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FROM PROMISE TO PROMINENCE
CELEBRATING 40 YEARS

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

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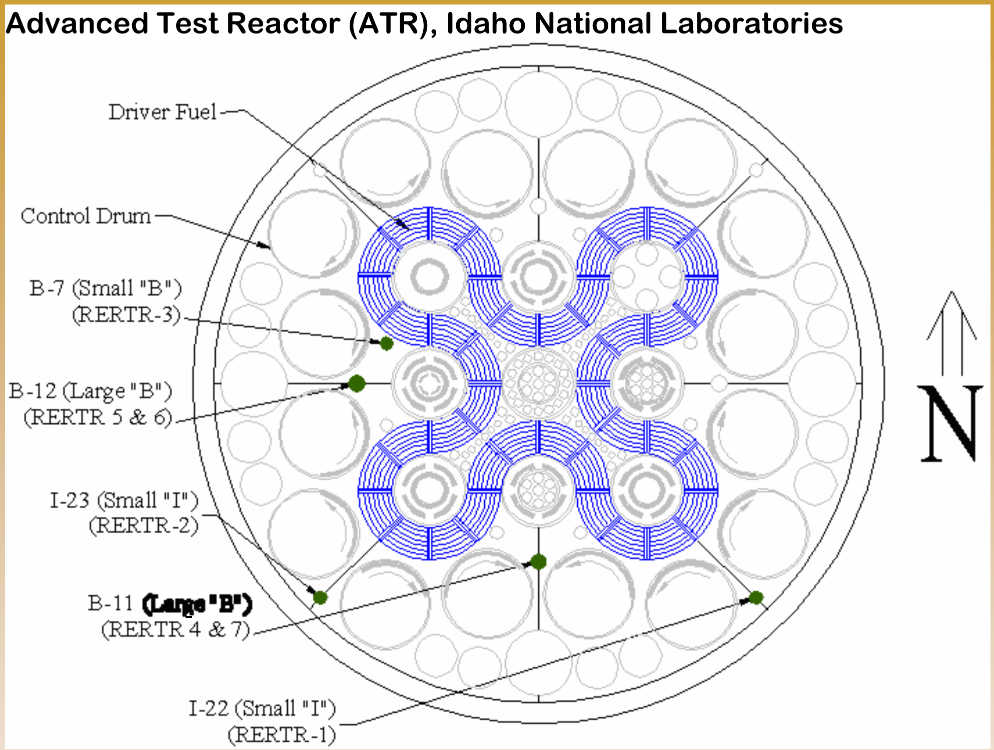
“Bridging the technology gap”

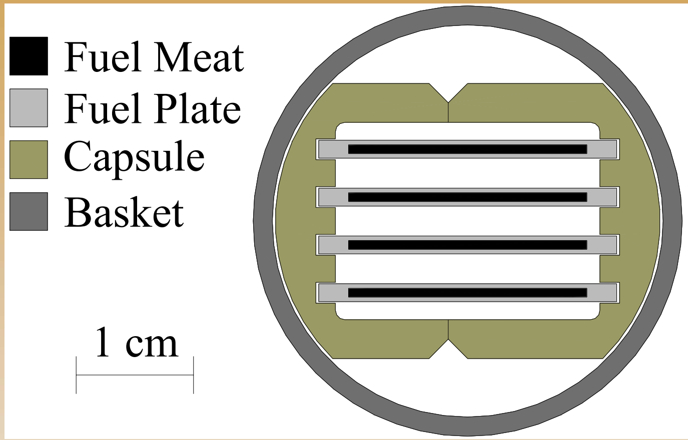
Objectives

- To examine the growth and composition of the intermetallic compound layers that develop in U-Mo/Al system though 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%).

Research Test Reactors

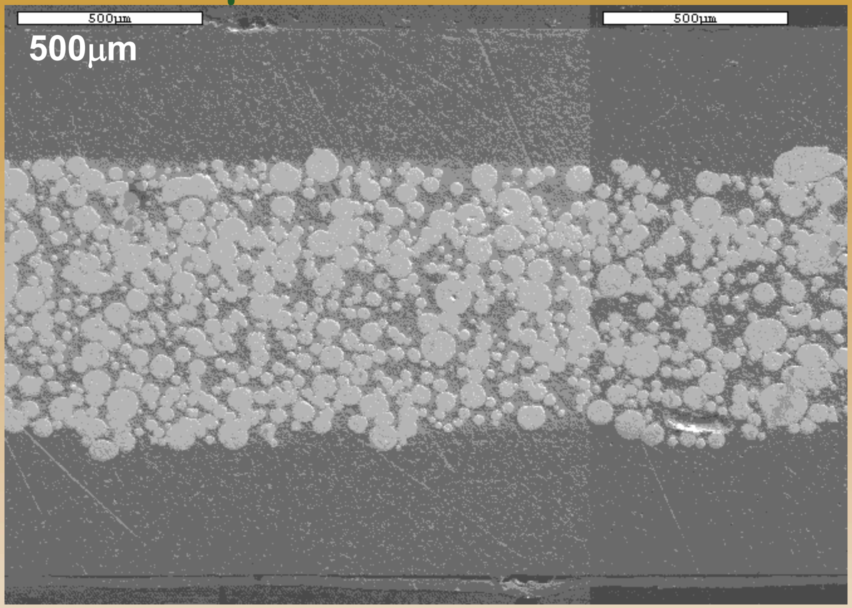




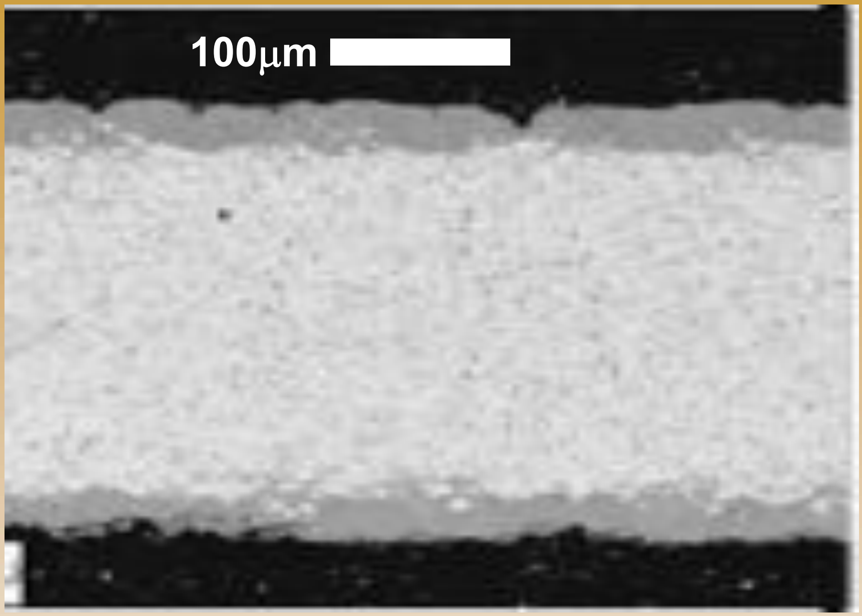
- **U-Mo dispersion/monolithic fuels in Al-alloys matrix are being developed to fulfill the requirements of low enriched uranium in research reactors under the Reduced Enrichment for Research Test Reactors (RERTR) program.**
  - **Experiments can be loaded into the reactor and exposed to high radiation levels. Higher than typical power generation reactors.**

Reactor Fuel Systems

Dispersion Fuel Plate



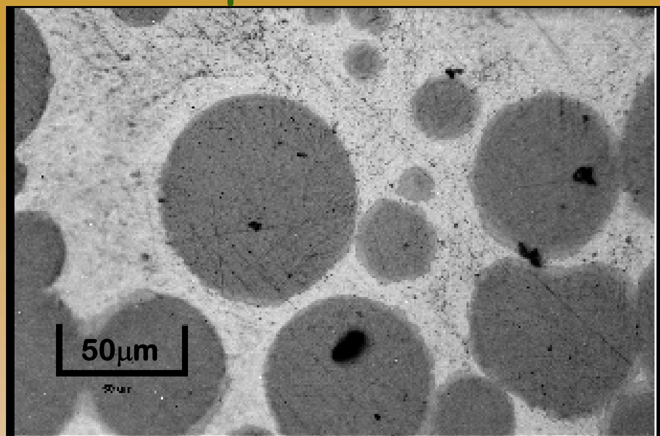
Monolithic Fuel Plate



- 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.

Motivation

Dispersion Fuels | **Monolithic Fuels**



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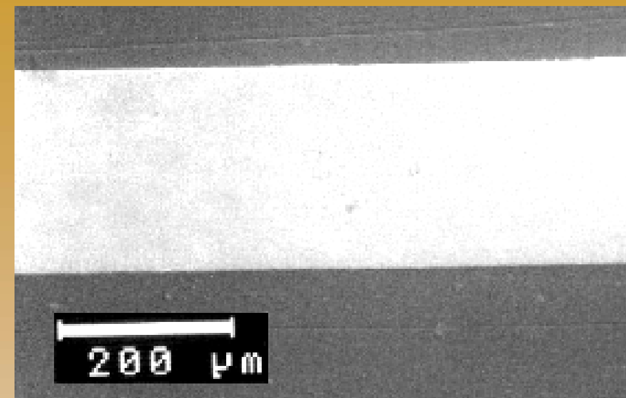

