CHARACTERIZATION OF THE MICROSTRUCTURE OF IRRADIATED U-MO DISPERSION FUEL WITH A MATRIX THAT CONTAINS SI

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ABSTRACT

RERTR U-Mo dispersion fuel plates are being developed for application in research reactors throughout the world. Of particular interest is the irradiation performance of U-Mo dispersion fuels with Si added to the Al matrix. Si is added to improve the performance of U-Mo dispersion fuels. Microstructural examinations have been performed on fuel plates with Al-2Si matrix after irradiation to around 50% LEU burnup. Si-rich layers were observed in many areas around the various U-7Mo fuel particles. In one local area of one of the samples, where the Si-rich layer had developed into a layer devoid of Si, relatively large fission gas bubbles were observed in the interaction phase. There may be a connection between the growth of these bubbles and the amount of Si present in the interaction layer. Overall, it was found that having Si-rich layers around the fuel particles after fuel plate fabrication positively impacted the overall performance of the fuel plate.

1. Introduction

The United States Reduced Enrichment for Research and Test Reactors (RERTR) Fuel development program is actively developing low enriched uranium (LEU) fuels for the world’s research reactors that are currently fueled by uranium enriched to more than 20% 235U [1].

To assess the performance of U-Mo dispersion fuels with Si-doped matrices, different reactor experiments have been conducted using the Advanced Test Reactor (ATR). For the purpose of conducting scanning electron microscopy examinations on irradiated samples, the RERTR-6 experiment has been of particular interest since the experiment was extracted from ATR over three years ago, and as a result, small samples can be handled in the Electron Microscopy Laboratory (EML). This paper will discuss results of recent microstructural characterization using a scanning electron microscope (SEM) that was performed on a fuel plate with Al-2Si matrix, irradiated as part of the RERTR-6 experiment. Optical metallography (OM) data will also be included. Focus will be given to the partitioning behavior of Si amongst the microstructural phases and how the presence of Si affects the development of interaction layers at the U-7Mo/matrix interface. Comparisons are made to the other irradiation experiments that have been reported for fuel plates with U-7Mo fuel dispersed in Al-2Si matrix.

2. Experimental

2.1 Irradiation Testing

The RERTR-6 experiment was the first experiment to test “second generation” U-Mo fuels designed to overcome the fuel performance problems encountered in U-Mo/Al dispersions [2]. In this experiment, the fuel materials were tested to high burn-up under moderate flux and moderate temperature conditions. The fuel plates were positioned edge-on to the core, and as a result had a neutron flux across the widths of the fuel plates.
Previous papers discussed characterization results for other RERTR-6 fuel plates with Si-containing fuel meat matrices [3, 4]. This paper focuses on U-7Mo dispersion fuel plates with Al-2Si matrix. The irradiation conditions for the two characterized fuel plates with Al-2Si matrix are enumerated in Table 1. One plate (R2R020) was characterized using only OM and the other plate (R2R010) was characterized using OM and SEM. R2R010A is a punching taken from the low flux side of the fuel plate that was used for SEM, and R2R010B was taken from the high flux side. Included in this table are the irradiation conditions for other reported experiments that used plate-type, U-7Mo dispersion fuel with Al-2Si matrix.

Table 1. Irradiation conditions for U-7Mo/Al-2Si dispersion fuel plates irradiated as part of RERTR-6, IRIS-3 [5], and IRIS-TUM [5] reactor experiments.

