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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 either Al-0.2Si or 4043 Al (~4.8% Si) alloy matrix in the as-fabricated and/or as-irradiated condition using optical metallography and/or scanning electron microscopy. Fuel plates with either matrix can have Si-rich layers around the U-7Mo particles after fabrication, and during irradiation these layers were observed to grow in thickness and to become Si-deficient in some areas of the fuel plates. For the fuel plates with 4043 Al, this was observed in fuel plate areas that were exposed to very aggressive irradiation conditions.

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% ^{235}U.

To assess the performance of U-Mo dispersion fuels with Si-doped matrices, different reactor experiments have been conducted using the Advanced Test Reactor. Experiments have been run with dispersion fuels that have Al-0.2Si, Al-2Si, Al-5Si, 6061 Al and 4043 Al alloy matrices. This paper will discuss results of recent microstructural characterization that was performed on fuel plates that were irradiated as part of the RERTR-6 and RERTR-7 experiments that have either Al-0.2Si or 4043 Al alloy as the 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 [1]. In this experiment, the fuel materials were tested to high burn-up under moderate flux and moderate temperature conditions. The RERTR-7 experiment was a more aggressive test and employed fuel enriched to 58% ^{235}U. RERTR-7 was divided into two parts: RERTR-7A and RERTR-7B [2].

In Table 1, the irradiation conditions for some specific plates from the RERTR-6 and RERTR-7 experiments are presented. These plates had either Al-0.2Si or 4043 Al as the matrix and are the plates focused on in this paper. Chemical analysis of the 4043 Al revealed a composition of 4.81Si-0.20Fe-0.14Ti-0.16Cu-0.01Cr-0.01Mn-bal Al. Less than 0.01 wt% of Zn and Mg was measured.

Table 1. Irradiation conditions for fuel plates R5R020, R3R030, R3R040, and R3R050.

Fuel Plate Label	Exper.	Matrix	Peak Temp. (˚C)	Ave. Fission Density (10^{21} f/cm^3)	Ave. Fission Rate (10^{14} f/cm^{3}s)	Peak Heat Flux (W/cm^2)
R5R020	RERTR-6	Al-0.2Si	117.1	3.30	2.83	130.52
R3R030	RERTR-6	4043 Al	97.5	3.26	2.80	101.5
R3R040	RERTR-7	4043 Al	N/A	5.03	6.46	N/A
R3R050	RERTR-7	4043 Al	139.9	4.90	6.30	299.3

2.2 Microstructural Characterization

For the as-fabricated fuel, microstructural characterization was performed on transverse cross-sections using scanning electron microscopy with energy dispersive spectroscopy and wavelength dispersive spectroscopy (SEM/EDS/WDS).

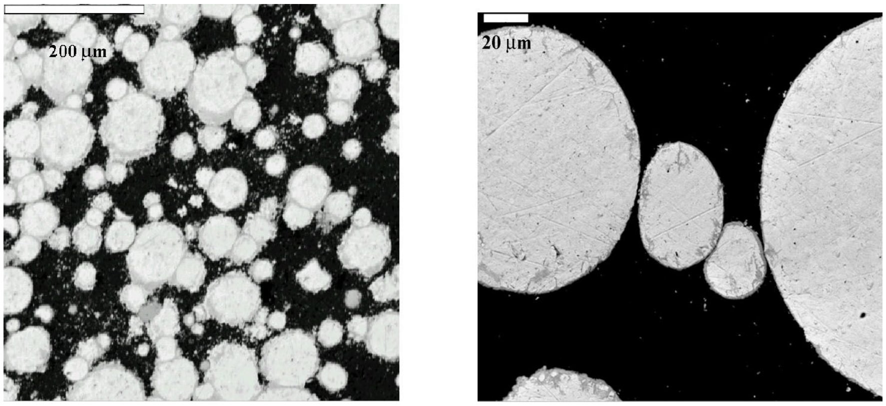
For the as-irradiated fuel plates, optical metallography (OM) was performed on transverse cross section taken from the mid-plane of the fuel plate. For the SEM/EDS/WDS analysis of the as-irradiated plates, a punching process was first used in the Hot Fuel Examination Facility to generate one-mm-diameter cylinders that contained a sampling of the fuel meat, and then in the Electron Microscopy Laboratory these cylinders were mounted, polished, and coated with a thin layer of Pd [3]. SEM/EDS/WDS analysis was performed to characterize the microstructure and determine how different fuel and matrix components partitioned between the different phases during irradiation.