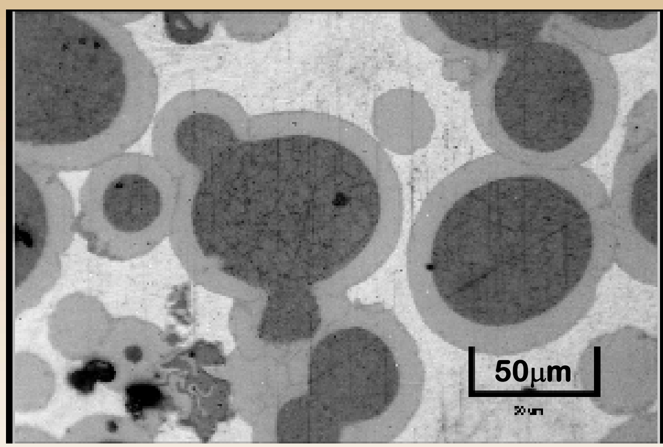

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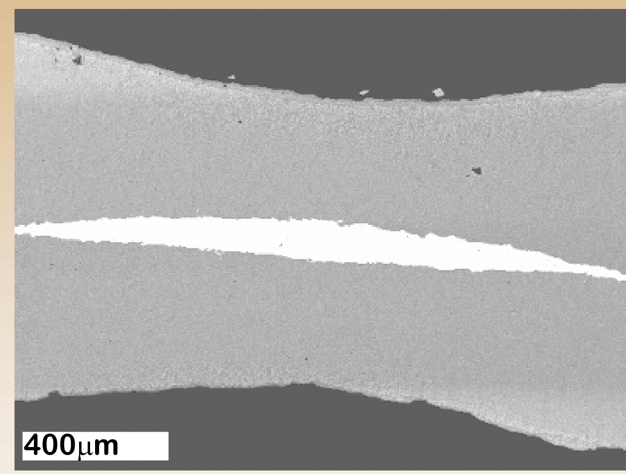
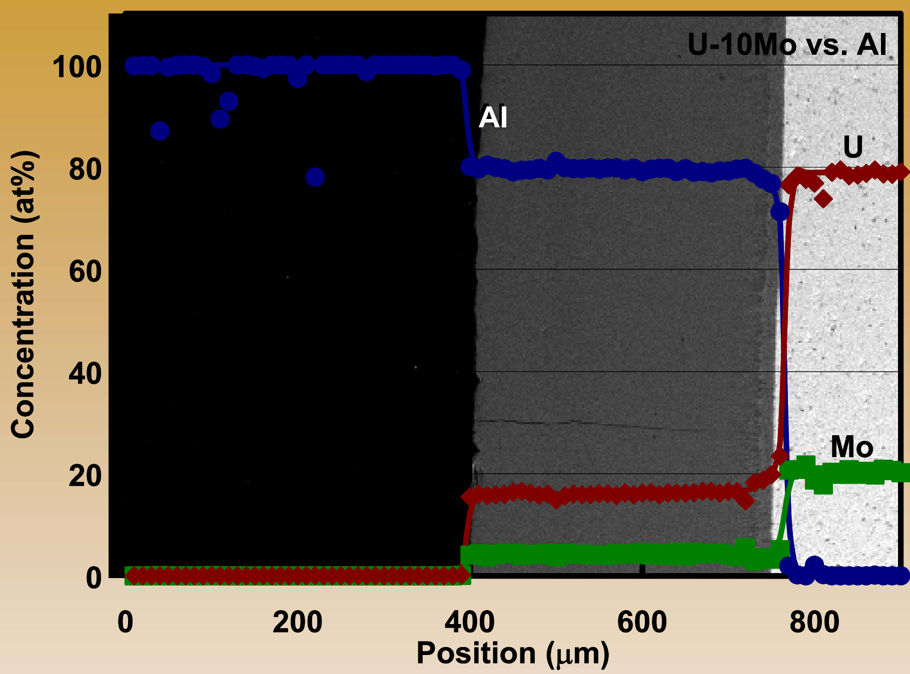
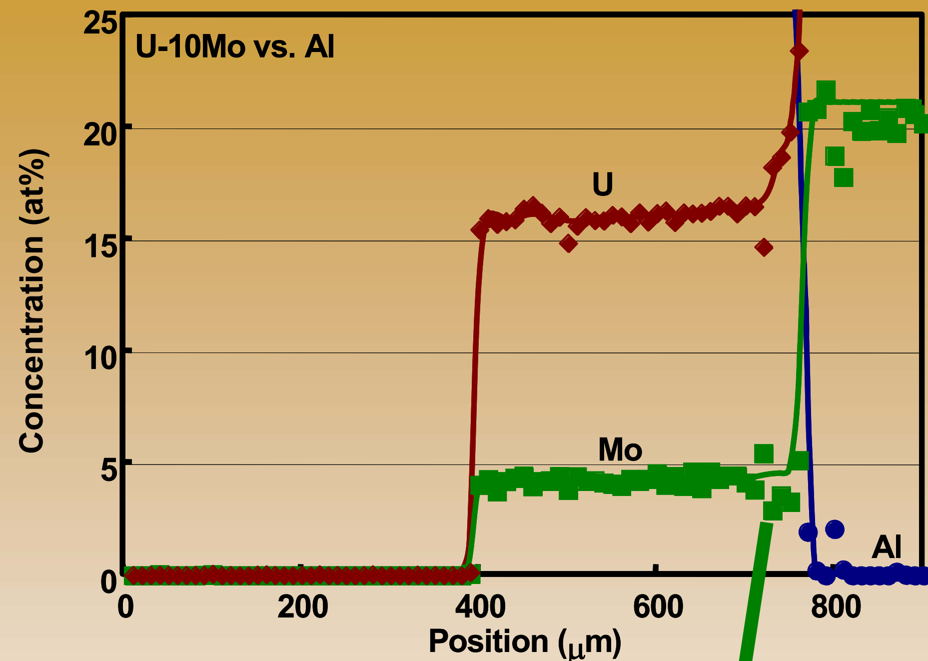


- **Dispersion and monolithic fuels in research test reactors suffer from detrimental interactions between the UMo fuel and Al matrix that lead to premature failure of the fuel systems.**
  - **Interaction zones develop complex multiphase microstructures.**
  - **Volumetric expansion takes place due to difference in the densities of the intermetallic phases.**
  - **The intermetallic phases have lower thermal conductivities than the fuel or the Al-matrix.**

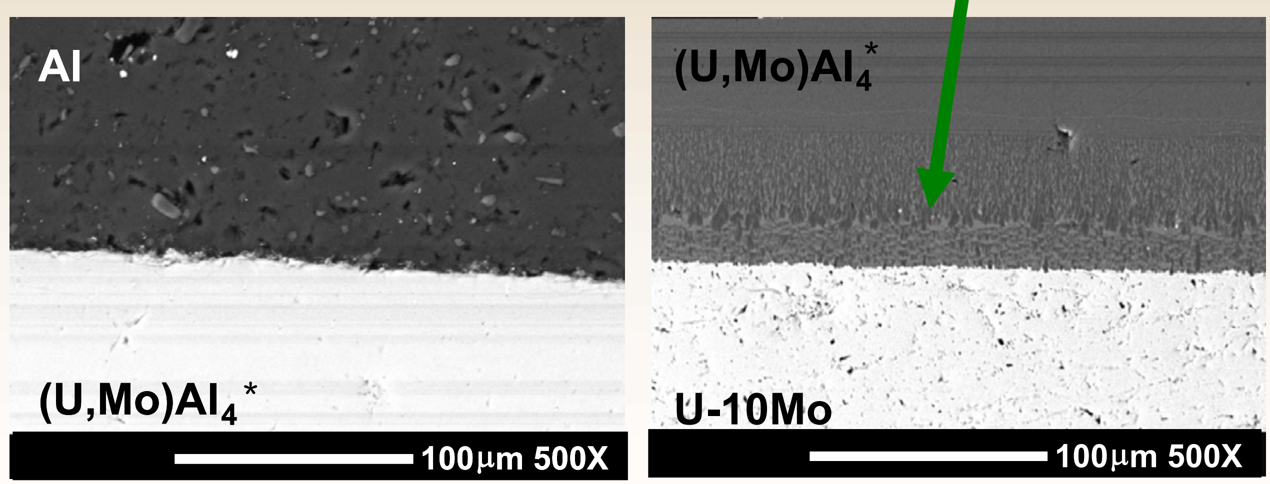
Review: U-10Mo vs. Al Diffusion Couple (600°C, 24hr)





- The interdiffusion zone developed an average non-fluctuating composition through most of the interaction zone.

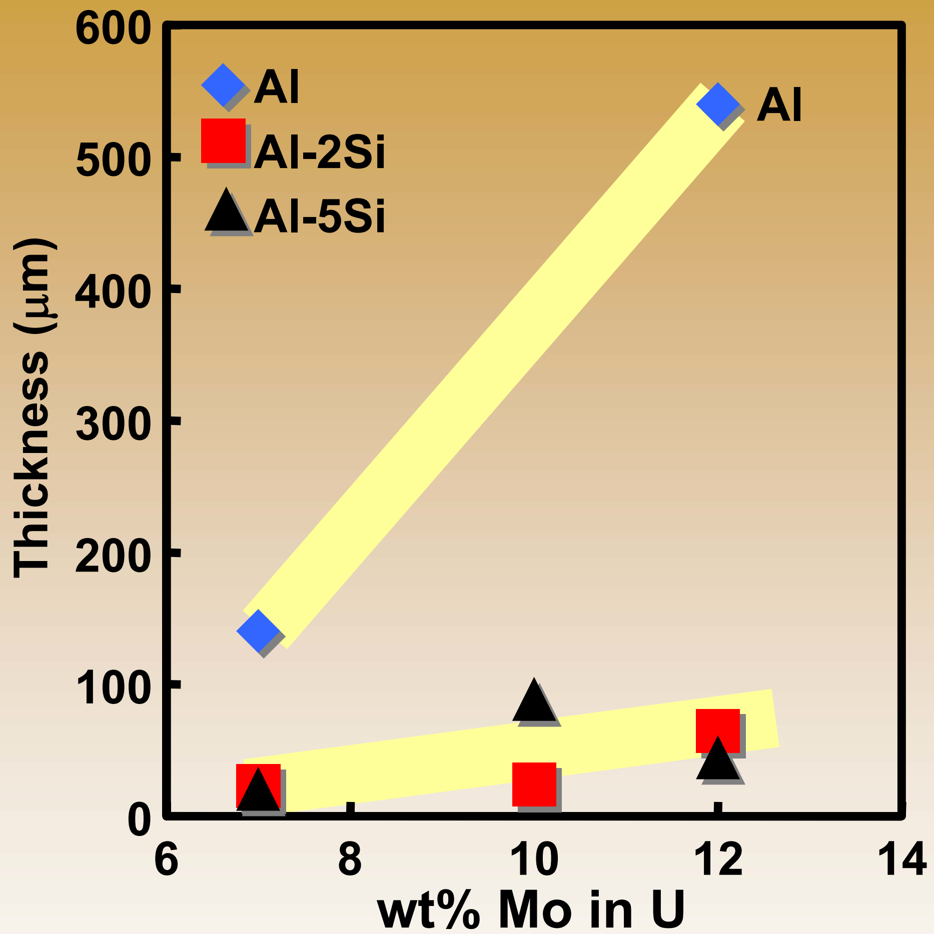
- The U-rich side of the interdiffusion zone developed an area with varying microstructure and composition.

