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ABSTRACT

The starting microstructure of a dispersion fuel plate can have a dramatic impact on the overall performance of the plate during irradiation. To improve the understanding of the as-fabricated microstructures of dispersion fuel plates, SEM and TEM analysis have been performed on RERTR-9A archive fuel plates, which went through an additional hot isostatic procsssing (HIP) step during fabrication. The fuel plates had depleted U-7Mo fuel particles dispersed in either Al-2Si or 4043 Al alloy matrix. For the characterized samples, it was observed that a large fraction of the γ-phase U-7Mo alloy particles had decomposed during fabrication, and in areas near the fuel/matrix interface where the transformation products were present significant fuel/matrix interaction had occurred. Relatively thin Si-rich interaction layers were also observed around the U-7Mo particles. In the thick interaction layers, $(U)(Al,Si)_3$ and $U_6Mo_4Al_{43}$ were identified, and in the thin interaction layers $U(Al,Si)_3$, $U_3Si_3Al_2$, U_3Si_5, and $USi_{1.88}$-type phases were observed. The $U_3Si_3Al_2$ phase contained some Mo. Based on the results of this work, exposure of dispersion fuel plates to relatively high temperatures during fabrication impacts the overall microstructure, particularly the nature of the interaction layers around the fuel particles. The time and temperature of fabrication should be carefully controlled in order to produce the most uniform Si-rich layers around the U-7Mo particles.

1. Introduction

In order to improve the irradiation performance of U-Mo dispersion and monolithic fuels, approaches to mitigate the affect of fuel/matrix and fuel/cladding interactions are being pursued. For the dispersion fuels, Si is being added to the matrix so that a Si-rich interaction phase develops at the U-Mo/matrix interface that exhibits good irradiation performance. For the monolithic fuels, a Zr barrier is inserted between the U-Mo foil and the cladding. During the fabrication of each fuel type, it has been demonstrated that the

time at high temperature will impact the final as-fabricated microstructure. [1,2]. These microstructures can contain a variety of different phases depending on the fabrication history.

For U-Mo dispersion fuel plates with Al matrices that contained Si, an investigation has been performed using scanning electron microscopy (SEM) and transmission electron microscopy (TEM) to identify the exact phases that are contained in the interaction zones that develop around the U-7Mo particles during fuel plate fabrication. The dispersion fuel plates that were analyzed were exposed to significant times at high temperatures and exhibited thin Si-rich layers along with relatively thick Si-depleted interaction layers. This allowed for the opportunity to investigate how Si-rich layers change during fabrication to form the Si-depleted layers.

This paper will describe the specific phases that were found in the different archive U-Mo dispersion fuel plates. Comments will be made about how the development of different phases may affect the overall irradiation performance of a fuel plate.

2. Experimental

The production of fuel plates for the RERTR-9A experiment involved the standard dispersion fuel fabrication steps. As a result, these fuel plates were exposed to 500°C for up to 1 hour during rolling, and to 485°C for 30 minutes during the blister-annealing step. In addition, an extra hot isostatic press (HIP) step (30 minutes at 500°C) was employed on the fuel plates to improve the bonding at the interface between the fuel meat and the 6061 Al cladding for these high-density (8 gU/cc) dispersion plates. The HIP process constituted a 75 minute heat-up to 500°C for a 30 minute soak, followed by a 75 minute cool-down to room temperature.