3. Results

3.1 As-Fabricated Plates

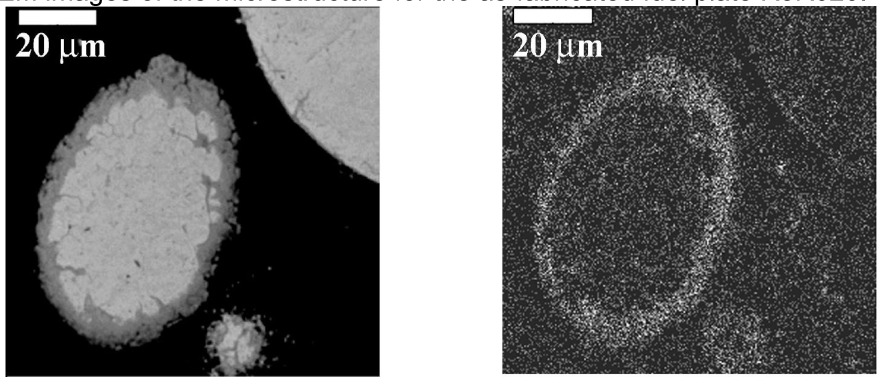
During the fuel fabrication campaign for the RERTR-6 experiment, archive fuel plates were produced that were later characterized to determine the starting microstructure of the fuel before irradiation. R3R020 was the fuel plate that was characterized to determine the starting microstructure of a fuel plate with U-7Mo fuel particles and 4043 Al matrix. For the Al-0.2Si matrix fuel, no as-fabricated fuel plate was produced to serve as an archive due to the aggressive fabrication schedule being followed to get all the plates that comprised the RERTR-6 experiment into reactor. Results from diffusion experiments using U-7Mo and low-Si Al-Si alloys at temperatures representative of fuel fabrication temperatures can be looked at to get an idea of how these plates would look after fabrication [4].

An SEM image of the microstructure of the R3R020 fuel plate is presented in Figure 1. Thin fuel/matrix interaction layers are present around all the fuel particles. These interaction layers were a result of the exposure of the fuel plates to relatively high temperatures during the rolling and blister annealing steps that was a part of the fuel fabrication process. During rolling, the plates were exposed to around 500˚C for up to one hour. During blister annealing the plates were exposed to 485˚C ±20˚C for 30 minutes [5]. Also, during fuel fabrication, the original γ−phase U-7Mo alloy apparently decomposed to α-U and γ’. This resulted in some localized fuel/cladding interaction, as shown in Fig. 1b. X-ray maps that were produced (see Fig. 2) show that these interaction layers were enriched in Si. The maximum Si content of the interaction layer was measured by SEM/EDS to be 45 at% with a maximum (Al+Si) concentration of 69 at%, and the (Al+Si)/(U+Mo) ratio varied between 1.7 and 2.2. The layers were on the order of 1-2 μm thick. U, Mo, and Al were also mapped and showed U and Mo in the fuel; U, Mo, and Al in the interaction layer; and, Al in the matrix. No oxygen enrichment was observed in the interaction layers.



(a) (b)

Fig. 1. SEM images of the microstructure for the as-fabricated fuel plate R3R020.



(a) (b)

Fig. 2. (a) An SEM image of fuel particles in as-fabricated plate R3R020, and (b) a Si X-ray map showing the enrichment of Si in the interaction layer.

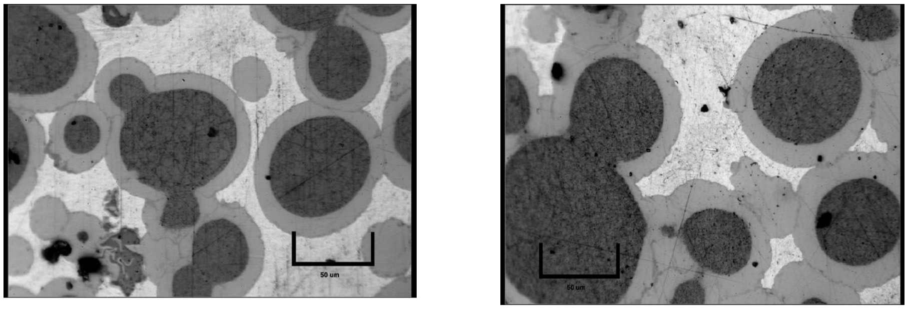
For the RERTR-7 fabrication campaign, archive plates with U-7Mo-2Zr and U-7Mo-1Ti fuel particles in 4043 Al matrices were examined with SEM/EDS/WDS. The observed microstructures were very similar to those shown in Figs. 1 and 2, and the U, Mo, Al, and Si partitioning behavior was very similar.