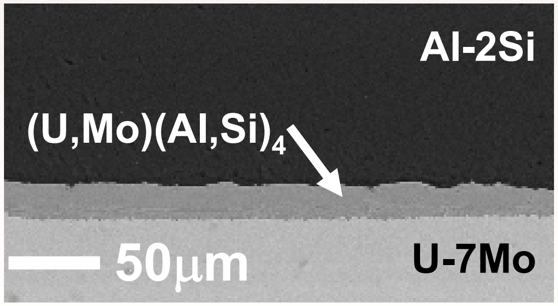


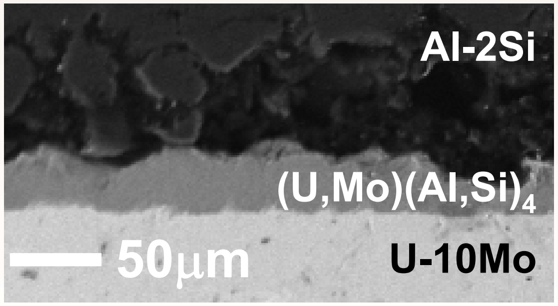
*Intermetallic average composition

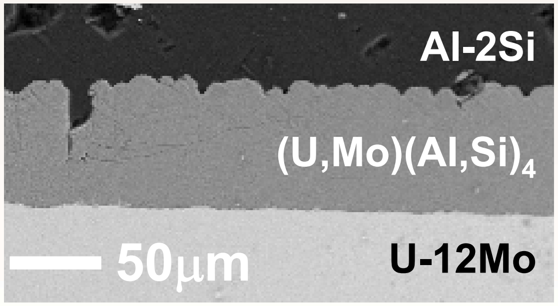
Review: Composition Dependent Growth Kinetics of Intermetallic Phases at 550°C

- The thickness of intermetallic layer increases with increasing concentration of Mo in the UMo alloy.

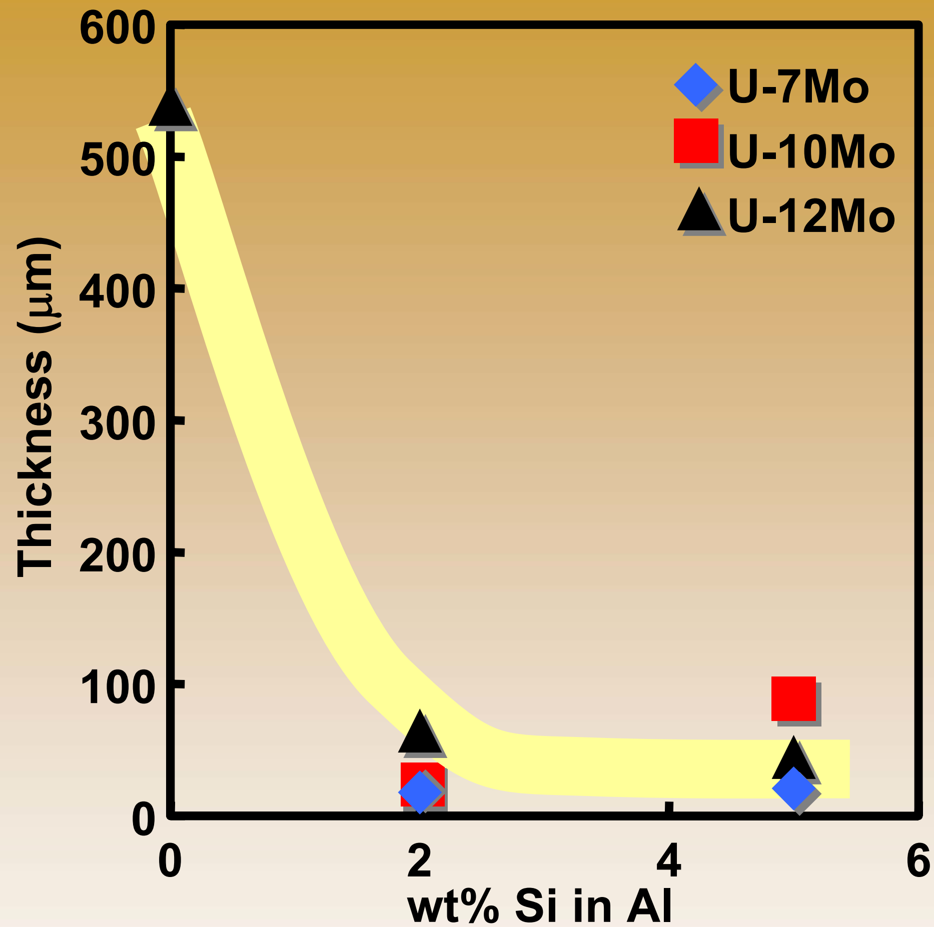


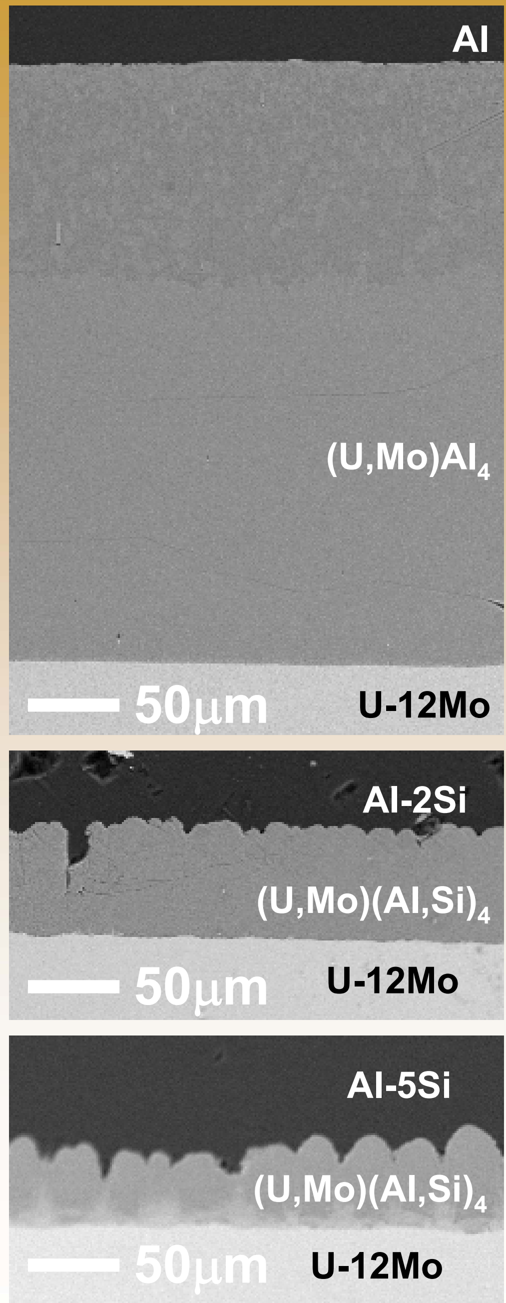






Review: Composition Dependent Growth Kinetics of Intermetallic Phases at 550°C





- The thickness of intermetallic layer in the couples with Al-Si alloys is an order of magnitude smaller than that with pure Al.

Experimental Details - Facilities


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 (θ-2θ)
  - 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.

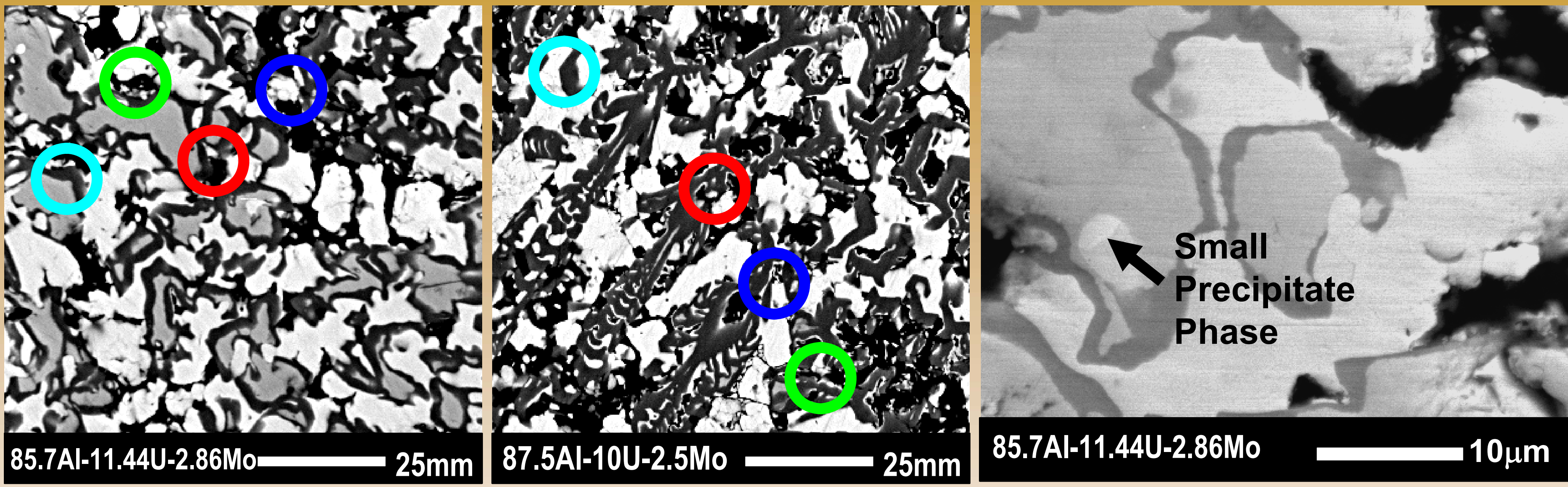
- Prior to assembly, the Al was treated with concentrated nitric acid to dissolve the Al_2O_3 surface layer.

- 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



1 **Black Phase** (Al Solid Sol.)

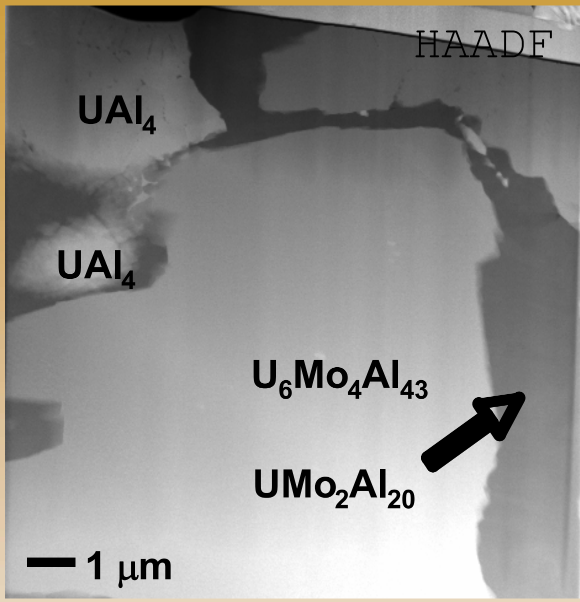
3 **Light Gray Phase** ($U_6Mo_4Al_{43}$)

