temperature is calculated to be ~90°C. Radioactivity of the fuel punching measured at the contact is ~80 R/β and ~300 mR/γ. A standard TEM sample was prepared by mounting the fuel punching inside a 3.0 mm diameter molybdenum ring using epoxy in a glovebox at the Electron Microscopy Laboratory (EML), followed by mechanical polishing, electrical jet-polishing, and precision ion polishing. Microstructural characterization was conducted using a 200 KV Jeol2010 TEM with a LaB_6 filament.

3. Results and Discussion

The major difference between the irradiation experiments of RERTR-6 and RERTR-7 fuel plates is that the time-averaged fission rate is higher for the latter. However, the previous TEM characterization of an irradiated RERTR-6 fuel plate was performed on a punching sample taken from the high flux side. The work presented in this paper was performed on a fuel punching from the low flux side of RERTR-7 fuel plate. As a result, the RERTR-7 fuel punching has ~27% lower fission density, ~13% higher time-averaged fission rate, and ~17°C lower local fuel centerline temperature (~90°C vs. ~107°C).

Three dimensionally ordered superlattice of fine fission gas bubbles in the crystalline region of U-7Mo fuel particles at low and high resolution are shown in Figure 1. These bright field TEM images were taken at zone <011> of bcc U-7Mo. The presence of a ring pattern in the selected area diffraction (SAD) is due to oxides formed on the sample surface. The average bubble diameter and its standard deviation is 3.1 ± 0.4 nm with measurements over 1066 bubbles. Bubble superlattice is responsible for the satellite spots in a magnified SAD at zone <011> shown in Figure 2. From the previous TEM work on RERTR-6 irradiated fuel, it was found that the bubble superlattice has a fcc structure coherent with the bcc U-7Mo host metal. The orientation relationship among bubble superlattice image, U-7Mo SAD of zone <011> and the satellite spots shown in Fugure 2 is consistent with the previous work [5]. Therefore, the fcc lattice constant for the observed bubble superlattice can be measured from both bubble images or satellite spots. The lattice constants of bubble superlattice measured from bubble images and SAD satellite spots are 12.07 ± 0.06 nm and 12.06 ± 0.15 nm, respectively.

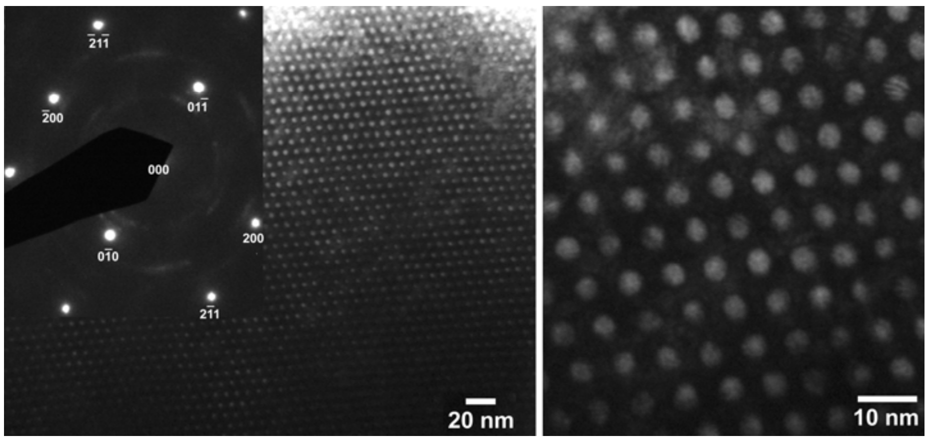

Figure 1. Superlattice of fission gas bubbles shown in low (left) and high (right) resolution bright field images. The inset showing the orientation of bcc U-7Mo fuel at zone <011> where bubble superlattice images were taken. The ring pattern is due to oxidation on sample surface.