3.2 As-Irradiated Plates

3.2.1 Optical Metallography

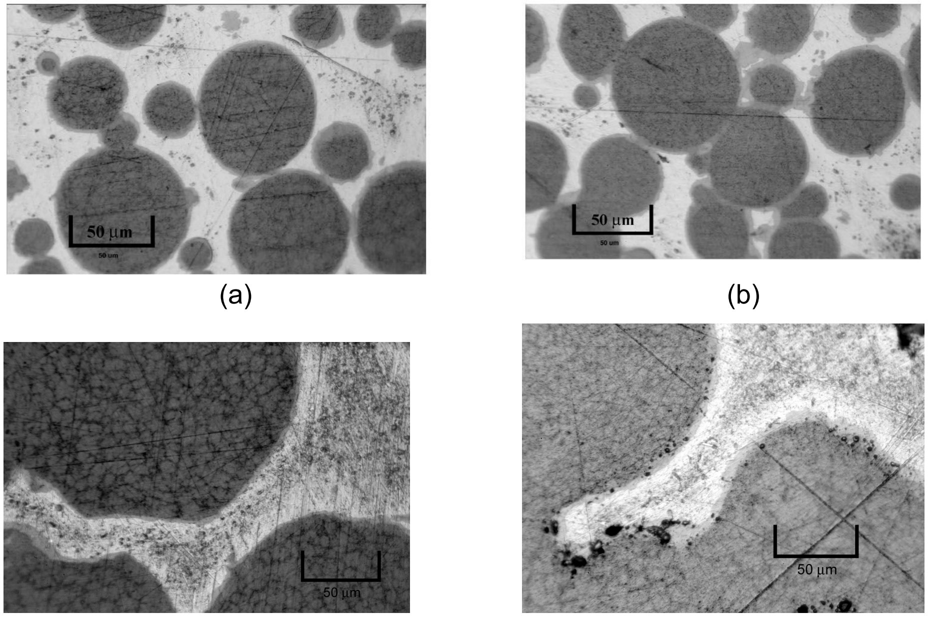
Fuel plate R5R020 was a fuel plate with an Al-0.2Si matrix that was irradiated as part of the RERTR-6 experiment. OM images that were taken in different areas of a full transverse cross section taken at the mid-plane of the as-irradiated microstructure are presented in Fig. 3. Due to the fission density gradient that was present across the width of the fuel plates for the RERTR-6 experiment, there was a variation in the interaction layer thickness that was observed around the fuel particles. The thickest layers (~10 μm) were observed at the highest-burnup edge of the plate.

The fuel plates with 4043 Al alloy matrix that were irradiated in RERTR-6 or RERTR-7 experiments included R3R030, R3R040, and R3R050. Representative OM images of the microstructures observed along full transverse cross sections taken at the mid-plane of R3R030 and R3R050 fuel plates are presented in Fig. 4. Figs 4a and 4b show the relatively narrow interaction layers observed across the mid-plane of R3R030 (around 1 to 2 μm). Figs 4c and 4d show the thicker layers (up to 10 μm) that were observed across the mid-plane of R3R050. R3R040 exhibited interaction layer thicknesses that were similar to those observed for R3R050. The thickest layers were observed at the edge of the plates that were exposed to the highest burnup.



(a) (b)

Fig. 3. OM images of the R5R020 fuel plate microstructure observed at the edges of the fuel plate with the (a) lowest and (b) highest burnups.



(a) (b)

(c) (d)

Fig. 4. OM images of the fuel microstructures observed for fuel plate R3R030 towards the (a) highest and (b) lowest burnup edges of the plate and for plate R3R050 towards the edges with the (c) lowest and (d) highest burnups.