SEM analysis was performed on archive dispersion RERTR-9A fuel plates using a Zeiss Ultra-55 field emission SEM with X-ray energy dispersive spectroscopy (XEDS). Specimens for TEM were prepared with a focused ion beam (FIB) in-situ lift-out (INLO) technique using a FEITM 200 TEM. Initially, a Pt layer was deposited onto the selected area of interest, to protect the surface of the sample from the accelerated Ga+ ion beam. The high energy Ga+ beam was utilized to mill material creating a trench on both sides. The edges of the sample were then milled leaving only a small bridge of material so that the sample remained attached to the bulk alloy. A W omniprobe was then lowered in and Pt welded to the bridge connecting the sample and the bulk alloy. The partially attached edge of the specimen was then milled completely to release the sample. The W omniprobe, with the TEM specimen still welded to it, was then lifted away from the stage and lowered toward a slotted copper TEM grid. The sample was then Pt welded to the copper grid. Each specimen was thinned further, milled to about 100 nm in thickness in order to obtain electron transparency. The FEI/Tecnai F30 300keV TEM/STEM equipped with a Fischione high angle annular dark field (HAADF) detector was used for the TEM analysis. For each specimen, both low and high magnification HAADF STEM images were collected for bulk and detailed analysis. Selected area and convergent beam diffraction patterns (SAED and CBED) were collected and indexed to identify the phase constituents.

3.   Results
3.1   SEM Analysis

The results from an earlier SEM analysis of the RERTR-9A archive samples [1] are summarized in Table. 1.

Table 1. Results of SEM Compositional Analysis of As-Fabricated RERTR-9A U-7Mo Dispersion Plates [1].

Matrix	Fabrication Details	Si-rich Layer Description	Notes
4043 Al (4.81 wt%Si)	Roll bonded and HIPed (500˚C/15 ksi/30 min)	Up to 7 μm thick. Uniform layer. Al: 30.2 (14.1)* Si: 24.5 (8.5) Mo: 5.8 (3.2) U: 39.6 (16.7)	Bulk interaction (~10% area).** Al: 56.9 (7.2)*** Si: 0.8 (0.5) Mo: 5.6 (1.1) U: 36.8 (6.6)
Al+2Si (mixture of pure Al and pure Si powders)	Roll bonded and HIPed (500˚C/15 ksi/30 min)	Varies from 2 to 5 μm thick. Not uniform. Al: 28.0 (23.1)* Si: 10.7 (10.3) Mo: 7.9 (3.5) U: 53.4 (22.4)	Bulk interaction (~20% area). Al: 53.2 (5.2)* Si: 1.8 (0.6) Mo: 5.9 (1.3) U: 39.7 (4.0)
Al-2Si (alloy powders)	Roll bonded and HIPed (500˚C/15 ksi/30 min)	From 2 to 5 μm thick. Not uniform. Al: 36.5 (25.0)* Si: 11.4 (11.0) Mo: 6.6 (3.6) U: 45.6 (23.7)	Bulk interaction (~20% area). Al: 53.2(5.2)*** Si: 1.2 (0.7) Mo: 5.9 (1.3) U: 39.8 (4.0)

* layer composition in at% (standard deviation), based on 10 measurements.
**   Bulk interaction refers to the formation of the Si-deficient layer.
*** layer composition in at% (standard deviation), based on 7 measurements.

From Table 1, it can be seen that the samples contained Si-rich layers around the fuel particles, along with some layers that contained little or no Si. Si X-ray maps generated for the different samples have confirmed the presence of Si-rich layers around the U-7Mo particles in the areas where the Si-deficient interaction layers were not present.