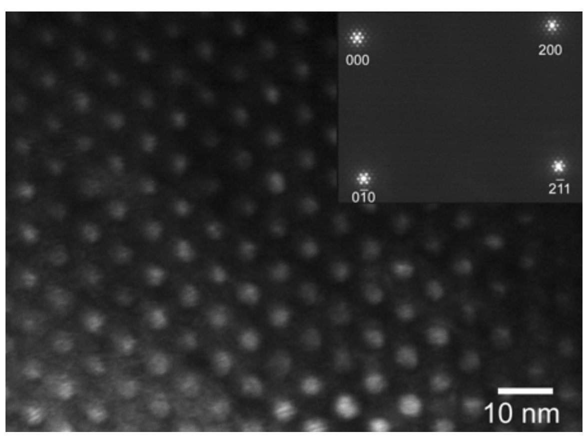

Figure 2. Orientation relationship between bcc U-7Mo alloy and fcc bubble superlattice in the U-7Mo fuel. Note the presence of satellite spots at each major differection spot of U-7Mo at zone <011> due to bubble superlattice.

The smaller size (3.1 vs. 3.5 nm) and larger superlattice constant (12.1 vs. 11.5 nm) in the RERTR-7 sample compared to the RERTR-6 sample may be the results of lower fission density and cooler centerline temperature. The estimated superlattice bubble concentration is

2.3×10^{24} m^{-3}. It is lower than 2.6×10^{24} m^{-3} for RERTR-6. Based on the fission density, if all of the fission gas atoms were to be stored in superlattice bubbles, the bubble size is estimated to be approximately 3.5 nm, which is larger than the measured 3.1 nm. It indicates the fission gas atoms of stable xenon and krypton stored in superlattice bubbles only accounts for 69% of the total stable Xe and Kr gas atoms produced from fissions. This is lower than the 85% (3.7 nm vs. 3.5 nm) found in RERTR-6 fule punching. It suggests that more fission gas atoms are stored in other forms, such as large fission bubbles, for RERTR-7 fuel particles.

It appears there are more large bubbles in the U-7Mo fuel particles. Figure 3 shows a montage of low magnification ($\times$ 2.5K) TEM images cross a relatively large area (> 40 μm) of a fuel particle. Various microstructural features can be indentified, including: Al-2Si matrix, fuel-matrix-interaction (F.M.I.) layer, amorphous and crystalline region of the U-7Mo fuel particle, and large bubbles in the fuel particle, etc. The F.M.I. layers remain thin with a thickness typically around 1~2 μm. All the F.M.I. are found to be amorphous. There are amorphous region of fuel particle near fuel/F.M.I. interface and it can extend into the fuel up to several μm. The energy dispersive x-ray spectroscopy (EDS) measurement indicates a relatively high Si content (> 10 at%) in the amorphous fuel region compared to a lower Si content in the crystalline fuel region (< 2 at% or none). The interfaces of amorphous/crystalline region are illustrated with marks in Figure 3. An abrupt change on SAD and Si content are found at all amorphous/crystalline interfaces in fuel particles. The amorphous region of the fuel particles was not identified from the previous work of RERTR-6 fuel sample even with a higher fission density. This is possibly a result of combined effect of higher fission rate and lower fuel plate centerline temperature on microstructural development.

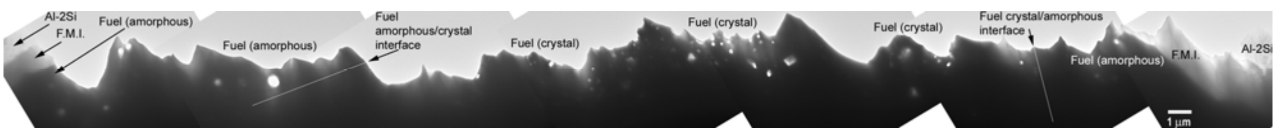

Figure 3. A montage of TEM images showing the Al-2Si matrix, F.M.I. layer, amorphous and crystalline regions of the fuel particle, and large fission gas bubbles in the fuel particle.