3.2.2 Scanning Electron Microscopy

SEM images of the microstructure observed for the Al-0.2Si matrix fuel plate R5R020 are presented in Fig. 5. Like was the case for the OM images (see Fig. 3), the thickness of the interaction layer was observed to be around 10 μm. X-ray mapping was employed to determine the partitioning behavior of fuel and cladding components (see Fig. 6 for Si). No concentration gradients for U, Mo, Al, or Si were observed in the interaction layer, and point-to-point composition analysis showed that the average composition (determined from fourteen points), in at%, of the interaction layer was around 83.1Al-2.7Mo-14.3U (± ~2 at%). As expected, based on the lack of Si in the generated Si X-ray maps, negligible Si was measured in the interaction layer. The Si was observed in precipitates that were present in the fuel meat matrix.

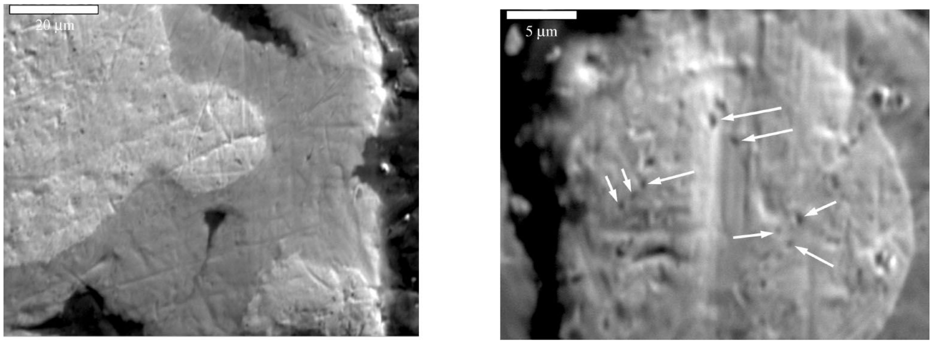
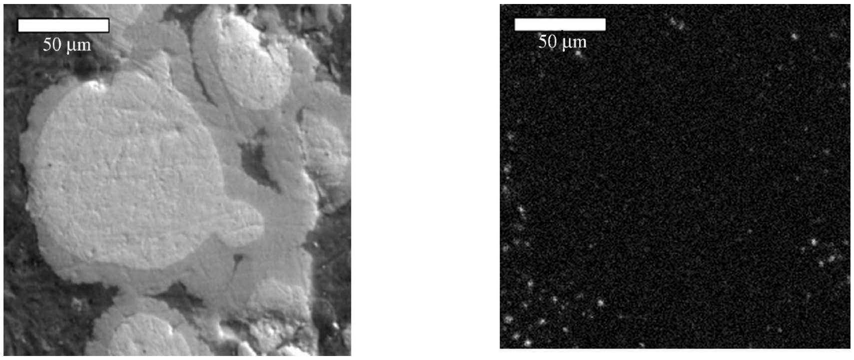


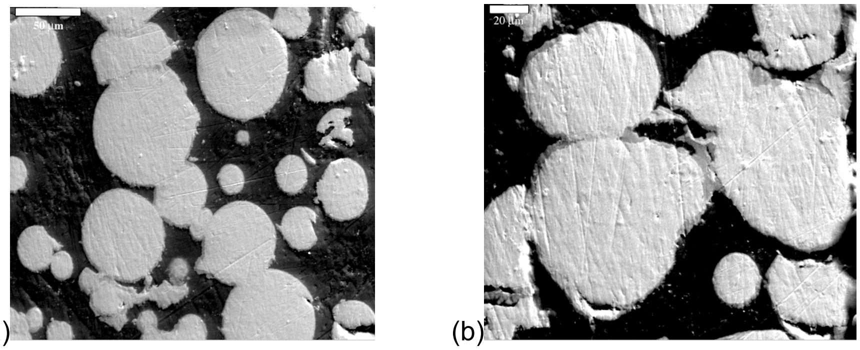
Fig. 5. SEM images of the microstructure observed for fuel plate R5R020. The arrows indicate pores observed in the fuel.