Fig. 1 shows backscattered electron images of the Si-deficient layers in the three RERTR-9A fuel plates. Up to 20% of the microstructural area consisted of the Si-deficient phase for the samples with Al+2Si mixture as the matrix or Al-2Si alloy as the matrix. The sample with 4043 Al alloy matrix had approximately 10% of the area that contained the Si-deficient layer.
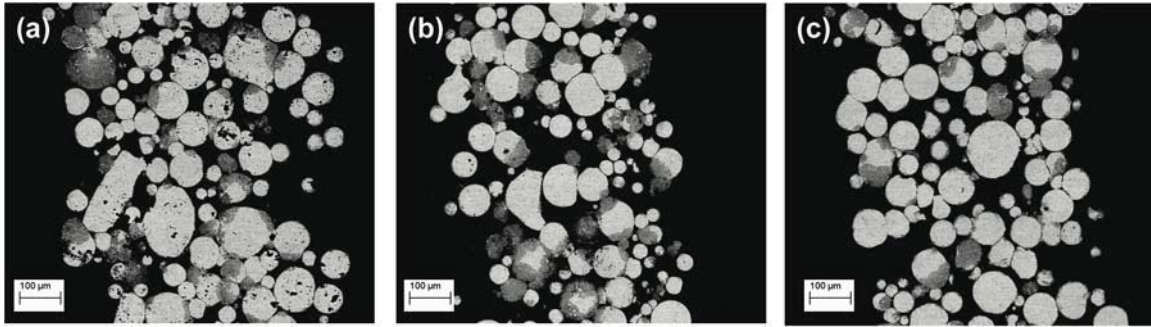
Fig. 1.Backscattered electron images of the microstructures observed for the as-fabricated RERTR-9A fuel plates with (a) 4043 Al, (b) Al+2Si, and (c) Al-2Si alloy matrices, respectively. The bright phase is U-7Mo alloy, the dark phase around the U-7Mo particles is the matrix, and the medium-gray phase is the Si-deficient interaction layer. Not observable are the thin, Si-rich interaction layers around the U-7Mo particles.

In Figure 2 are representative backscattered electron images of the microstructure observed for each of the three samples in areas where the interaction layer was relatively thin. Figure 3 shows a higher magnification image of areas in the U-7Mo fuel particles where islands of transformed U-7Mo could be observed and areas where the decomposed U-7Mo alloy was in contact with the matrix, which resulted in increased interaction. Some fuel particles were totally consumed by the interaction product (see Figure 4).
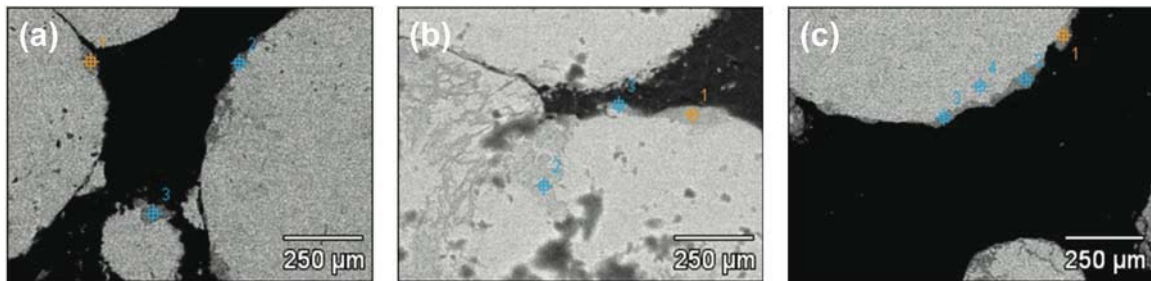