A more detailed view of the crystalline region of the fuel particle near the center of the montage in Figure 3 is shown in Figure 4. In addition to superlattice bubbles which can only be seen at high resolution in the crystal region, large bubbles are clearly visible in the transparent areas. Although a quantitative comparison is not available, it is noted that there are more large bubbles found in RERTR-7 than in RERTR-6. The distribution of these bubbles is not uniform as shown in Figure 3. These bubbles are found in both amorphous and crystalline region of the fuel particles.

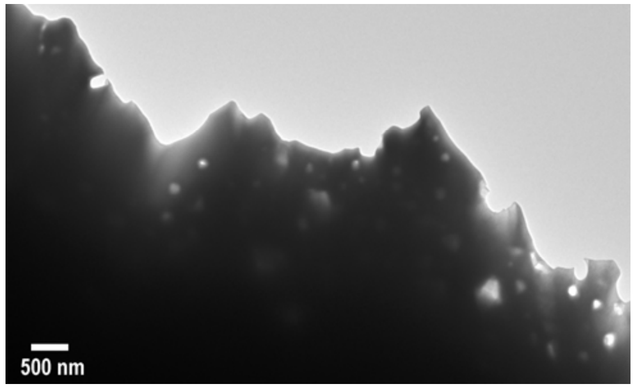

Figure 4. TEM image showing details of the crystalline region of U-7Mo fuel particle near the center of Figure 3. Note that large bubbles found in both amorphous and dcrystalline region of the U-7Mo fuel.

The SAD patterns of crystalline and amorphous regions of the fuel particles and F.M.I. are shown in Figure 5. The ring for amorphous region of the fuel is slightly smaller than that of F.M.I, suggesting a larger nearest-neighbor distance in fuel. There is no noticeable difference in amorphous ring pattern in F.M.I. between RERTR-6 and RERTR-7 fuels. The previous work on RERTR-6 fuel particles did not reveal amorphous regions in the transparent area. The role of high Si content on the formation of amorphous region near the surface of U-7Mo particles is not clear. It is known that Si can react with U to form U-Si compound U_3Si, U_3Si_2, and U-Si. These phases are unstable and can become amorphous under irradiation. The EDS measurements for amorphous regions of fuel particles give (U,Mo)/(Si,Al) ratio to be approximately three. It is speculated that U_3Si type phase may form in the RERTR-7 fuel particles near particle surface area that becomes amorphous under irradiation. It is known that, while U_3Si_2 and USi perform in an acceptable fashion, U_3Si performs poorly under irradiation by development of large fission gas bubbles and bubble interlink [9].

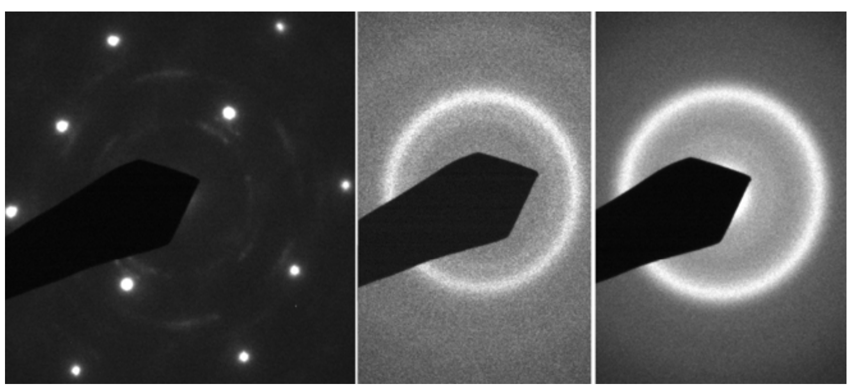

Figure 5. SAD patterns of crystalline region at zone <011> (left); the amorphous region (center) of the U-7Mo fuel particle; and the fuel-matrix-interaction layer (right).