<table>
<thead>
<tr>
<th>Fuel Plate Label</th>
<th>Peak Temp.(°C)</th>
<th>Ave. Fission Density (10^{21} f/cm^3)</th>
<th>Ave. Fission Rate (10^{14} f/cm^3s)</th>
<th>Peak Heat Flux (W/cm^2)</th>
</tr>
</thead>
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<tr>
<td>R2R010A</td>
<td>94</td>
<td>2.4</td>
<td>2.0</td>
<td>98</td>
</tr>
<tr>
<td>R2R010B</td>
<td>109</td>
<td>4.5</td>
<td>3.8</td>
<td>148</td>
</tr>
<tr>
<td>R2R020</td>
<td>104</td>
<td>3.1</td>
<td>2.7</td>
<td>139</td>
</tr>
<tr>
<td>IRIS-3</td>
<td>83</td>
<td>3.4</td>
<td>3.0</td>
<td>196</td>
</tr>
<tr>
<td>IRIS-TUM</td>
<td>103</td>
<td>4.2</td>
<td>3.6</td>
<td>~250</td>
</tr>
</tbody>
</table>

2.2  Microstructural Characterization

For as-irradiated fuel plates, OM was performed on a transverse cross section taken from the mid-plane of a fuel plate. To conduct SEM analysis on as-irradiated plates, a punching process was first used in the Hot Fuel Examination Facility to generate one-mm-diameter cylinders that contained a sampling of the fuel meat. These samples were then transferred to EML where they were mounted, polished, and coated with a thin layer of Pd. SEM analysis, with energy and wavelength dispersive spectroscopy (EDS/WDS), was performed on the mounted samples to characterize the microstructure and to determine the partitioning behavior, during irradiation, of different fuel and matrix components between the different fuel meat phases.

3. Results and Discussion

3.1 Optical Metallography

For fuel plates R2R010 and R2R020, an OM image of a full transverse cross section taken at the mid-plane of the as-irradiated microstructure, along with higher magnification images at the low and high flux side of each fuel plate, are presented in Figs. 1 and 2. The overall microstructures for both fuel plates were very similar.

Fig. 1. A low magnification optical micrograph (a) of the R2R010 fuel microstructure and an image at the (b) low and (c) high flux side of the fuel plate.
3.2 Scanning Electron Microscopy

SEM analysis was performed on punching samples generated from fuel plate R2R010. Images of the microstructure observed for a sample produced at the low flux side of the fuel plate are presented in Fig. 3. Like was the case for the OM images (see Fig. 1), the nominal thickness of the interaction layer was observed to be around 1-2 μm. X-ray mapping was employed to determine the partitioning behavior of fuel and matrix components (see Fig. 4). Si was enriched in the interaction layer, except in some discrete locations where “nodules” were present in the interaction layer (see Fig. 4e). A higher magnification image of a “nodule” is presented in Fig. 3d. For the gaseous fission products, Kr was observed in the fuel and interaction layer phases, and Xe and Cs could also be found in the fuel meat matrix. Some of the highest Xe concentrations were found at the interface between two contacting U-7Mo particles. The solid fission products Nd and Ru were primarily found in the fuel and interaction layer phases, and small concentrations seemed to be present in the fuel meat matrix.
For the punching from the high flux side of the fuel plate, relatively narrow Si-rich interaction layers were also observed around the U-7Mo fuel particles. SEM micrographs of the microstructure are presented in Fig. 5. Like was the case for the sample from the low flux side of the plate, some local regions in the interaction layer were observed to be relatively thicker. Compositional analysis indicated that these regions contained negligible Si. Detailed analysis of one of these regions (area 1 in Fig. 5a) revealed the presence of fission gas bubbles in the interaction layer. This microstructural feature has never been observed before in any other RERTR-6 samples characterized to date. Composition analysis along a line shown in Fig. 6a indicated that where Si concentrations remained relatively high in the interaction layer no fission gas bubbles could be resolved up to high magnifications. On the other hand, where the Si concentration was significantly lower, easily resolvable fission gas bubbles could be observed. It was only in this very localized region of the sample that fission gas bubbles could be resolved in the interaction layer.
Fig. 5. Backscattered electron images of the high flux side of the R2R010 fuel microstructure. Location 1 in (a) is where fission gas bubbles were observed in the interaction layer, and location 2 is near an area of a fractured fuel particle where fission gas bubbles could be resolved in the U-7Mo.