Fig. 2.Backscattered electron images of the microstructures observed for the as-fabricated RERTR-9A fuel plates with (a) 4043 Al, (b) Al+2Si, and (c) Al-2Si alloy matrices, respectively. Most of the interaction layers in these areas of the samples are relatively thin and Si-rich.
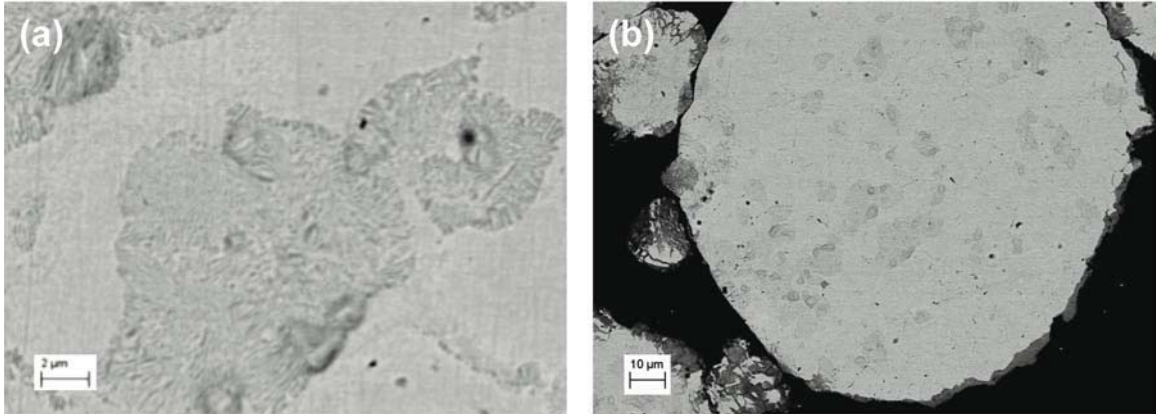
Fig. 3.Backscattered electron images for the U-7Mo/Al-2Si sample showing (a) decomposition in the U-7Mo alloy and (b) the morphologies of the interaction layers around different fuel particles.
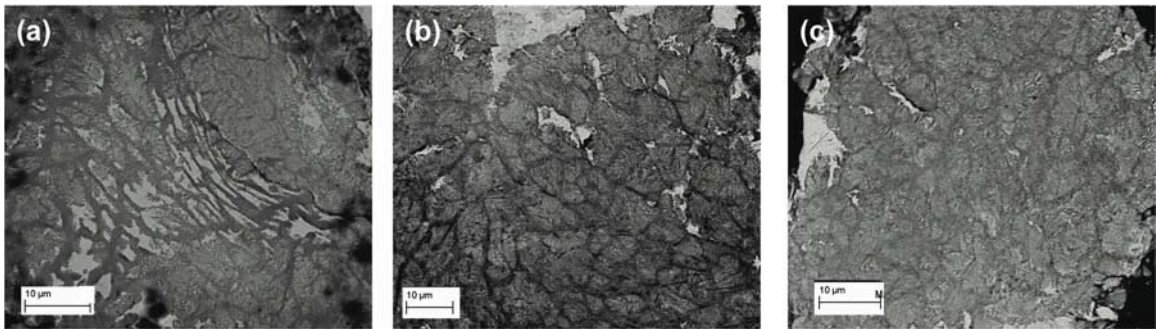

Fig. 4.Backscattered electron images of the microstructures observed for the as-fabricated RERTR 9A fuel plates with (a) 4043 Al, (b) Al+2Si, and (c) Al-2Si alloy matrices, respectively, where some fuel particles were completely consumed by interaction products.

3.2 TEM Analysis

TEM analysis was performed in two different regions of the U-7Mo/4043 Al sample, one at a location where there was thick interaction layer and another where the interaction layer was relatively thin. One region of the U-7Mo/Al-2Si sample where the interaction layer was relatively thin was also analyzed.

