MICROSTRUCTURE CHARACTERIZATION OF RERTR FUEL

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ABSTRACT

A variety of phases have the potential to develop in the irradiated fuels for the reduced enrichment research test reactor (RERTR) program. To study the radiation stability of these potential phases, three depleted uranium alloys were cast. The phases of interest were identified including $U(Si,Al)_3$, $(U,Mo)(Si,Al)_3$, UMo_2Al_{20}, UAl_4, and $U_6Mo_4Al_{43}$. These alloys were irradiated with 2.6 MeV protons at 200ºC up to 3.0 dpa. The microstructure is characterized using SEM and TEM. Microstructural characterization for an archive dispersion fuel plate (U-7Mo fuel particles in Al-2%Si cladding) was also carried out. TEM sample preparation for the irradiated dispersion fuel has been developed.

1 Introduction

The RERTR program is to develop new low enrichment uranium fuels to replace the high enrichment fuels for research and test reactors worldwide. This is a collaborated effort among many countries to ensure a safe and secured use of research and test reactors to meet non-proliferation requirements. An important part of the fuel development program is to study fuel performance under irradiation. Radiation stability of the potential fuel-cladding interaction product may play an important role in fuel performance. Microstructural characterization using transmission electron microscopy (TEM) is capable to provide key information on the microstructure (crystal structure, precipitates, defects, various interfaces, and microchemistry, etc) with resolution down to the nanometer.

A variety of phases have the potential to develop in irradiated RERTR fuels as a result of fuel/cladding interaction. To study the effects of radiation on the potential fuel/cladding interaction product, three depleted uranium (DU) alloys are arc-cast with the compositions of 67U-5Si-28Al (alloy-A), 48U-5Mo-47Al (alloy-B), and 69U-4Mo-20Al-7Si (alloy-D) at Idaho National Laboratory (INL). 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 interdiffusion process, then it is likely that a $(U,Mo)(Si,Al)_3$ phase will form and remain stable under irradiation, much 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. In addition to DU alloys, an archive dispersion fuel plate (U-7Mo in Al-2%Si alloy) was also analyzed using TEM. This work will provide an important reference to the upcoming TEM analysis of the irradiated dispersion fuel plate.

2 Experiments and Results

2.1 Characterization of Proton Irradiated Du Alloys

The SEM images of microstructure for three DU alloys after heat treatment at 500ºC for 200 hours are shown in Fig. 1. TEM disc samples (3.0 mm diameter, ~300 μm thick) are prepared from DU alloys. The SRIM code was used for the calculation of displacements per atom

(dpa) for proton irradiation [4]. These discs were irradiated with 2.6 MeV protons at 200ºC to doses of 0.1, 1.0, and 3.0 dpa at the University of Wisconsin. Proton irradiation produced a uniform damaged layer approximately 25-30 μm depending on the phase composition. A schematic of TEM sample preparation for proton irradiated DU alloys is shown in Fig. 2. The irradiated TEM discs were mechanically wet polished from the unirradiated side down to a thickness of ~120 μm, followed by electrical jet-polishing to perforation, and additional ion polishing (4.0 KV Ar ions at incident angles 7º- 9º) to produce large thin areas around the perforation. A JEOL2010 transmission electron microscope was used for microstructure analysis. The composition and crystal structure for phases identified in these alloys are listed in Table 1.

