NE 591: Advanced Reactor Materials

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Last Time

- USi interaction with Al leads to U(Al,Si)3 phases which can amorphize
- Fission gas bubbles largely are retained in the fuel, with bubbles appearing in the interaction layer at very high burnups
- UMo dispersion fuels allow for greater U density
- Mo stabilizes the gamma phase, and the gamma is additionally stabilized under irradiation
- Fuel swelling is critical in UMo fuels, with the unique feature of a fission gas superlattice
- Subsequent recrystallization leads to breakaway swelling
- Addition of Si can suppress the interaction layer formation in UMo particles

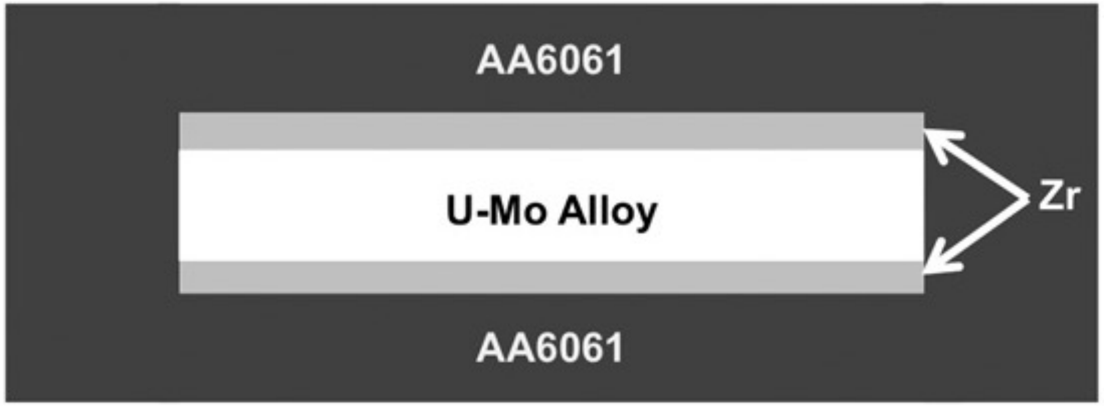
RESEARCH REACTORS

UMo Dispersion to Monolithic

- Higher power, high-burnup testing of aluminum matrix U-Mo dispersion fuel revealed a pattern of breakaway swelling behavior at intermediate burnup
- Breakaway swelling is defined by a rapid or unpredictable transition to high swelling behavior
- This swelling behavior was due to interaction of the fuel particles with the Al matrix, forming (U,Mo)Al compounds
- Large fission gas bubbles can form in these phases, weakening the fuel and exerting gas pressure, potentially resulting in fuel failure
- To remove this (U,Mo)Al phase, we can: 1) coat the particles; 2) modify chemistry (such as by adding Si); 3) substitute the Al matrix with another materials; 4) remove the matrix

Monolithic Fuels

- The U–Mo monolithic fuel, in which a U–Mo thin foil is sandwiched between a Zr liner and directly bonded to cladding, is currently under development
- This fuel design has the advantage of providing higher U density than the dispersion form while essentially eliminating the problem related to reaction products between the fuel and matrix
- Si additives were tried, but led to delamination of the fuel from cladding



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Foil/Plate Fabrication

- Any new nuclear fuel first requires the development of conceptual fabrication technology to prepare samples for irradiation testing
- Monolithic fuel is fabricated, in general by (1) alloying and casting, (2) foil rolling, 3) bonding of the cladding, and (4) finishing and inspection for quality assurance
- Fabrication processes are still being refined
- Alloying is typically performed through vacuum induction melting (VIM) or arc melting of uranium and molybdenum metals at a temperature of 1300 1500C.
- The molten alloy is typically cast into a coated graphite book mold
- The casting is surface machined into a ‘coupon’ of the appropriate size for reduction into a foil though hot and cold rolling