Figure 5 shows HAADF TEM micrographs from an area along a decomposed U-7Mo particle/matrix interface for the U-7Mo/4043 Al sample. A relatively large interaction layer developed in this location. CBED patterns were examined at specific locations in the interaction layer that confirmed the existence of cubic $U(Al,Si)_3$, orthorhombic $U_6Mo_4Al_{43}$, and orthorhombic α-U phases. The $U(Al,Si)_3$ phase contained small amounts of Mo, based on XEDS analysis via TEM.
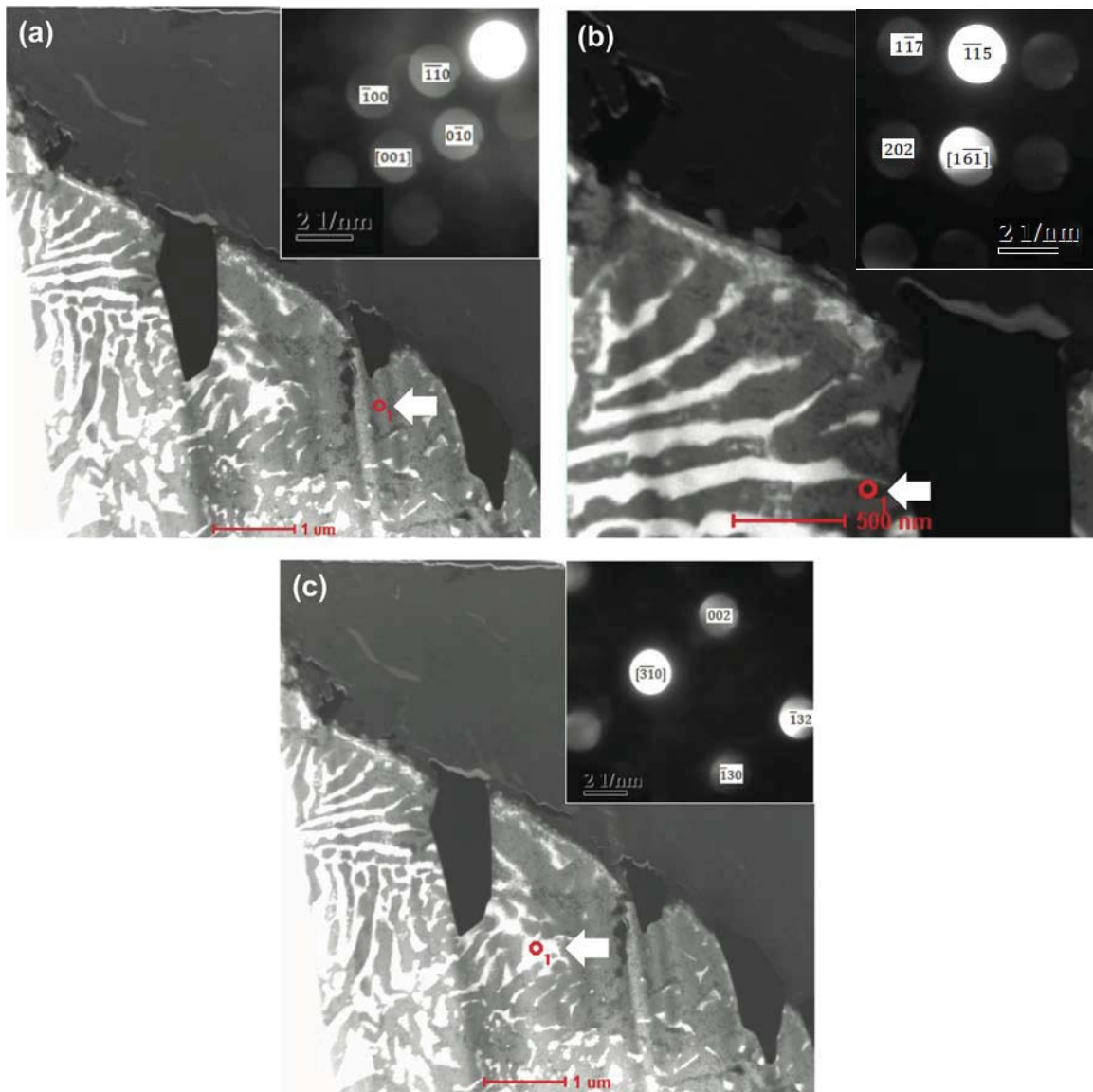
Fig 5. HAADF TEM micrographs and corresponding CBED patterns from the U-7Mo/4043 Al sample at a location along a decomposed U-7Mo particle/matrix interface where the particle had reacted with the matrix to form reaction product (medium gray): (a) a region consisting of $U(Al,Si)_3$ phase; (b) a region where $U_6Mo_4Al_{43}$ was identified; and (c) a fuel region where α-U was identified.