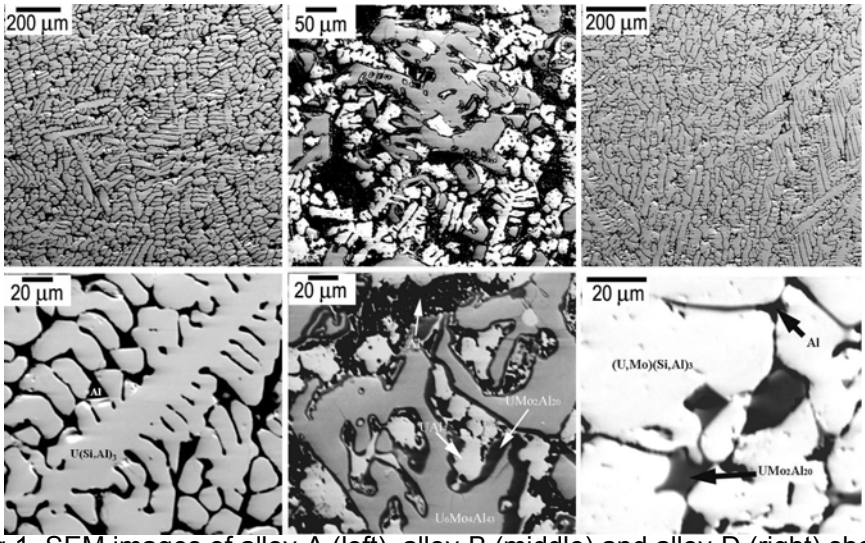

Fig 1. SEM images of alloy-A (left), alloy-B (middle) and alloy-D (right) show low magnification (top) and the high magnification (bottom) views.

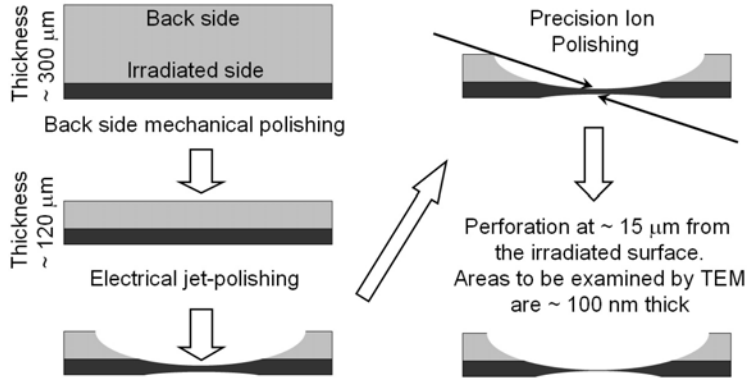

Fig 2. Schematic of TEM sample preparation for proton irradiated DU alloys.

Phase	Alloy	Crystal Structural Information
$U(Si,Al)_3$	A	Cubic, $L1_2$ ordered Cu_3Au type, a=b=c=0.426 nm, α=β=γ=90°, Pearson symbol: cP4, Space group: 221
$(U,Mo)(Si,Al)_3$	D	Cubic, $L1_2$ ordered Cu_3Au type, a=b=c=0.426 nm, α=β=γ=90°, Pearson symbol: cP4, Space group: 221
UMo_2Al_{20}	B, D	Cubic, a=b=c=1.4506 nm, α=β=γ=90°, Pearson symbol: cF184, Space group: 227
$U_6Mo_4Al_{43}$	B	Hexagonal, a=b=1.0966 nm, c=1.7690 nm, c/a=1.613, α=β=90º, γ=120°, Pearson symbol: hp106, Space group: 193
UAl_4	B	Body-centre orthrohomic, a=0.6270 nm, b=1.3710 nm, c=0.4410 nm, α=β=γ=90°, Pearson symbol: oI20, Space group: 74

Table 1: Crystal structural information for the phases identified in three DU alloys.

The [001] zone diffraction pattern for $(U,Mo)(Si,Al)_3$ and $U(Si,Al)_3$, shown in Fig. 3, indicates an ordering at 8 times of the lattice spacing (3.408 nm) in the $U(Si,Al)_3$ phase. No precipitate

was found in these two phases. There is no discernible change in these zone patterns after proton irradiation to 3.0 dpa compared to the unirradiated microstructure. Radiation induced amorphization was not found in any of the five phases. For the UMo_2Al_{20} phase, a bright field image of high density stacking faults and a high resolution lattice fringe image of the {111} plane projection in the unirradiated condition are shown in Fig. 4. High density stacking faults are only found in UMo_2Al_{20} in alloy-B. The same phase in alloy-D shows scattered stacking faults at much lower density. Dislocation loop was not found in UMo_2Al_{20} phase irradiated up to 3 dpa.

