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ABSTRACT

Three depleted uranium alloys are successfully cast for the radiation stability studies of the fuel-cladding interaction product using proton irradiation. Scanning electron microscopy (SEM) analysis indicates the presence of the phases of interest: $U(Si,Al)_3$, $(U,Mo)(Si,Al)_3$, and a mixture of UMo_2Al_{20}, $U_6Mo_4Al_{43}$, and UAl_4. Irradiation with 2.6 MeV protons at 200ºC to the doses of 0.1, 1.0, and 3.0 displacement per atom (dpa) are carried out.

1. Introduction

The RERTR Fuel Development Program is tasked with developing new low enrichment uranium (LEU) nuclear fuels that can be employed to replace existing high enrichment uranium (HEU) fuels currently used in some research reactors throughout the world. An important part of the fuel development program is an effort to conduct irradiation testing to better understand in-reactor fuel performance and to provide key data that can be incorporated into computer models that are used to model in-reactor fuel performance. For dispersion type fuels, radiation stability of the interaction layers at the interface of fuel particle and cladding matrix plays an important role in fuel performance. Proton irradiation studies of these interaction products will provide important insights on the microstructure stability under irradiation damage.

A variety of phases have the potential of forming in irradiated RERTR fuels as a result of fuel/matrix or fuel/cladding interactions. To study the radiation stability of these potential phases, three depleted uranium alloys are fabricated with compositions of 67U-5Si-28Al (alloy-A), 48U-5Mo-47Al (alloy-B), and 69U-4Mo-20Al-7Si (alloy-D). The first alloy composition selected is close to that of a $U(Si,Al)_3$ phase. This phase has been observed to form in uranium-silicide dispersion fuels and exhibits stable performance under irradiation [1]. The second composition is near that of $(U,Mo)Al_7$, a composition observed in interaction layers of the current version of U-Mo dispersion fuels that use Al as the matrix, which showed poor irradiation performance at very high burnup [2]. In order to improve the performance of U-Mo dispersion fuels, the RERTR program has been investigating the use of Si additions to the cladding matrix to influence fuel/matrix interaction such that a more stable interaction product will form. The idea is that by having Si participate in the inter diffusion process, then it is likely that a $(U,Mo)(Si,Al)_3$ phase will form and remain stable under irradiation, like the $U(Si,Al)_3$ phase did in the uranium-silicide fuels [3]. As a result, the third alloy has a composition near that of a $(U,Mo)(Si,Al)_3$ phase.

The first objective of this work was to verify that the microstructural response of $U(Si,Al)_3$ phase and the phase mixture that exist with a composition of $(U,Mo)Al_7$ under proton irradiation were consistent with the fuel performance reactor tests. The second was to investigate if the radiation stability of the interaction product, $(U,Mo)(Si,Al)_3$ phase was similar to $U(Si,Al)_3$ phase in the silicide fuel. Finally, phase stability, amorphization, and cavity formation and distribution as a function of irradiation dose were to be established.

2. Experiment

Three depleted uranium (DU) alloys were cast using arc melt. Ingot for each alloy weighs approximately 30 grams. High purity Mo, Al, and Si at 99.999% were used for alloy fabrication. These ingots were wrapped in Ta foil, sealed in a stainless steel tube, and homogenized at 500°C for 200 hours. Table 1 lists the material information for this proton irradiation study.

Table 1. DU alloys cast for proton irradiation studies.

Sample designation	A	B	D
Composition (wt%)	67U-5Si-28Al	48U-5Mo-47Al	69U-4Mo-20Al-7Si
Composition (at%)	$U_{19}Si_{12}Al_{69}$	$U_{10}Mo_3Al_{87}$	$U_{22}Mo_3Al_{56}Si_{19}$
To study F.C.I. product	$U(Si, Al)_3$	UMo_2Al_{20}, UAl_4, $U_6Mo_4Al_{43}$ mixture	$(U, Mo)(Si, Al)_3$
Found in fuel type	H.E.U.	L.E.U. (U-Mo)	L.E.U./Al-Si Clad
Anticipated performance	Good	Not good	Good
Microstructure stability	Stable	Cavity + Swelling	Stable (?)