Figure 6 shows an area where a narrower interaction layer developed at the interface between U-7Mo and 4043 Al. CBED analysis indicated the presence of tetragonal $U_3Si_3Al_2$ in the layer. XEDS analysis via TEM indicated up to 3 at% Mo was present in this phase. Near the thin layer (Fig. 6b), an area with increased interaction was observed. This appeared to be at a location of the sample where the original γ-(U,Mo) phase had decomposed, and in this localized area where the αU was present, a relatively high rate of interdiffusion with Al had taken place. This accelerated interdiffusion has been observed

in other studies looking at heat-treated U-Mo dispersion samples, where the α-U in the U-Mo powders displayed a relatively high rate of interdiffusion with Al. [3,4].
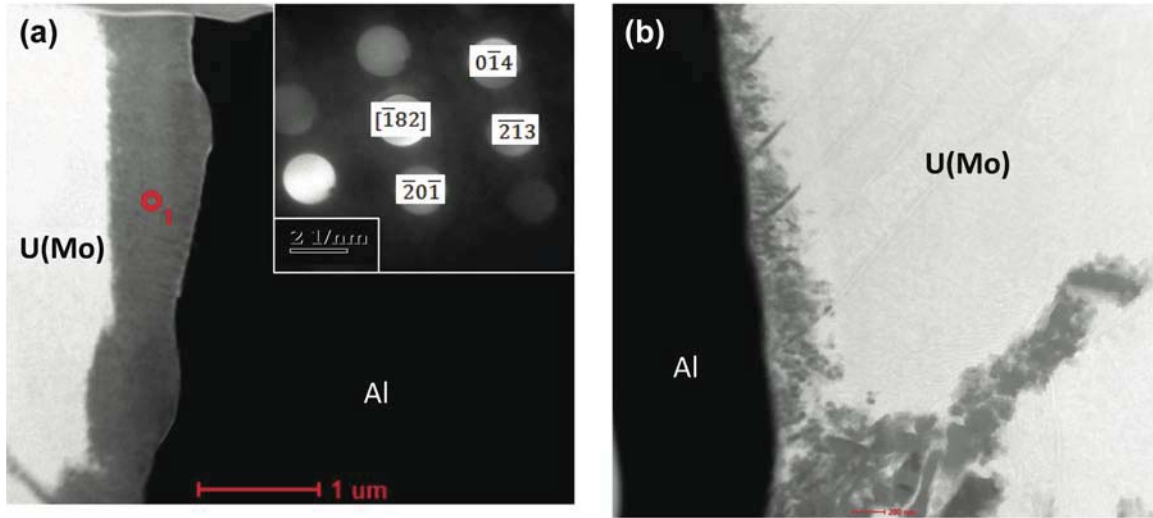

Fig 6. HAADF TEM micrographs and the corresponding CBED pattern from the U-7Mo/4043 Al sample at the U-7Mo (bright)/matrix (black) interface where a relatively thin interaction layer (medium gray) was observed: (a) CBED of region 1 corresponds to $U_3Si_3Al_2$; (b) onset of transformation and accelerated interaction.

Figure 7 shows an area where a thicker Si-rich interaction layer developed at the interface between U-7Mo and 4043 Al. Four different phases were identified in this layer: hexagonal U_3Si_5, $U(Al,Si)_3$, Al, and another very minor uranium-silicon phase (maybe USi).
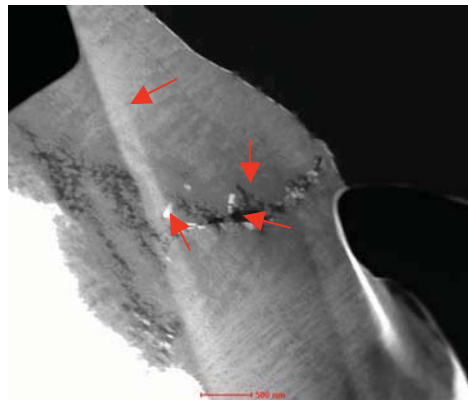

Fig 7. HAADF TEM micrograph showing four locations in a relatively thick Si-rich interaction layer where separate phases were identified. The dark phase to the right and white phase to the left are Al and U-7Mo, respectively.