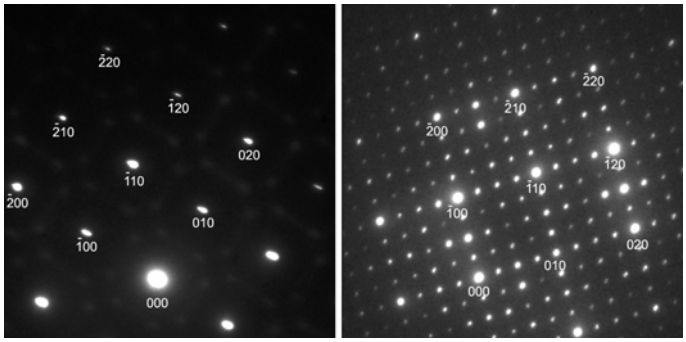

Fig 3. The [001] zone diffraction patterns for $(U,Mo)(Si,Al)_3$ (left) and $U(Si,Al)_3$ (right). Both show the $L1_2$ ordered structure (indexed spots) with well defined extra fine spots found in $U(Si,Al)_3$.

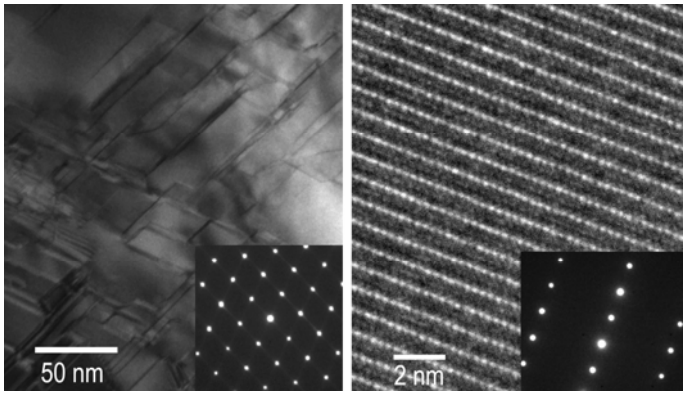

Fig 4. Bright field image of UMo_2Al_{20} phase near zone [011] (left) shows the high density of stacking faults and a high-resolution lattice fringe image at zone [123] (right) shows {111} plane projection.

2.2 Development of TEM analysis for Archive Dispersion Fuel

Microstructural characterization of an archive dispersion fuel was carried out using TEM. A schematic of TEM sample preparation for the dispersion fuel is shown in Fig. 5. A 1.0 mm diameter punching from the dispersion fuel plate is glued to a 3 mm diameter Mo ring using epoxy. The Mo ring was then mechanically polished to remove the Al cladding from both sides, resulting in a thickness of ~120 μm. An electrical jet-polishing from both sides for a fixed amount of time reduces the thickness in the centre to approximately 10-20 μm. The TEM disc sample was then ion polished with 4.0 KV Ar+ ions at ±7° incident angle to perforation. The resultant TEM sample is proven to be adequate for microstructural analysis with a 200 KV electron beam.

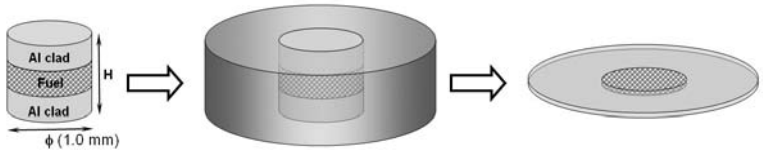

Fig 5. Schematic of TEM sample preparation for RERTR dispersion fuel.