SEM analysis was performed to identify the phases of interest. Samples were mounted and polished through 1 μm polishing compound. The mounted samples were inserted into a ZEISS Model 960A scanning electron microscope that was equipped with an Oxford wavelength dispersive spectrometer (WDS) and energy dispersive spectrometer (EDS) that employed ISIS LINK software. Secondary electron images were generated to determine the alloy microstructures, and the WDS and EDS spectrometers were employed to generate X-ray maps and to perform point-to-point compositional analysis.

For proton irradiation, the ingots were cut into 300-400 μm thin slices with a low speed saw and core drilled to the disc samples of 3.0 mm diameter. The disc samples were mechanically wet polished to the 1200 grit finish. An average of 12-15 disc samples were loaded on the irradiation stage for each irradiation. A liquid metal interface was used between the back of the disc samples and the stage to improve the thermal conduction for improved temperature control. Proton irradiations were conducted using a tandem accelerator at the University of Wisconsin. The 2.6 MeV proton beam was rastering over an area of 10x16 mm^2 on the irradiation stage. The irradiation temperature was monitored through three thermocouples and controlled at 200±20°C. The rate of atomic displacement damage is estimated to be approximately 1.0 $x10^{-5}$ dpa/s using STRIM2006 calculation with the default displacement energy of 25 eV [4]. The irradiation target

chamber and the irradiation stage are shown in Figure 1. Disc samples of three DU alloys were irradiated to doses of 0.1, 1.0, and 3.0 dpa. Microstructural analysis for both the unirradiated and the irradiated DU alloys will be performed using transmission electron microscopy (TEM). The results of TEM analysis are not available for this paper.

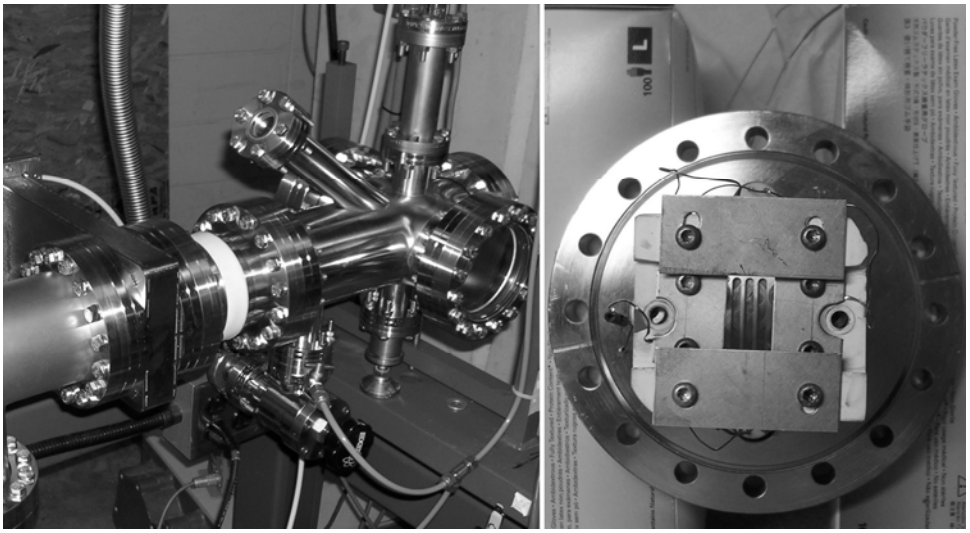

Figure 1. Irradiation chamber and stage for proton irradiation of DU alloys.

3. Results and Discussion

Secondary electron images of the Sample A microstructure are presented in Figure 2. Two phases comprise the alloy microstructure: a $U(Al,Si)_3$ phase and an Al phase. The $U(Al,Si)_3$ phase has an approximate composition in at% of: U_{27}-Al_{60}-Si_{13} (±1 at%).

