U-Mo/Al Alloys Diffusion Couples: Fuel/Cladding Interactions

E. Perez¹, Y.H. Sohn¹, D.D.Keiser²

¹Advanced Materials Processing and Analysis Center
Department of Mechanical, Materials and Aerospace Engineering
University of Central Florida, Orlando, Fl
²Idaho National Laboratory, Idaho Falls, ID

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Objectives

To examine the growth and composition of the intermetallic compound layers that develop in U-Mo/Al system through diffusion couple experiments.

Examine the phase development of the Al-rich UMoAl ternary systems based on cast alloys with nominal compositions of 85.7Al-11.44U-2.86Mo, 87.5Al-10U-2.5Mo (at%).
U-Mo dispersion/monolithic fuels in Al-alloys matrix are being developed to fulfill the requirements of low enriched uranium in research reactors under the Reduced Enrichment for Research Test Reactors (RERTR) program.

Experiments can be loaded into the reactor and exposed to high radiation levels. Higher than typical power generation reactors.
Dispersion and monolithic fuel system designs are being considered.

- UMo particles are dispersed in an Al matrix.
- UMo foil cladding between Al-alloy sheets.

These alloy systems show promising results due to their high uranium density.
Dispersion and monolithic fuels in research test reactors suffer from detrimental interactions between the UMo fuel and Al matrix that lead to premature failure of the fuel systems.

- Interaction zones develop complex multiphase microstructures.
- Volumetric expansion takes place due to difference in the densities of the intermetallic phases.
- The intermetallic phases have lower thermal conductivities than the fuel or the Al-matrix.
The interdiffusion zone developed an average non-fluctuating composition through most of the interaction zone.

The U-rich side of the interdiffusion zone developed an area with varying microstructure and composition.

*Intermetallic average composition
The thickness of intermetallic layer increases with increasing concentration of Mo in the UMo alloy.
The thickness of intermetallic layer in the couples with Al-Si alloys is an order of magnitude smaller than that with pure Al.
Glove Box

Vacuum System

Lindberg/Blue 3-Zone horizontal tube furnace

Quartz Capsules

TEM FEI Tecnai F30

FIB FEI 200 TEM

SEM Hitachi 3500N
Experimental Details

- Alloys with nominal compositions 85.7Al-11.44U-2.86Mo and 87.5Al-10U-2.5Mo (at.%) were by arc melting of high purity Al, U and Mo.

- The alloys were re-melted three times to ensure homogenization.

- The alloys were then annealed at 500°C for 200 hours.

Analysis of phase constituents and microstructure:

- X-Ray Diffraction ($\theta$-2$\theta$)

- Scanning Electron Microscopy:
  - Backscatter Electron Microscopy
  - Energy Dispersive Spectroscopy

- Transmission Electron Microscopy and Scanning TEM:
  - Site-Specific Specimen Preparation via Focused Ion Beam In-Situ Lift-Out (FIB-INLO)
  - High Angle Annular Dark Field (HAADF) Imaging
  - Selected Area Diffraction
Experimental Details

Solid-Solid diffusion couples were assembled using U-7Mo, U-10Mo and U-12Mo with pure Al (99.999%), and were heat-treated in Ar-atmosphere at 600°C for 24 hours.

Diffusion couple alloys were sectioned, polished and assembled under a controlled Ar atmosphere in a glove box.

Prior to assembly, the Al was treated with concentrated nitric acid to dissolve the Al$_2$O$_3$ surface layer.

Diffusion couples were encapsulated in quartz capsule in Ar atmosphere after Argon flush for heat treatment. Ta foil was placed in the capsule as an oxygen trap.

Diffusion anneal performed using a Lindberg/Blue 3-Zone horizontal tube furnace.

Diffusion microstructures structures examined by:

- Scanning Electron Microscopy:
  - Backscatter Electron Microscopy
  - Energy Dispersive Spectroscopy

- Transmission Electron Microscopy and Scanning TEM:
  - Site-Specific Specimen Preparation via Focused Ion Beam In-Situ Lift-Out (FIB-INLO)
  - High Angle Annular Dark Field (HAADF) Imaging
  - Selected Area Diffraction
Cast 85.7Al-11.44U-2.86Mo and 87.5Al-10U-2.5Mo (at%) Ternary Alloys

Four phases were observed in bulk alloys via SEM/BEI: Phase equilibrium was not obtained, or cooling effects may be present.

A fifth phase (UAi₃) was found in very small quantities in the 85.7Al-11.44U-2.86Mo alloy.
TEM Analysis of the 85.7Al-11.44U-2.86Mo Alloy

In the bulk of the alloys, the TEM electron diffraction showed the presence of:

- Al solid solution
  - cF4
- UAl4 with little solubility for Mo
  - ol20 Imma
- U6Mo4Al43
  - hP106 P63/mcm
- UMo2Al20
  - cF184 Fd-3m
TEM electron diffraction pattern showed that the fifth phase found in very small quantities was the UAl$_3$ phase.
TEM Analysis of the 87.5Al-10U-2.5Mo Alloy

The data from the 85.7Al-11.44U-2.86Mo alloy was used to identify the phases on this alloy.

- Al solid solution
- UAl$_4$ with little solubility for Mo
- UMo$_2$Al$_{20}$
- U$_4$Mo$_6$Al$_{43}$

The UAl$_3$ Phase was not observed in this alloy.
XRD Analysis of UMoAl alloys

The phases observed by XRD in 85.7Al-11.44U-2.86Mo and 87.5Al-10U-2.5Mo alloys were:

- Al, UAl₄, U₄Mo₆Al₄₃ and UMo₂Al₂₀.
- The UAl₃ phase volume fraction in the alloys was very small and not detectable by XRD.

XRD of the alloys confirmed the presence of the phases found by TEM electron diffraction.
Diffusion Couples Annealed at 600°C for 24 Hours

Layer Thickness: 265μm

U-7Mo vs. Al 600°C 24hr 70X BSE

Layer Thickness: 542μm

U-10Mo vs. Al 600°C 24hr 70X BSE

Layer Thickness: 352μm

U-12Mo vs. Al 600°C 24hr 70X BSE
**Diffusion Couples Typical Microstructural Development**

**Typical Microstructure:**

- Two-Phase regions composed of intermetallic phases
- High U–phase at the UMo/Intermetallic interface
Electron diffraction patterns have been collected on selected areas of the U-10Mo vs. Al diffusion couple annealed at 600°C for 24 hours.
EDX compositional analysis identifies a U-solid solution depleted of Mo.
Based on the development of ternary UMoAl alloys, the phases found by EDX may be the $\text{UMo}_2\text{Al}_{20}$ and the $\text{U}_6\text{Mo}_4\text{Al}_{43}$. 
U-10Mo vs. Al 600C 24 hr Diffusion Couple
Selected Area Electron Diffraction
Due to the small grain size of the developed phases, polycrystalline electron diffraction patterns were collected on the center of the interaction zone.
Summary

Alloys with nominal compositions 85.7Al-11.44U-2.86Mo and 87.5Al-10U-2.5Mo (at.%) have been examined using XRD, SEM/EDS and TEM/STEM for identification of phase constituents and analysis of the microstructures.

The fcc Al solid solution, cubic-UAl$_3$, orthorhombic-UAl$_4$, hexagonal-U$_6$Mo$_4$Al$_{43}$ and diamond cubic-UMo$_2$Al$_{20}$ phases were observed.

Diffusion Couples of U-7, U-10 and U-12Mo vs. Al were assembled and annealed at 600°C for 24 hours for detailed characterization of the phases that develop in the interaction zones.
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