Fig. 6. Backscattered electron images (a,b) of an area of the R2R010 fuel microstructure on the high flux side of the fuel plate. The line in (a) indicates the location where a linescan compositional analysis was taken, and (b) shows how the right side of the line, which passed through a Si-depleted region of interaction layer, coincided with an area where relatively large fission gas bubbles could be observed.

Characterization of as-fabricated dispersion fuel plates before insertion into ATR has shown that Si-rich layers are present around the fuel particles after fabrication of dispersion fuel plates with Al-2Si matrix [3], and based on the results reported here for the as-irradiated samples with Al-2Si matrix, these initial layers do not exhibit an overall significant increase in thickness and do not develop significantly large areas of gross porosity. This agrees well with the SEM characterization that has been performed on other fuel plates with matrices that contain significant Si (0.88 wt% or greater) that were irradiated as part of the RERTR-6 experiment [3, 4]. This type of interaction layer behavior during irradiation is a significant improvement compared to the RERTR fuel plates that were tested in ATR with just Al as the matrix, which developed wide interaction layers and large pores at the interaction layer/Al interface [6]. For another plate-type, dispersion fuel with Al-2Si matrix (IRIS-3) that was irradiated in the OSIRIS reactor, an irradiation behavior that differed from RERTR-6 fuel plates was observed [7]. Characterization of the IRIS-3 fuel plate indicated that the fuel/matrix interaction layers around the U-7Mo particles were similar to those that have been observed for U-7Mo dispersion plates with just Al, in that most of the interaction layers that formed did not contain significant Si. However, it was observed that in regions near Si-rich matrix particles an enrichment of Si was present in the interaction layer. Similarly, a pin-type fuel element with Al-2Si matrix (KOMO-3) that was irradiated developed interaction layers without significant Si [8]. However, the characterization results for both these fuel plates seemed to indicate that there was a positive effect of having Si present in the matrix in that it reduced the amount of fuel/matrix interaction.
The results of a third reported irradiation test, which used ground U-7Mo powders and Al-2Si matrix (IRIS-TUM), also indicated a reduction in interaction layer growth because of Si in the matrix [9]. One potentially significant difference between the RERTR-6, Al-2Si-matrix plates and those irradiated in IRSIS-3 and KOMO-3 is that Si-rich interaction layers were present in the RETR-6 plates before irradiation, whereas the KOMO-3 and IRIS-3 plates did not have these layers, since they were processed at lower temperatures. This suggests that having Si-rich fuel/matrix interaction layers present after fabrication may play a significant role in improving the performance of U-Mo dispersion fuel plates, because these layers do not change much in thickness and do exhibit stable behavior under the conditions of the RERTR-6 experiment.

The fact that some resolvable porosity could be found in the interaction layer of the sample from the high flux side of R2R010 suggests that there is a link between the Si concentration of the interaction layer and the stability of the very fine fission gas bubbles that typically cannot be resolved using an SEM. When the local Si concentration in the interaction layer is below a certain threshold under certain reactor conditions, the fission gases present in very fine bubbles may begin to agglomerate to form larger bubbles, which in turn may become mobile resulting in movement to the interaction layer/Al-Si alloy matrix interface. In [10], it was suggested that 5 at% Si is required in the interaction layer to retain stable behavior. Future characterization of irradiated samples will continue investigating how the Si concentration in the interaction layer affects the behavior of fission gas bubbles.

4. Conclusions

1. Si-enriched fuel/matrix interaction layers produced during fabrication of U-Mo dispersion fuels with Al-2Si matrix exhibit overall stable behavior when fuel plates are irradiated up to the conditions of the RERTR-6 experiment (moderate power and burnup). This means that the layers remain relatively thin and do not develop areas of significantly large porosity.

2. The local Si content within the fuel/matrix interaction layers may affect the behavior of the fission gases that are dissolved. Depending on the reactor conditions of an experiment, there may be a minimum Si content required to keep the behavior of dissolved fission gases stable. If concentrations fall below a certain level, then relatively large fission gas bubbles can grow, and the fission gases could become more mobile.

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References
[6]. G. L. Hofman et al., RERTR 2003, Chicago, IL, October, 2003