Figure 6 reveals bubbles in the F.M.I. and the fuel particle. The light area in the lower left corner is an Al-2Si matrix. The areas with marks in F.M.I. and fuel are where the EDS measurement were performed. The results from EDS measurement are listed in Table 1. Note that aluminum content across the F.M.I. remains constant while Si decreases and Mo and U increase as EDS probe moving towards the fuel particle. There is a sudden increase in Si content from 1.8 at% to

8.7 at% across the interface of F.M.I. and fuel. The Si content remains high in the amorphous region of the fuel (typically > 10 at.%) and then drops to nearly zero when EDS probe is in the crystalline region in the fuel particle. The average atomic ratio of (U,Mo)/(Si,Al) in amorphous fuel is ~3.1. The average size for large bubbles in the fuel particles (161 bubble counts) and F.M.I. layers (430 bubble counts) are measured to be 159 $\pm$ 87 nm and 54 $\pm$ 45 nm, respectively. The large standard deviations are due to a wide range of bubble sizes with nonuniform distribution.

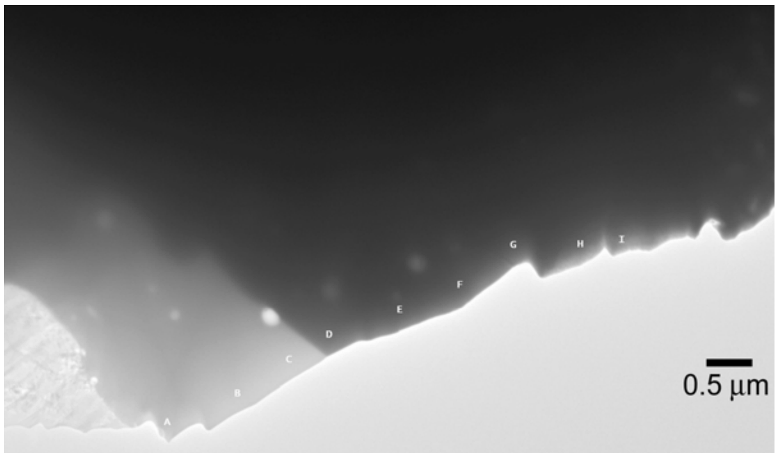

Figure 6. EDS profile shwoing the location of Al-2Si (low left), F.M.I. (grey); amorphous region (D-F) of the fuel and crystalline region (G-I and the right side) of the fuel.

Table 1. EDS measurement (at%) at various locations shown in Figure 6.

Spot	Al	Si	Mo	U	Note
A	78.01	5.06	4.36	12.57	F.M.I. near Al-2Si side
B	78.71	3.45	4.87	12.97	F.M.I.
C	77.60	1.85	5.23	15.32	F.M.I. near fuel side
D	15.98	8.73	17.74	57.54	Fuel, amorphous
E	11.75	10.20	16.79	61.26	Fuel, amorphous
F	11.29	15.31	16.25	57.15	Fuel, amorphous
G	9.16	2.37*	20.98	67.49	Fuel, crystal

* It indicates a relatively large uncertainty due to low signal/noise ratio for Si.

4. Conclusions

TEM characterization of a punching sample from the low flux side of an irradiated RERTR-7 dispersion fuel plate shows the fcc superlattice of fine fission gas bubbles in the crystalline regions of the fuel particles. The average size of superlattice bubbles and the fcc superlattice constant are 3.1 nm and ~12.1 nm, repectively. It is estimated that these bubbles account for approximately 69% of the total stable Xe and Kr gaseous atoms produced from fission. Amorphous regions in fuel particles are identified that are associated with a relatively high Si content next to the F.M.I. The formation of amorphous region in fuel next to F.M.I. is likely a result of higher fission rate and lower fuel centerline temperature. Large bubbles are found in both amorphous and crystalline regions of the fuel particles as well as in F.M.I. The average bubble size in fuel particles and F.M.I. are measured to be approximately 159 nm and 54 nm, respectively.

Acknowledgments

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