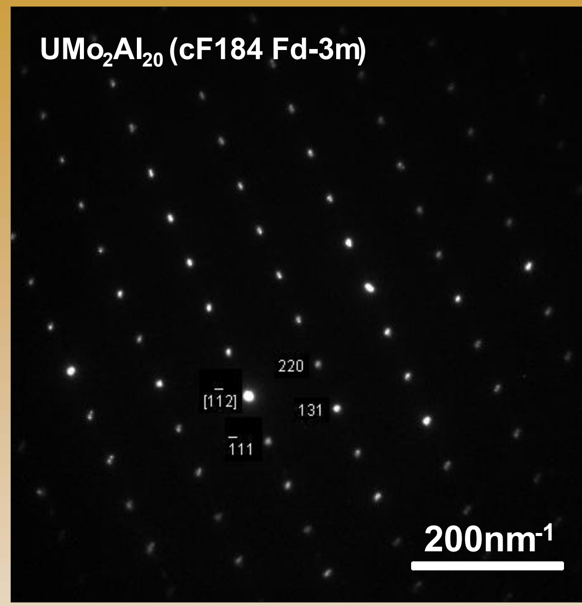
2 **Dark Gray Phase** (UMo_2Al_{20})

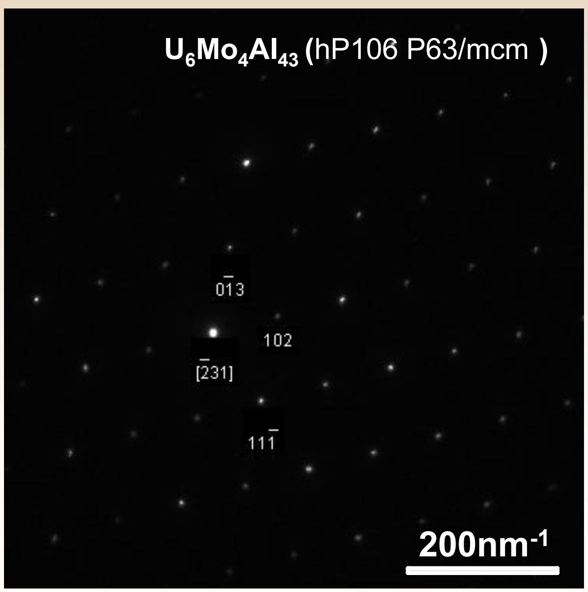
4 **White Phase** (UAl_4)

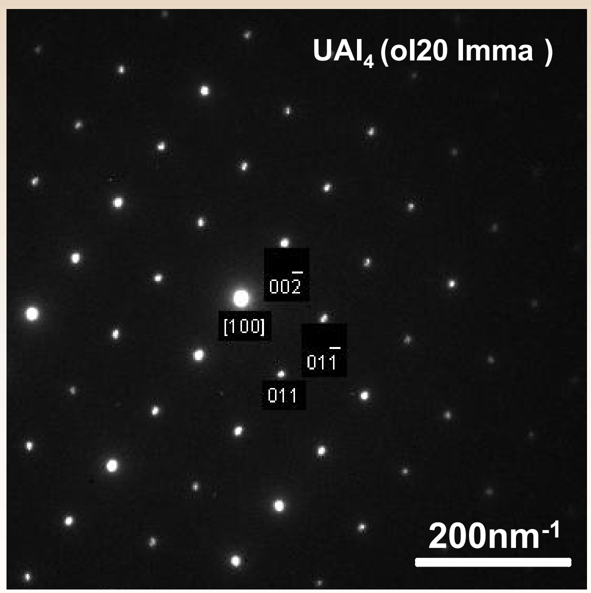
- **Four phases were observed in bulk alloys via SEM/BEI: Phase equilibrium was not obtained, or cooling effects may be present.**
- **A fifth phase (UAl_3) was found in very small quantities in the 85.7Al-11.44U-2.86Mo alloy.**

TEM Analysis of the 85.7Al-11.44U-2.86Mo Alloy





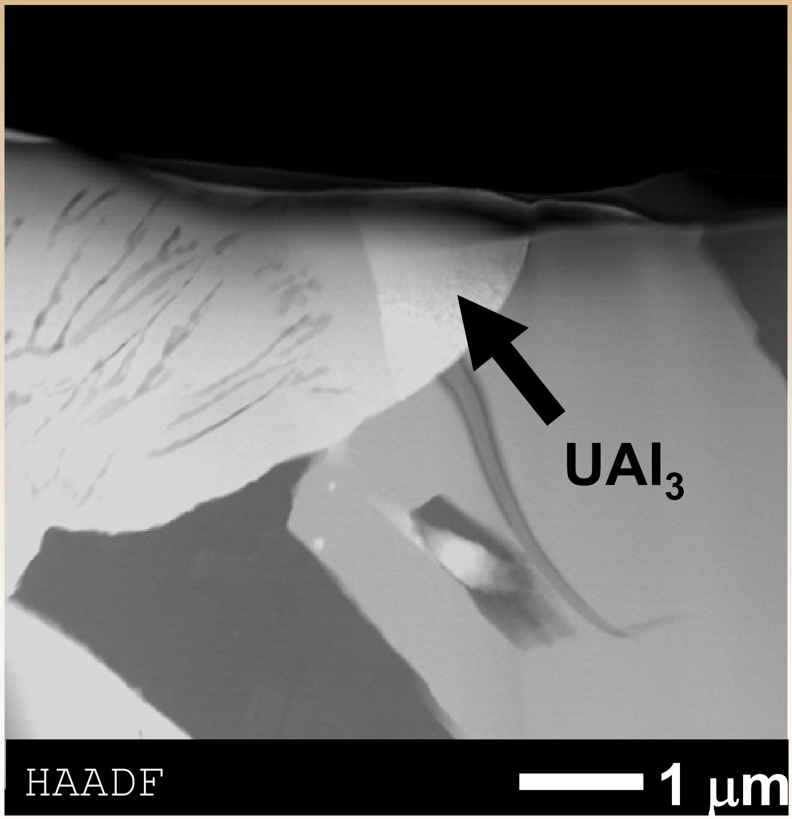


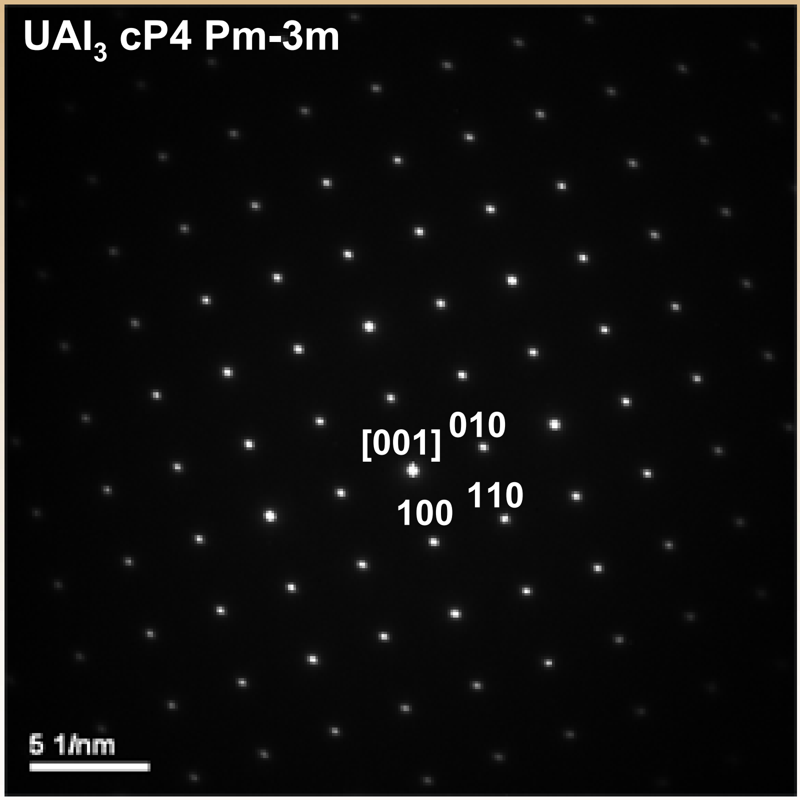


- In the bulk of the alloys, the TEM electron diffraction showed the presence of:
  - Al solid solution
    - cF4
  - UAl_4 with little solubility for Mo
    - oI20 Imma
  - $U_6Mo_4Al_{43}$
    - hP106 P63/mcm
  - UMo_2Al_{20}
    - cF184 Fd3m

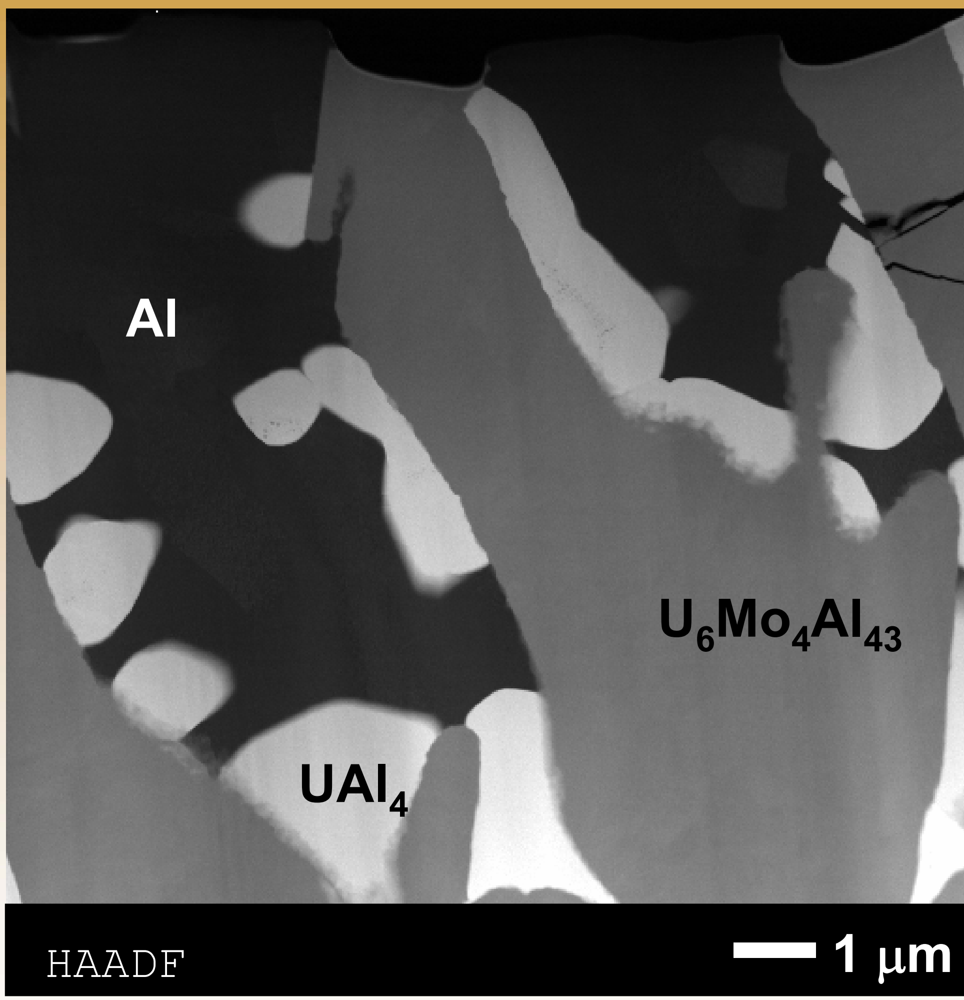
TEM Analysis of the 85.7Al-11.44U-2.86Mo Alloy