For the sample with U-7Mo particles dispersed in Al-2Si matrix, TEM analysis was performed at an area where relatively thin interaction layer was present. HAADF TEM micrographs showing the locations in the interaction layer where diffraction analysis was performed are presented in Figure 8. A tetragonal $U_{1.88}$-type of phase was present nearest the unreacted U-7Mo, and a cubic U(Al,Si)$_3$-type phase was present adjacent to the unreacted matrix as presented in Fig. 7. Negligible Mo was present in these phases.
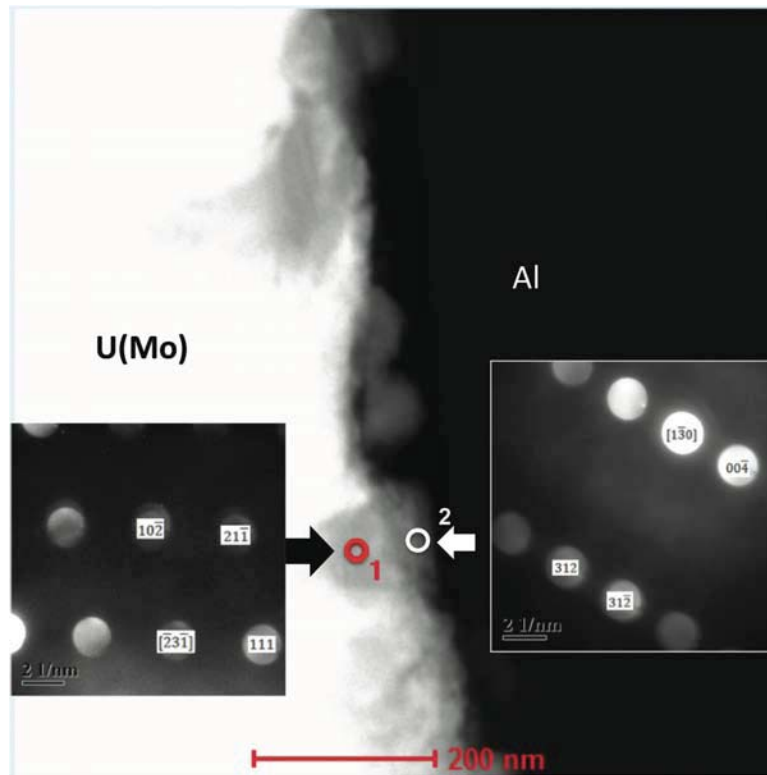

Fig 8. HAADF TEM micrographs from the U-7Mo/Al-2Si sample (a-c) at the U-7Mo (bright)/matrix (black) interface where a relatively thin interaction layer (medium gray) is present. Circles indicate regions where diffraction analysis identified the presence of (1) $U_{1.88}$ and (2) U(Al,Si)$_3$.

4.   Discussion

Based on the SEM and TEM analysis that was performed on RERTR-9A archive dispersion fuel plates, decomposition of the U-7Mo alloys during fabrication can have a dramatic impact on the thickness of the fuel/matrix interaction layers that can develop and the types of phases that are present in the layers for fuel plates with 4043Al and Al-2Si matrices. The markedly thicker interaction zones can be associated with locations where a decomposed fuel region was in contact with the matrix. The narrower Si-rich interaction layers did not appear to have areas of decomposition in close proximity. These zones were observed to contain ternary U-Al-Si phases (viz. U(Al,Si)$_3$ and $U_3Si_3Al_2$) and binary U-Si phases (USi$_{1.88}$ and U_3Si_5). It was found that within the thin Si-rich interaction layer, the tetragonal USi$_{1.88}$-type phase was present in a zone nearest the

unreacted U-7Mo, and the cubic $U(Al,Si)_3$-type phase was present in a zone adjacent to the unreacted matrix. Only $U(Al,Si)_3$ was found both in the thin Si-rich interaction layer along with the thicker Si-deficient interaction layer. $U_6Mo_4Al_{43}$ was found only in the thick Si-deficient interaction layer.