(a) (b)

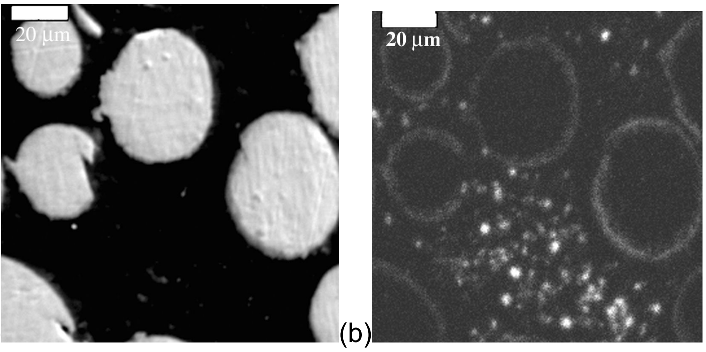
Fig. 6 Secondary electron image (a) and Si X-ray map (b) for fuel plate R5R020.

For fuel plate R3R030, two different types of microstructure were observed: one had around 1 to 2 µm-thick interaction layers and the other had layers that were around 10 µm thick, based on looking at the interaction layer thickness around the largest-diameter particles. The thickest layers coincided with regions of the fuel plate that had achieved around 100% LEU burnup. Fig. 7 shows the fuel plate microstructure, and Fig. 8 shows a Si X-ray map where the thinner layers were observed. Figs. 9 and 10 show the same where the microstructure displayed thicker layers. In the microstructure where the thinner layers were observed, the layers were enriched in Si, and there were precipitate free zones (PFZ) around many of the particles. These PFZs have been interpreted as the result of the recoil damage zones that extend around each of the U-Mo particles to a distance of around 10 µm, and it has been suggested that the Si-containing precipitates in these regions dissolve and the Si from the precipitates diffuses towards the fuel/matrix interface [6]. For the areas of the microstructure with the thicker interaction layers, negligible Si was observed in the layers. The original Si in the interaction layers appeared to have come out as precipitates in the matrix. Point-to-point composition analysis at fifteen different locations within the ~10 µm-thick-layer indicated an approximate composition, in at%, of 82.4Al-2.5Mo-15.1U (± ~2 at%). Because the Si-rich layer was smaller than the spatial resolution of the individual composition measurements, the composition of this layer could not be measured.



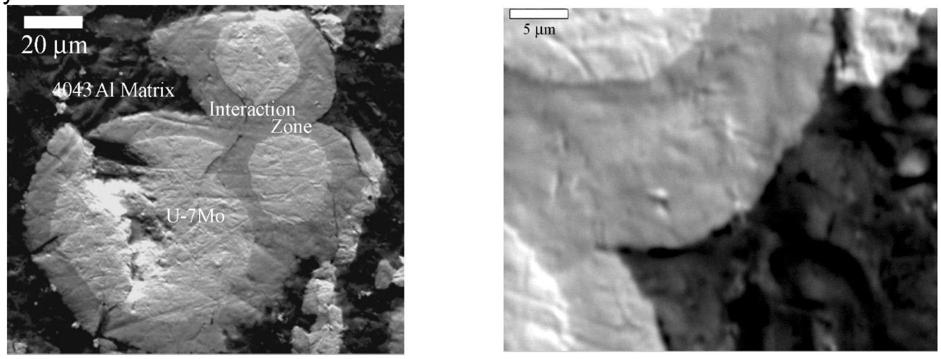
(a) (b)

Fig. 7. SEM images of the microstructure for fuel plate R3R030 where relatively thin interaction layers were observed.



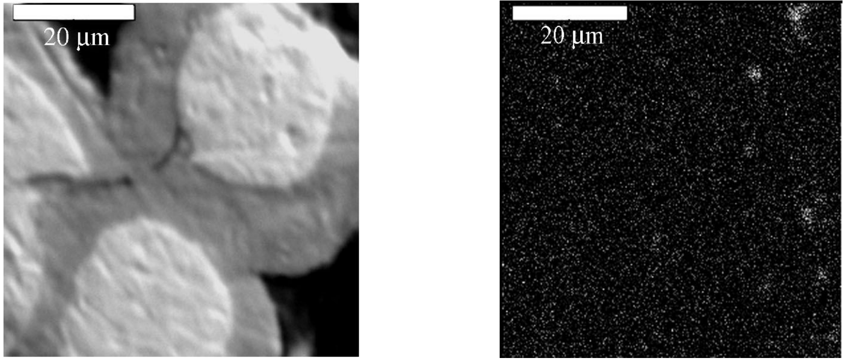
(a) (b)

Fig. 8. SEM image (a) and Si X-ray map (b) for R3R030 where interaction layers were relatively thin.