For the TEM characterization of reactor-irradiated RERTR dispersion fuels in the near future, the 1.0 mm diameter fuel punching will be done in an INL Hot Fuel Examination Facility (HFEF). The assembly of fuel punching into an Mo ring and the following mechanical polishing will be performed in a glove box at INL's Electron Microscopy Laboratory (EML). Both facilities are shown in Fig. 6. The mechanical polishing for TEM disc of the irradiated dispersion fuel will be using a local radiation shielding with two tungsten plates and a tungsten sample mounting block, as shown in Fig. 7. The sample surface inspection will be done by using a palm size portable digital microscope in the glove box.

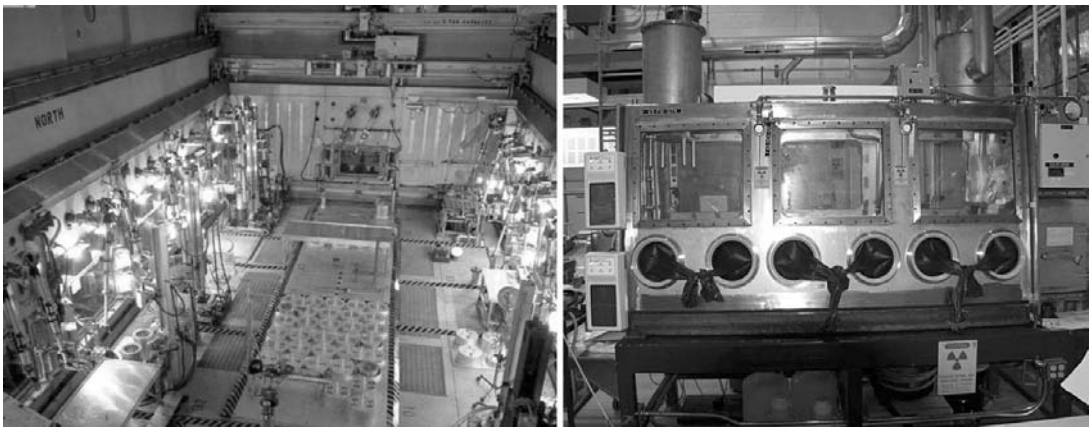

Fig 6. The HFEF (left) and the EML glove box (right) at Idaho National Laboratory.

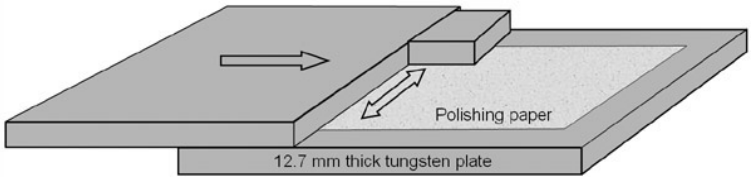

Fig 7. Schematic of mechanical polishing with local shielding for TEM sample preparation in a glove box for irradiated RERTR dispersion fuel.

3 Discussion

Three DU alloys consist of five intermetallic phases. Proton irradiation at 200°C up to 3.0 dpa did not produce any significant change in the irradiated microstructure characterized using TEM. This is likely due to the relatively low irradiation temperature and doses which inhabit the development of various defects, such as dislocation loops and micro cavities. The absence of amorphization at these irradiation conditions indicates a relative stable microstructure to the displacement damage from proton irradiation. Considering the radiation damage process for RERTR fuel in the reactor is driven by both fast neutrons and fission product, the latter may be more important in governing the microstructural development in fuels. This will be tested by upcoming heavy ion irradiation with Kr ions at INL. TEM characterization of the reactor-irradiated dispersion fuel will be crucial to reveal the details of microstructural development and their impact on fuel performance.

4 Conclusions

The TEM characterization shows no significant changes in the microstructures of five phases irradiated with protons at 200°C up to 3.0 dpa. Theses five phases are stable to the atomic displacement damage up to 3.0 dpa.

Acknowledgements

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