TEM results have also been reported for two other samples taken from the RERTR-9A fuel plate with Al-2Si matrix [1]. This previous analysis focused on the interface between decomposed U-7Mo particles and the matrix. An ordered fcc $U(Al,Si)_3$ phase was observed in the interaction layer that contained some Mo. No evidence of a $U_6Mo_4Al_{43}$ phase was observed. The presence of the $U(Al,Si)_3$ phase agrees with the results of this paper.

Comparing the results of this study with those reported for diffusion couple tests conducted at 500°C or 550°C using U-7Mo and other Si-containing matrices, viz. 6061 Al, there is good agreement in terms of the development of relatively thick interaction layers near decomposed U-7Mo and narrower interaction layers where decomposed U-7Mo was not apparent [5,6]. Composition analysis showed that both the thick and narrow interaction layers contained a Si enriched phase and a phase more enriched in Al. Results from an X-ray diffraction analysis of a U-7Mo/6061 Al couple annealed at 550°C for 3 hours [5] showed results for regions near the thicker interaction zone where $U(Al,Si)_3$ and $U_6Mo_4Al_{43}$ phases were identified. This agrees with the results of this paper where both of these phases were also identified near the thicker interaction zone. In a diffusion couple between U-7Mo and an Al A356 alloy (7.1 wt% Si) annealed at 550°C, $U(Al,Si)_3$, UMo_2Al_{20}, and U_3Si_5 phases were observed in the diffusion zone [7]. In [8], diffusion couples were annealed between U-7Mo and Al-Si alloys and then characterized using μ-XRD. In the diffusion couples that contained either an Al-2Si alloy or 4043 Al, two types of zones made up the formed interaction layer. The zone nearest the U-7Mo contained UAl_3 and $U_6Mo_4Al_{43}$, and the other zone contained $U(Al,Si)_3$ and UMo_2Al_{20}. For the couples with alloys that contained more than 5 wt% Si, it was observed that both U_3Si_5 and USi_2 developed in the interaction zone. Similar U-Si phases were observed in some fuel plate samples characterized in this study.

As discussed in [1], the fuel plates in the RERTR-9B experiment never went through the extra HIP step, as did the RERTR-9A plates. The result was interaction layers around the U-7Mo particle that were much thinner and more uniform after fabrication. Consequently, when the RERTR-9B fuel plates went into reactor very little, if any, of the $U_6Mo_4Al_{43}$ phase was present at the interface of the fuel particles. Instead, $U(Al,Si)_3$, $U_3Si_3Al_2$, U_3Si_5, and USi_2 were most likely present with possibly some other yet to be identified minor phases. The significance of the RERTR-9A having significant amounts $U_6Mo_4Al_{43}$ in the diffusion zone is that the presence of a phase with a composition like UAl_4 in an irradiated fuel plate has been linked to poor fuel performance [9]. Whereas, fuel plates with interaction layers that are relatively thin and Si-rich have been found to exhibit good irradiation performance [10].

5.   Conclusions

Based on the TEM characterization of RERTR-9A archive fuel plates with 4043Al and Al-2Si matrices that were exposed to high temperatures for relatively long times, comments can be made about how decomposition of the U-7Mo fuel affects interaction layer development. When fuel plates are exposed to temperatures near 500˚C for relatively long times, large interaction layers can develop adjacent to areas where the U-7Mo fuel has decomposed, and these layers can contain $U(Al,Si)_3$ and $U_6Mo_4Al_{43}$ phases. In areas along the U-7Mo/matrix interface where fuel decomposition is not apparent, relatively narrow, Si-rich layers develop that can contain $U(Al,Si)_3$, $U_3Si_3Al_2$, U_3Si_5, and $USi_{1.88}$. As a result, during fabrication of dispersion fuel plates the time at high temperatures should be controlled in order to produce interaction layers that are narrow and Si-rich.

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