- **TEM electron diffraction pattern showed that the fifth phase found in very small quantities was the UAl_3 phase.**



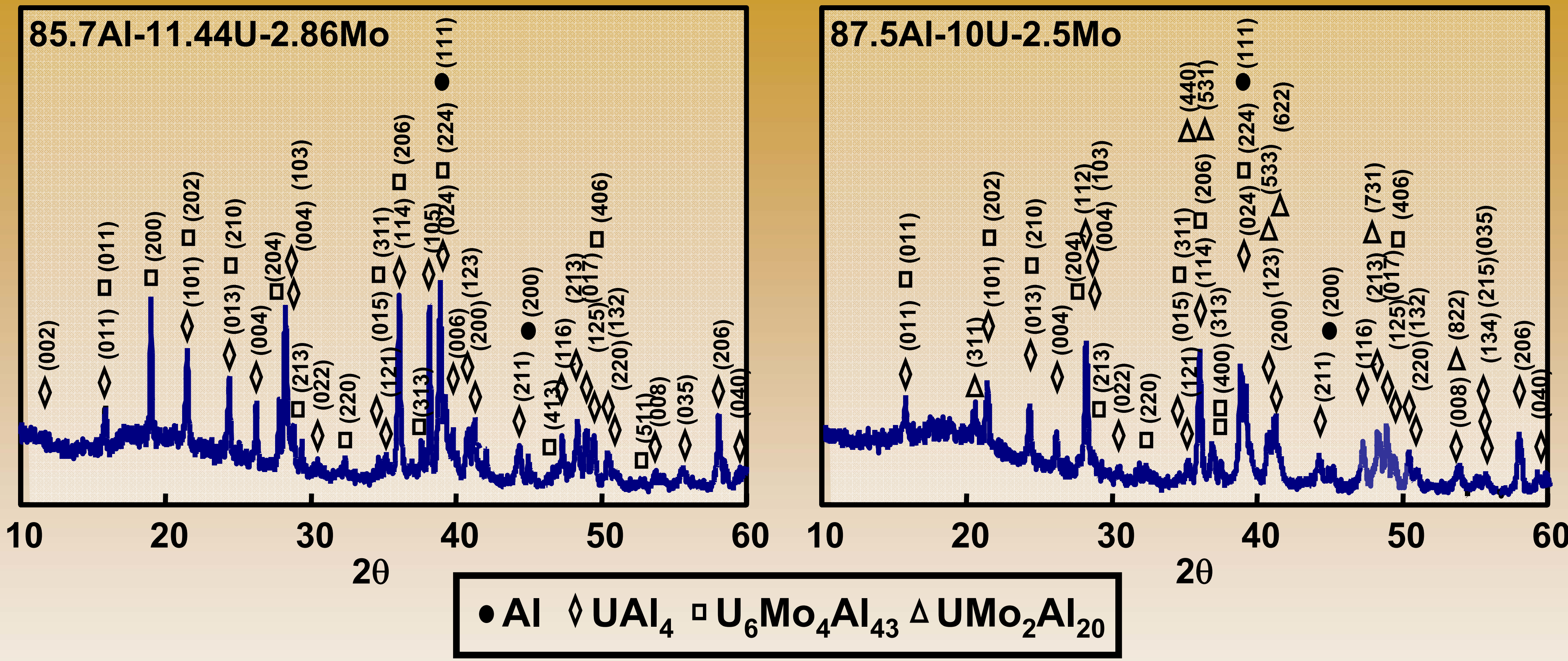


TEM Analysis of the 87.5Al-10U-2.5Mo Alloy



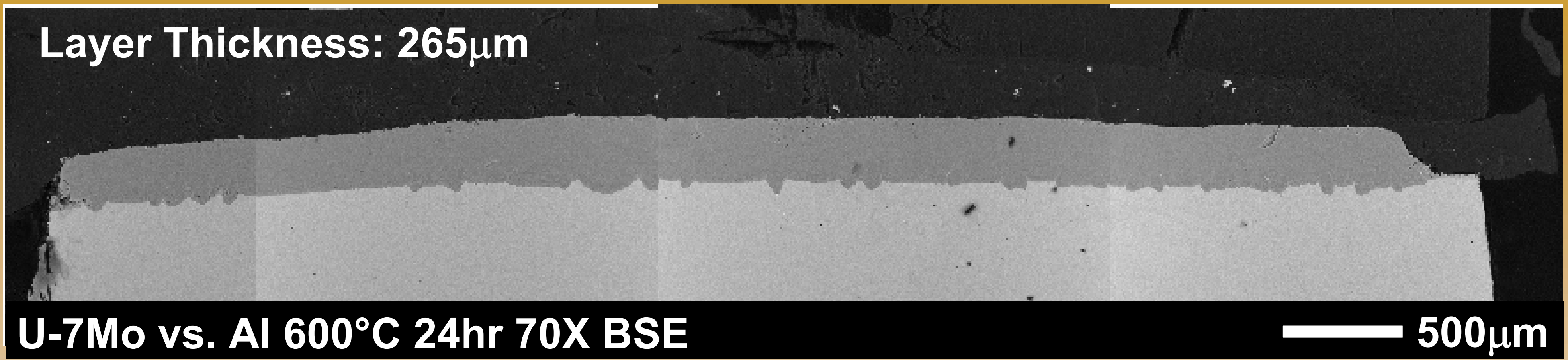
- The data from the 85.7Al-11.44U-2.86Mo alloy was used to identify the phases on this alloy.
  - Al solid solution
  - UAl_4 with little solubility for Mo
  - UMo_2Al_{20}
  - $U_4Mo_6Al_{43}$
- The UAl_3 Phase was not observed in this alloy.

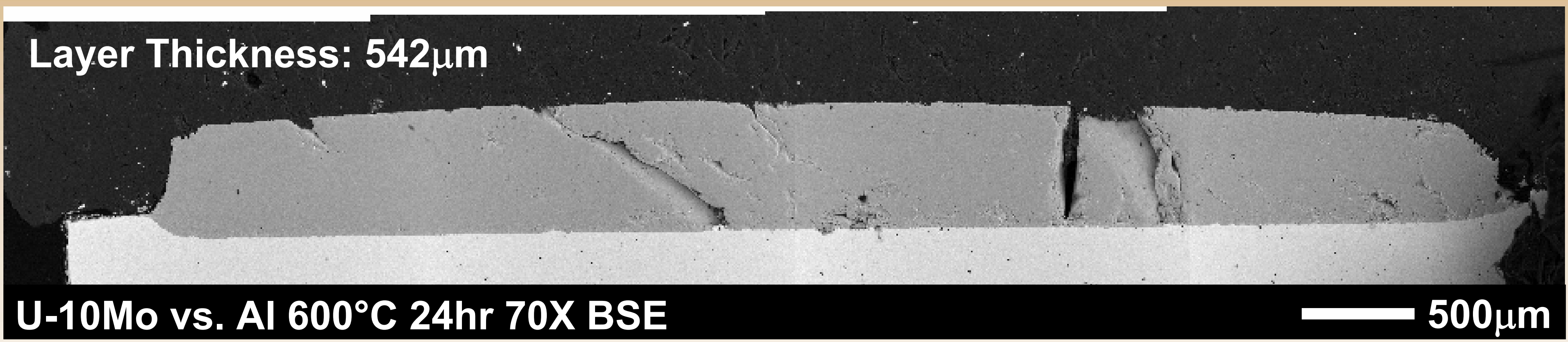
XRD Analysis of UMoAl alloys

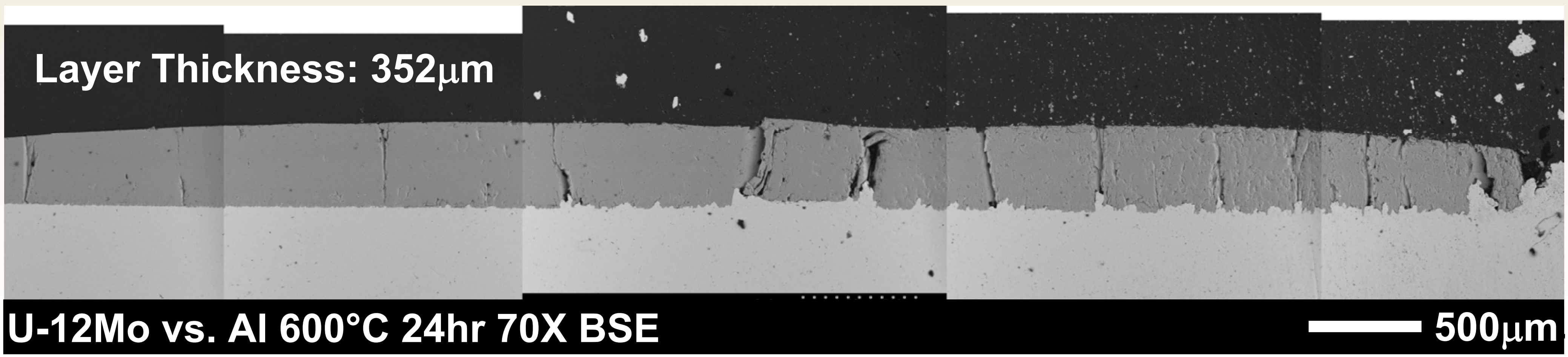


- The phases observed by XRD in 85.7Al-11.44U-2.86Mo and 87.5Al-10U-2.5Mo alloys were:
  - Al, UAl_4, $U_4Mo_6Al_{43}$ and UMo_2Al_{20}.
  - The UAl_3 phase volume fraction in the alloys was very small and not detectable by XRD.
- XRD of the alloys confirmed the presence of the phases found by TEM electron diffraction.