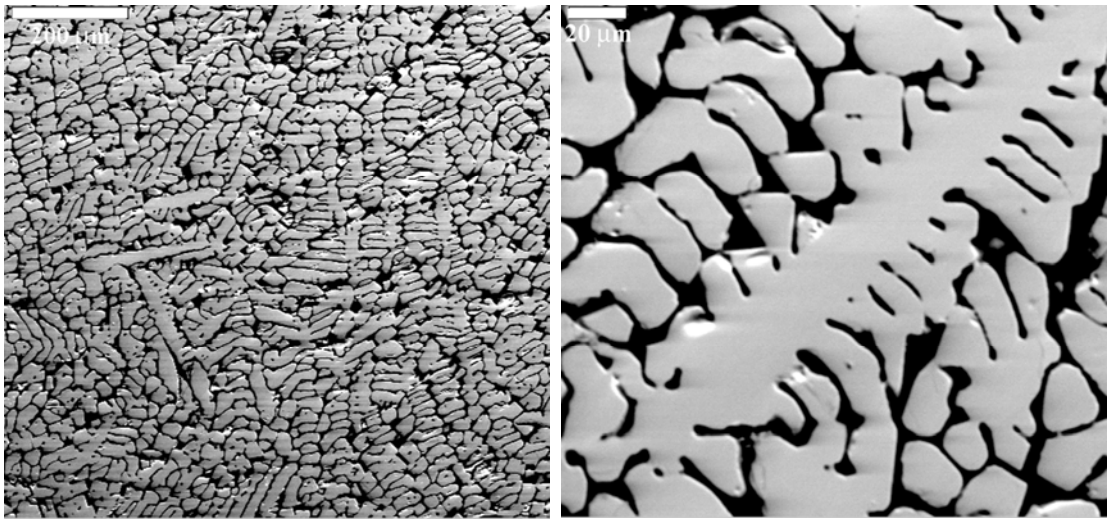

Figure 2. Secondary electron images at low (left) and high (right) magnification of the microstructure observed for alloy A. The black phase is Al, and the bright contrast phase is a $U(Si,Al)_3$ phase.

Secondary electron images of the Sample B microstructure are presented in Figure 3. Four phases comprise the alloy microstructure: UMo_2Al_{20}, $U_6Mo_4Al_{43}$, UAl_4, and Al. The approximate compositions for the UMo_2Al_{20}, $U_6Mo_4Al_{43}$, and UAl_4 in at% are: Al_{88}-Mo_7-U_5, Al_{79}-Mo_9-U_{12}, and Al_{78}-U_{22}, respectively.

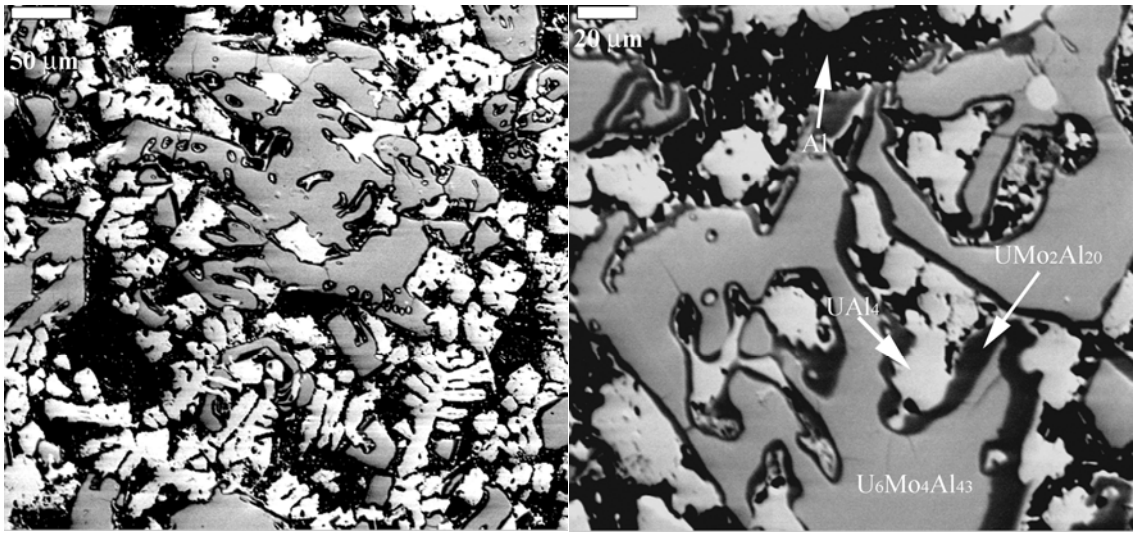

Figure 3. Secondary electron images at low (left) and high (right) magnification of the microstructure observed for Sample B.