(a) (b)

Fig. 9. SEM images of the microstructure where relatively thick interaction layers were observed in fuel plate R3R030.



(a) (b)

Fig. 10. SEM image (a) and Si X-ray map (b) in an area of the R3R030 fuel plate where relatively thick interaction layers were observed.

4. Discussion

Based on the characterization that was performed on as-fabricated plates that went into the RERTR-6 and RERTR-7 experiments, it is clear that Si-rich interaction layers were already present around the U-7Mo fuel particles before any of the fuel plates with 4043 Al alloy matrix were inserted into the Advanced Test Reactor. These interaction layers were a result of the exposure of the fuel plates to relatively high temperatures during the rolling and blister annealing steps that were a part of the fuel fabrication process. Based on interdiffusion studies that have been performed using U-7Mo and low-Si Al alloys [4], there is a good chance that the fuel plate with Al-0.2Si also had pre-existing Si-rich interaction layers.

Looking at the OM images for the irradiated fuel plates, it is clear that in some cases the relatively thin interaction layers that were present in the fuel plates before irradiation have grown in reactor, and in some cases have reached an approximate thickness of 10 μm. For the fuel plates with 4043 Al matrices, the interaction layer thickness can approach 10 μm in the areas of the fuel plates that achieved around 100% LEU burnup. Based on SEM analysis, the 10-μm-thick interaction layers contain negligible Si. Conversely, when fuel particles have retained the relatively thin interaction layers during irradiation and are characterized using the SEM, appreciable Si is observed. This suggests that during irradiation enough Si must diffuse to the interaction layers in order to keep the fabrication-generated layers stable (i.e., large pores do not form like for the U-Mo/Al matrix fuels). If this does not transpire, then the Si-rich layers become unstable, and the U-Mo-Al interdiffusion behavior that is typical during the irradiation of U-Mo dispersion fuels with Al as the matrix takes over. Other researchers have also concluded that it is important to have sufficient Si in the matrix of a U-Mo dispersion fuel in order to get good irradiation performance [7].

The Al-0.2Si matrix dispersion fuels do not appear to contain enough Si to keep the thin, Si-rich interaction layer stable. Only thick interaction layers were observed that contained negligible Si. This in combination with the information from the R3R030, R3R040, and R3R050 fuel plates would suggest that there is some Si concentration level between 0.2 wt% and 4.81 wt% where there would be enough Si in the matrix to keep the Si-rich interaction layer stable, resulting in good fuel plate irradiation behavior to high burnups. It has been shown that fuel plates with 2.0 wt% Si added to the matrix also exhibit good irradiation performance [8]. Even with 4.81 wt% Si in the matrix of a fuel element, porosity and 10-μm-thick interaction layers can be observed in some local areas of a fuel plate, but this is only observed where the fuel plates had been exposed to extremely high burnup levels (i.e., ~100% LEU burnup). These high burnup levels are beyond what a typical research reactor fuel would see, and even with these features present, the fuel plates displayed overall good irradiation behavior.

5. Conclusions

Based on the characterization of as-fabricated and irradiated U-7Mo dispersion fuel plates with either Al-0.2Si or 4043 Al alloy as the matrix, the following conclusions can be drawn:

1. Fuel plates that were inserted into the Advanced Test Reactor as part of the RERTR-6 and RERTR-7 experiments already had Si-rich interaction layers present around the fuel particles, due to the fuel plate fabrication process.

2. After irradiation, the RERTR-6 fuel plate with Al-0.2Si alloy matrix appeared to have developed only relatively thick fuel/matrix interaction layers that contained negligible Si.

3. The fuel plate with 4043 Al (4.81 wt% Si) matrix, irradiated as part of the RERTR-6 experiment, contained Si-rich interaction layers that were about the same thickness as those that were produced during fabrication, along with relatively thick layers that contained

negligible Si. The thick layers seemed to form in areas of the fuel plate that were exposed to the highest burnup. Thick interaction layers could also be found in fuel plates that had 4043 Al matrices that were irradiated in the aggressive RERTR-7 experiment.

4. In order for Si-rich fuel/matrix interaction layers to remain stable in U-Mo dispersion fuels during irradiation, it appears there needs to be a sufficient supply of Si in the matrix, and the optimal Si content is somewhere between 0.2 and the 4.81 wt%.

Acknowledgments

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