Diffusion Couples Annealed at 600°C for 24 Hours

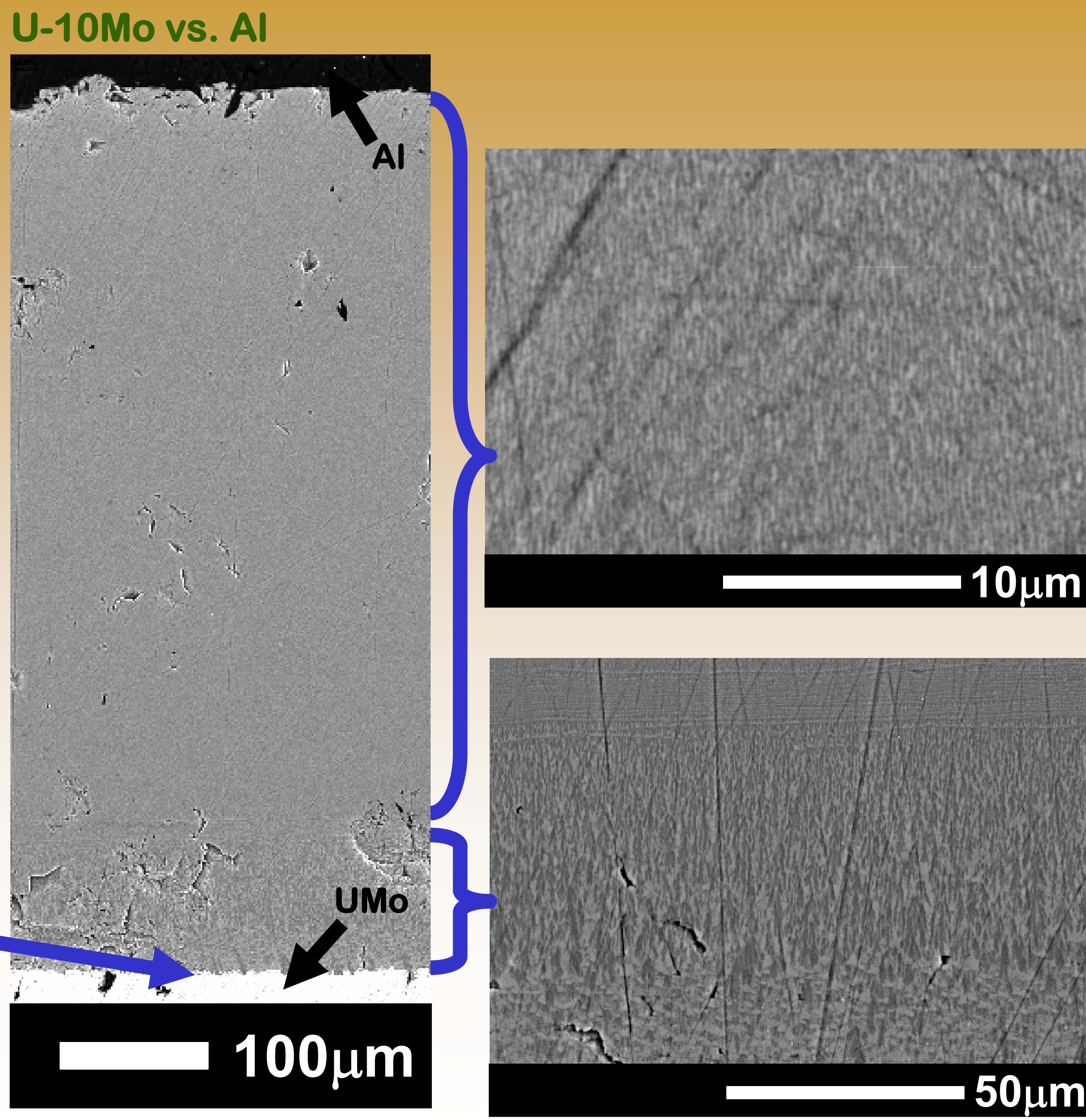


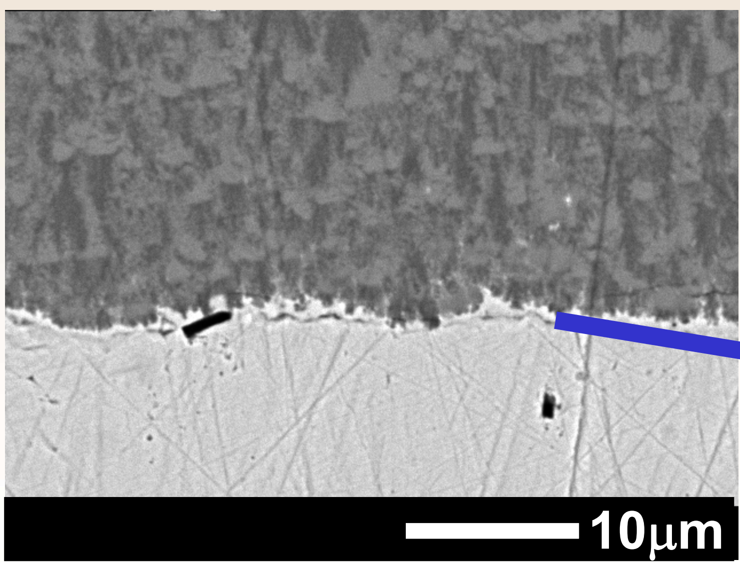




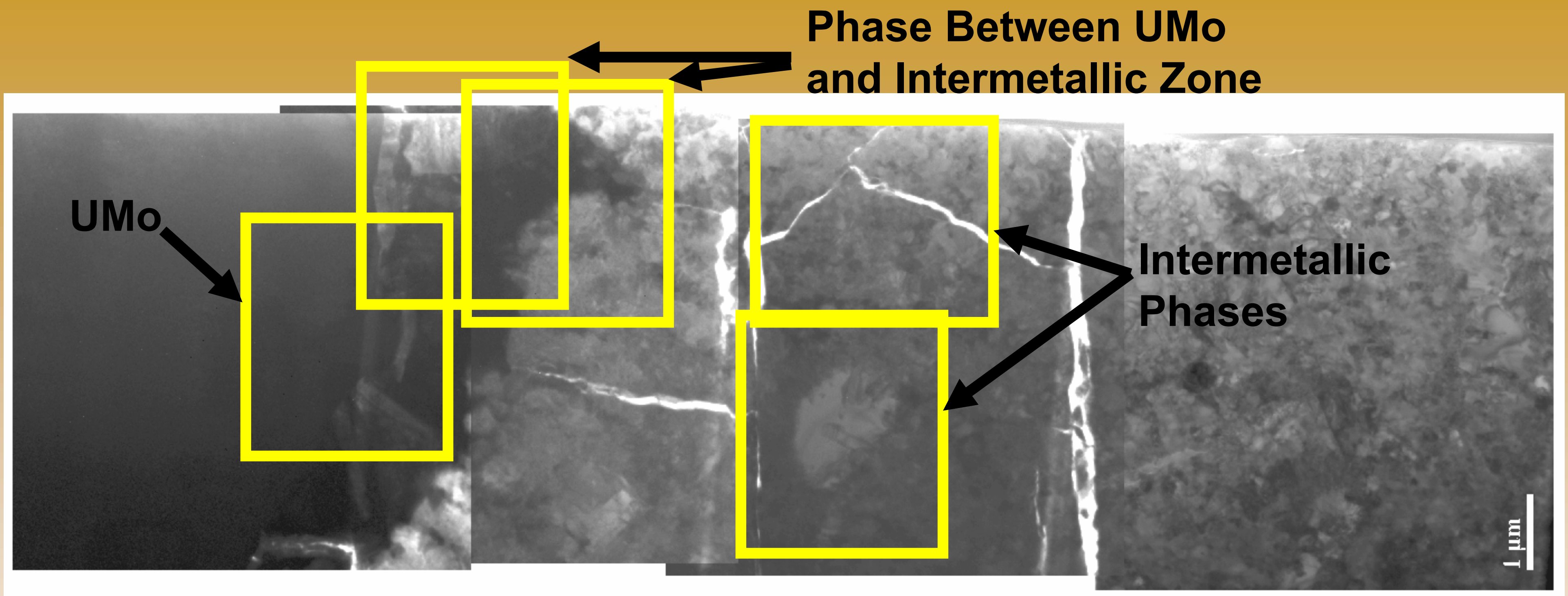
Diffusion Couples Typical Microstructural Development

- **Typical Microstructure:**
  - **Two-Phase regions composed of intermetallic phases**
  - **High U–phase at the UMo/Intermetallic interface**