Secondary electron images of the Sample D microstructure are presented in Figure 4. Three phases comprise the alloy microstructure: $(U,Mo)(Si,Al)_3$, UMo_2Al_{20}, and Al. The approximate compositions for the $(U,Mo)(Si,Al)_3$ and UMo_2Al_{20} in at% are: Al_{49}-Si_{19}-Mo_3-U_{27} and Al_{86}-Mo_8-U_6 respectively.

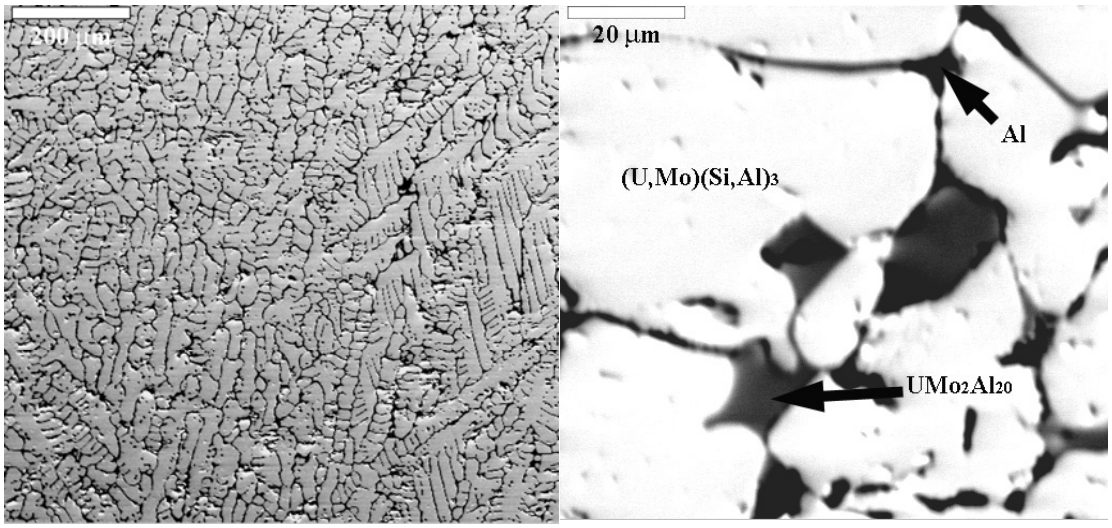

Figure 4. Secondary electron images at low (left) and high (right) magnification of the microstructure observed for Sample D.

For proton irradiation, the 0.1 dpa irradiation proceeded smoothly and was completed in approximately 3 hours. There is no problem for alloy "A" at 1 and 3 dpa. Some of the alloy "B" discs showed slight melting for 1 dpa and 3 dpa irradiation. The irradiation for alloy "D" discs at 1 dpa showed slight discoloration and crack development. The problem for alloy "D" discs becomes worse at 3 dpa. It is likely that the problem developed for the irradiated "D" alloy is associated with the microstructural changes which will be revealed through TEM analysis.

4. Conclusion

Three alloys have been successfully cast, homogenized, and characterized using SEM/EDS/WDS analysis. These alloys will work perfectly for their intended purpose as ion beam irradiation samples. Sample A contains only the $U(Al,Si)_3$ phase, besides some residual Al, which is the phase of interest to be studied. Sample B has U-Mo-Al phases that are assumed to influence the irradiation behaviour in the current versions of RERTR U-Mo dispersion fuels, and will be of interest as well. Finally, Sample D not only contains a quaternary phase that is of interest, i.e. the $(U,Mo)(Si,Al)_3$ phase, but it also contains one of the ternary U-Mo-Al phases (UMo_2Al_{20}) that is suspected to exhibit poor performance during irradiation. This will allow for a direct comparison, within one sample, of a phase that will hopefully exhibit favourable irradiation performance with a phase that is suspected to exhibit poor performance.

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