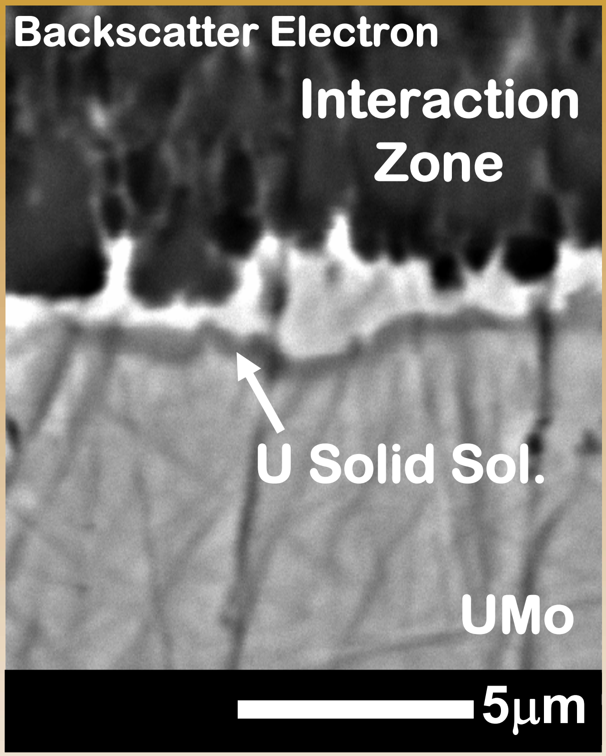


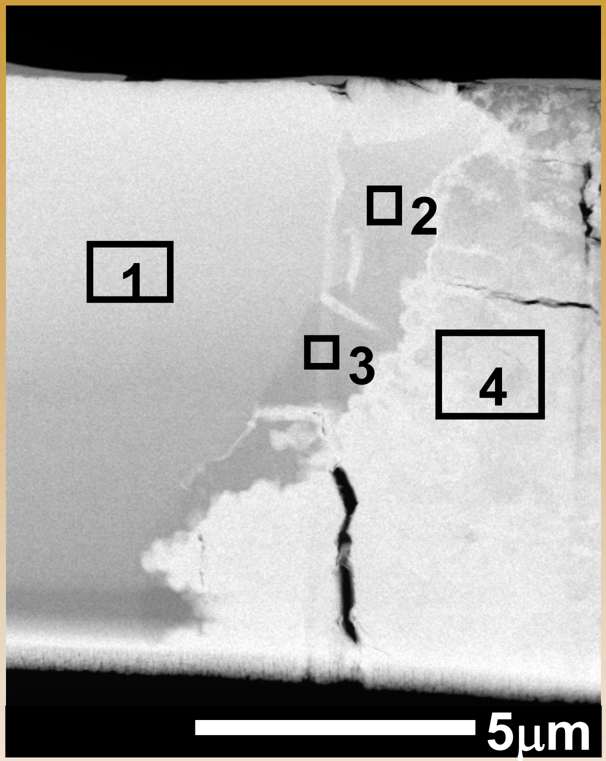
Electron diffraction

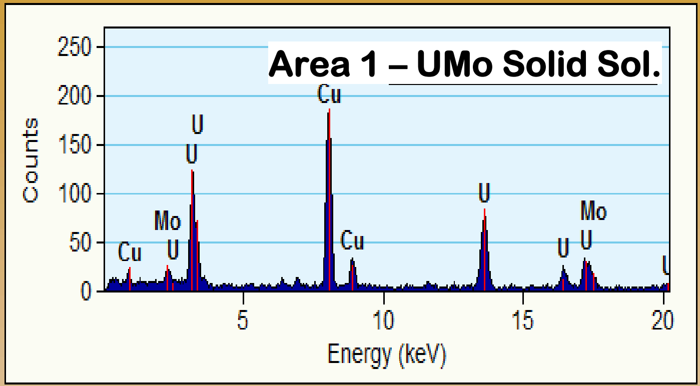


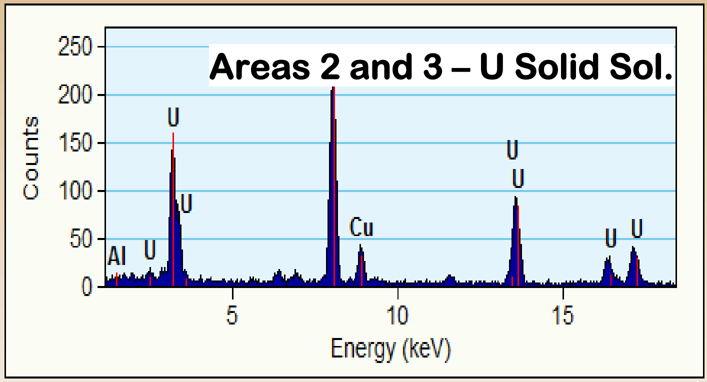
- Electron diffraction patterns have been collected on selected areas of the U-10Mo vs. Al diffusion couple annealed at 600°C for 24 hours.

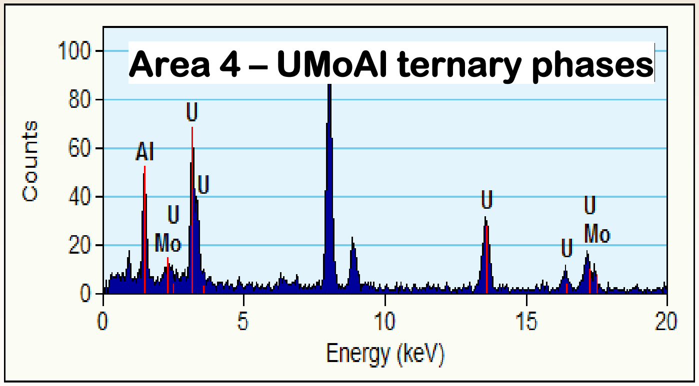
U-10Mo vs. Al HAADF and EDX 600°C 24hr UMo/Intermetallic Interface





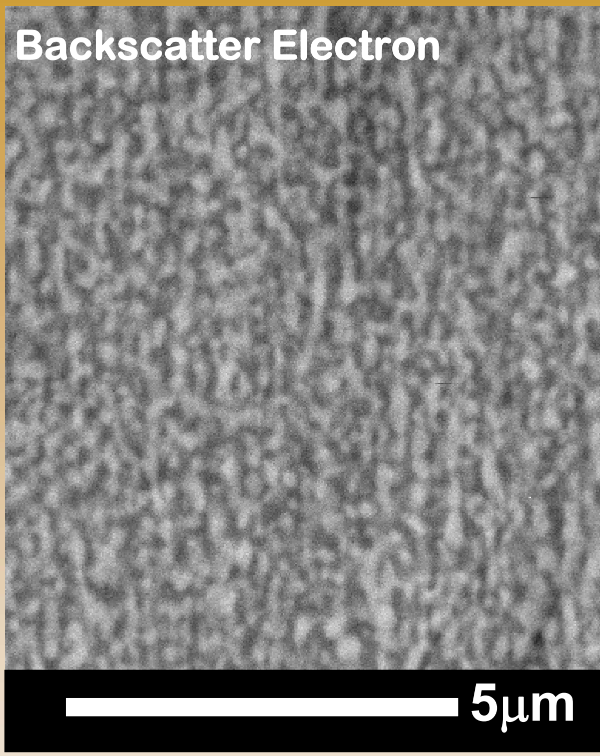


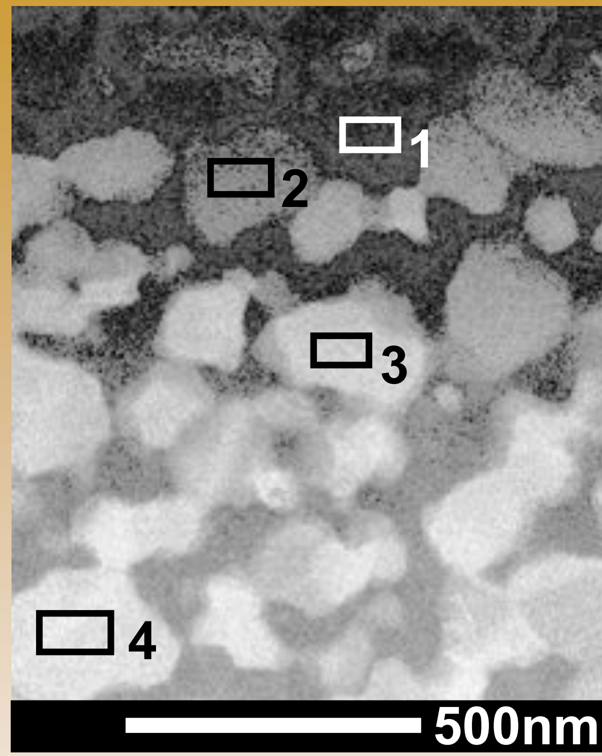


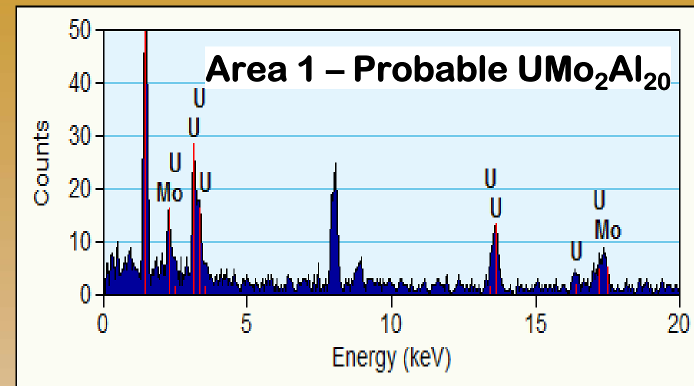


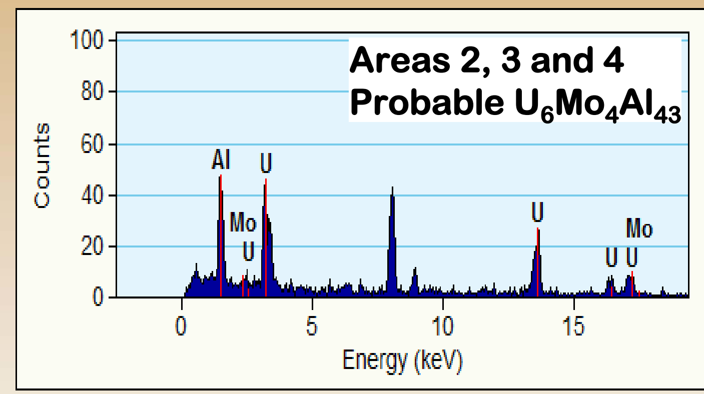
- **EDX compositional analysis identifies a U-solid solution depleted of Mo.**

U-10Mo vs. Al HAADF and EDX 600°C 24hr Center of Interaction Zone



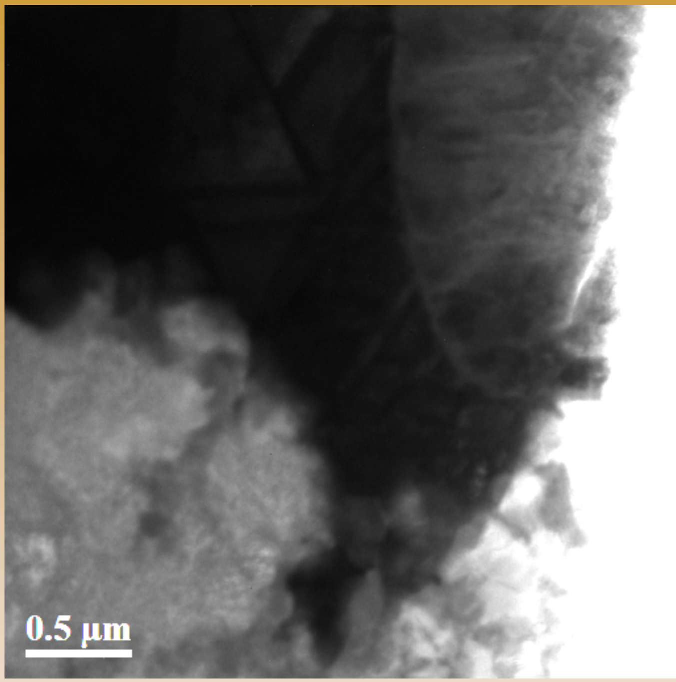


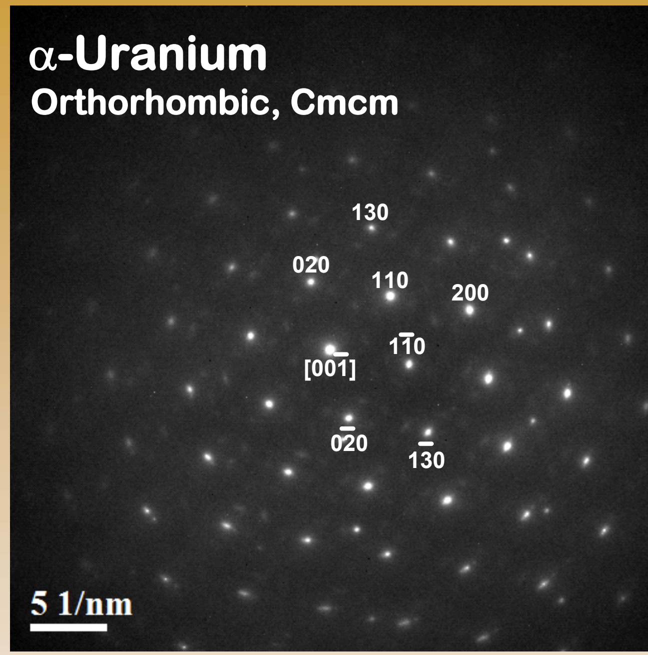


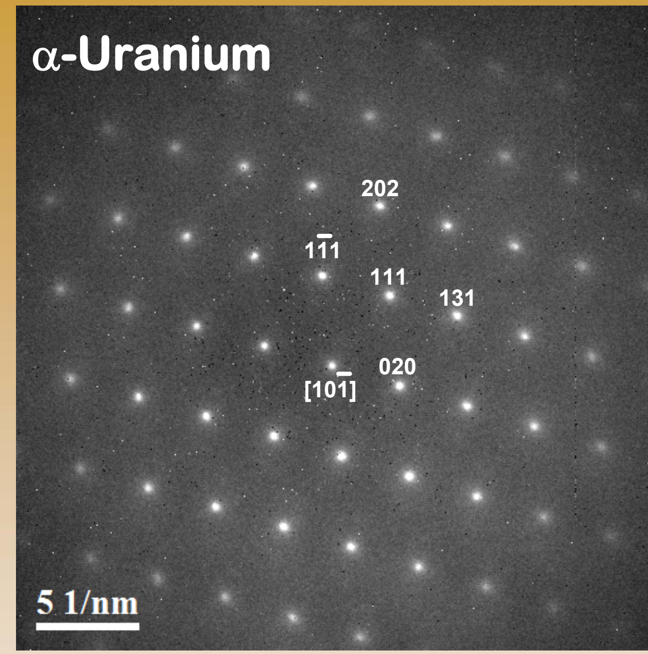


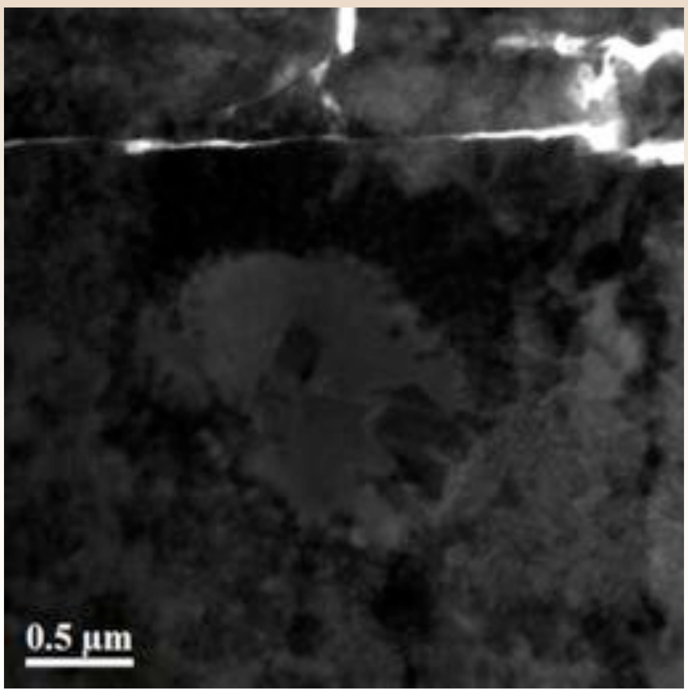
- Based on the development of ternary UMoAl alloys, the phases found by EDX may be the UMo_2Al_{20} and the $U_6Mo_4Al_{43}$.

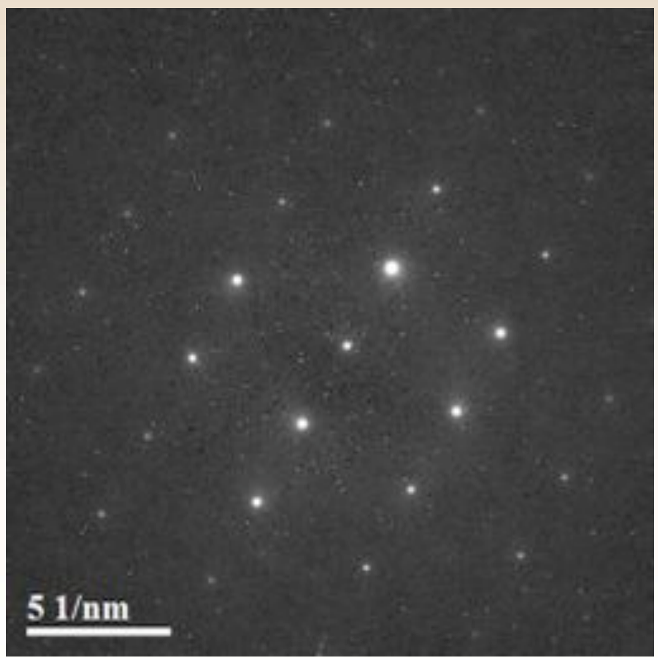
U-10Mo vs. Al 600C 24 hr Diffusion Couple
Selected Area Electron Diffraction

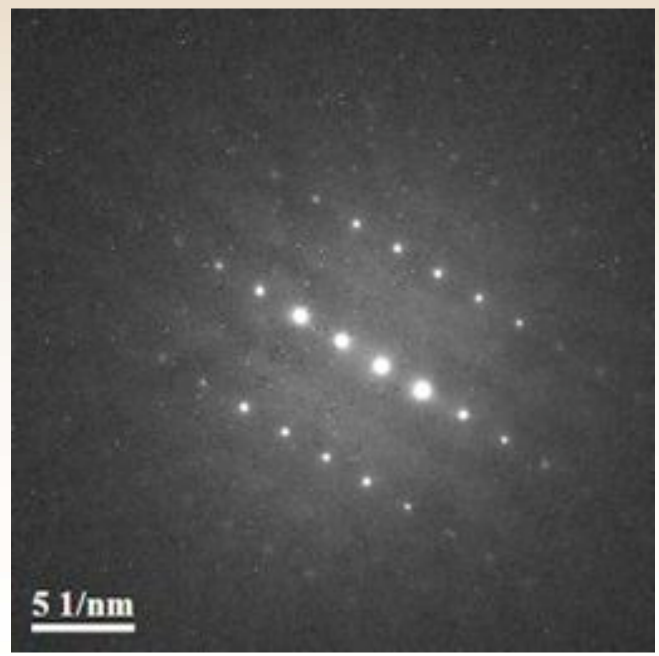




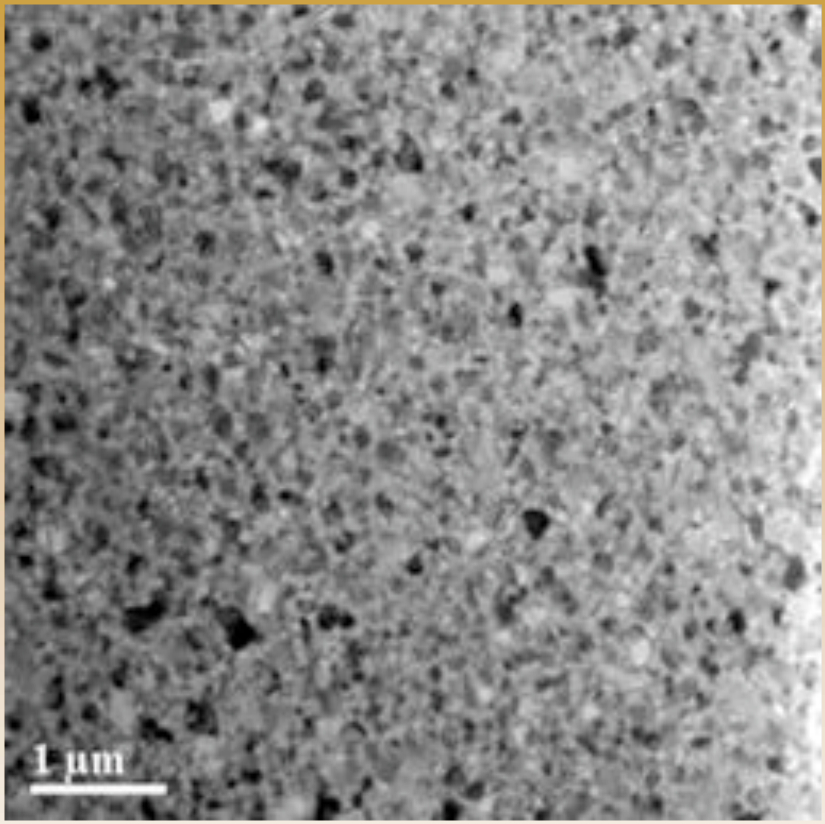


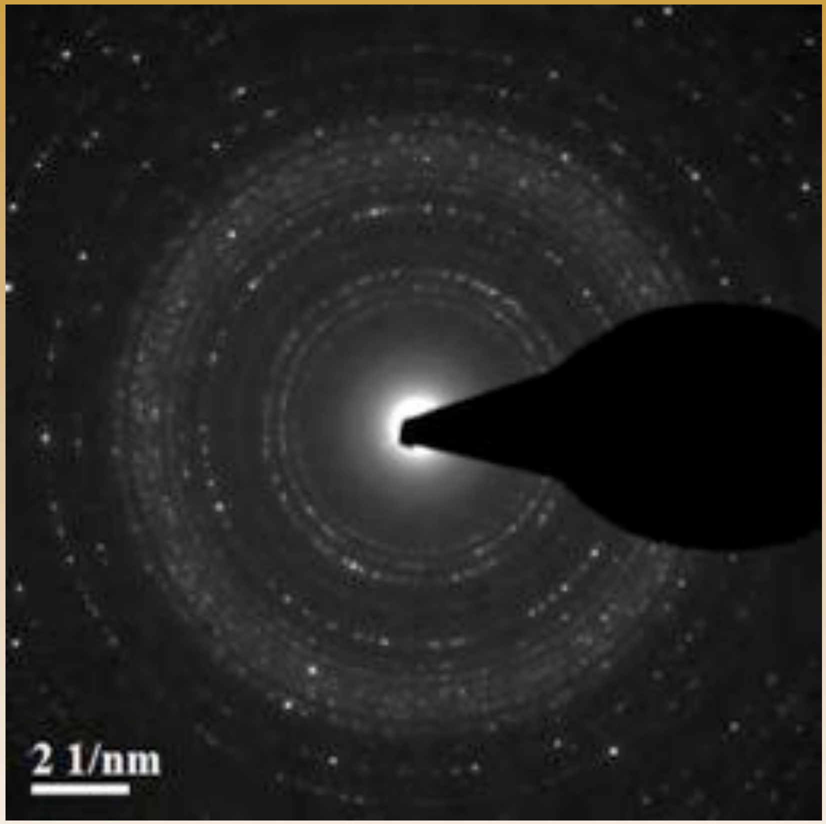






U-10Mo vs. Al 600°C 24hr
Polycrystalline Electron Diffraction Pattern





- 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-$U_6Mo_4Al_{43}$ and diamond cubic-UMo_2Al_{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.

Acknowledgments

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