Foil/Plate Fabrication

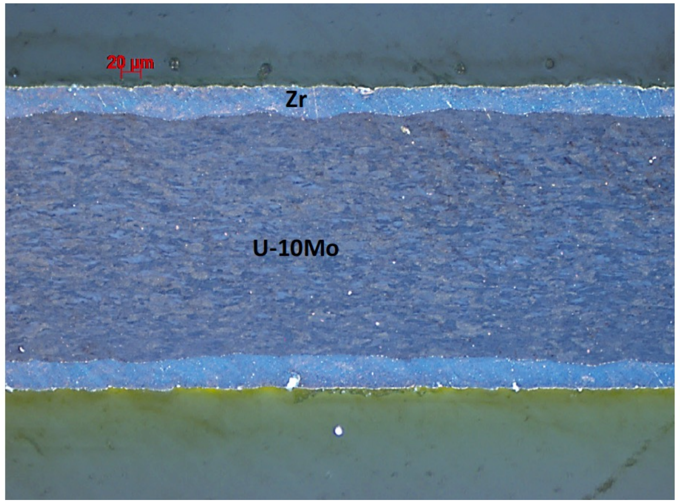
- Foil fabrication involves reducing the thickness of a cast alloy coupon to the desired fuel foil thickness while simultaneously bonding the Zr barrier layer to the surface of the U-Mo
- Hot rolling operations are conducted with the alloy temporarily encapsulated in a steel rolling can
- This allows the material to be heated to 650C in air without oxidizing the fuel alloy and the Zr
- After decanning, the resulting U-10Mo fuel foil, bonded to two thin (0.025 mm) layers of zirconium, sandwiched between two sheets of aluminum and placed in a steel can for HIP (Hot Isostatic Press) processing
- The HIP process is typically conducted at a temperature of 560C for a time of 90 minutes or longer
- The HIP can is cut open; the bonded plates are removed, and the plates are finished to final dimensions and inspected

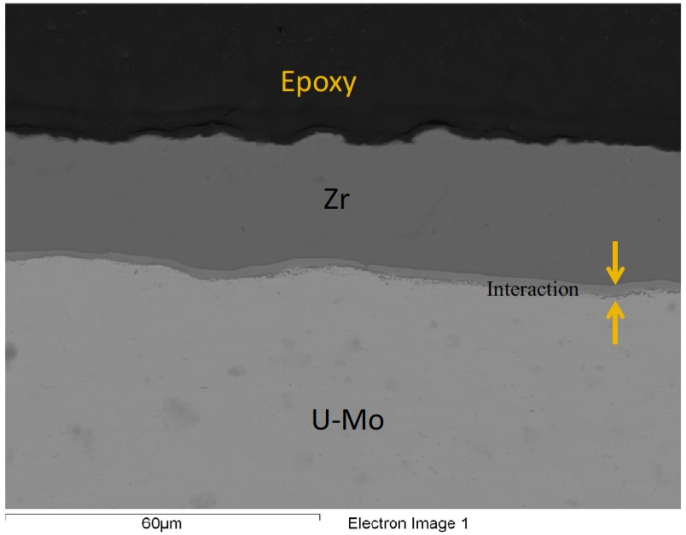
UMo Microstructure

- The microstructure of U-Mo foil depends on feedstock impurity content, the starting condition of the fuel coupon, and the rolling schedule
- The size and morphology of the grains depend on the thermo-mechanical treatments that are used during fuel-plate fabrication
- Molybdenum depleted and enriched regions in the as-cast microstructure resulting from solidification form elongated features during rolling
- Bands of differing Mo concentration can be observed in the final foil
- Decomposition of the original gamma (U,Mo) phase results in a lamellar structure comprised of alpha-U and gamma'-phase
- These lamellar structures can manifest themselves as bands within the foil microstructure, or as decomposed regions along the grain boundaries

Interaction Layer

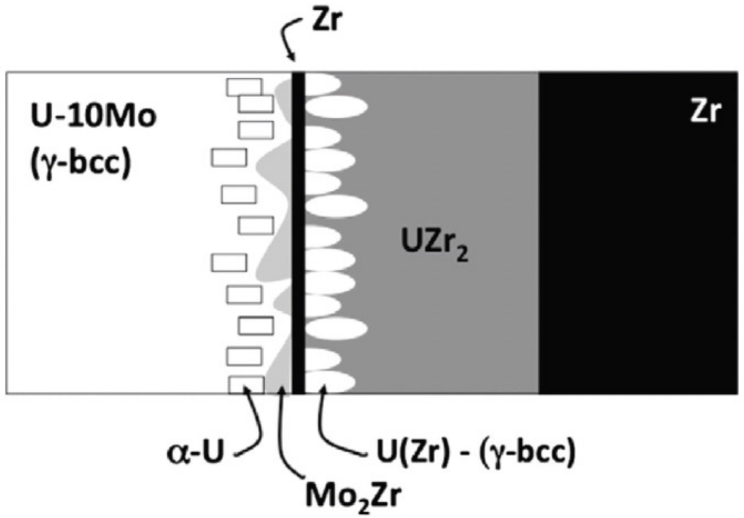
- A key feature that defines the irradiation performance of monolithic fuel is the interface between the fuel meat and the cladding
- The approach was to minimize the interaction between the U-Mo fuel foil and the cladding through introduction of a Zr diffusion barrier
- A barrier thickness of 25 μm was selected to exceed the maximum fission fragment recoil range (~9 μm) and to allow for variability in the manufacturing process

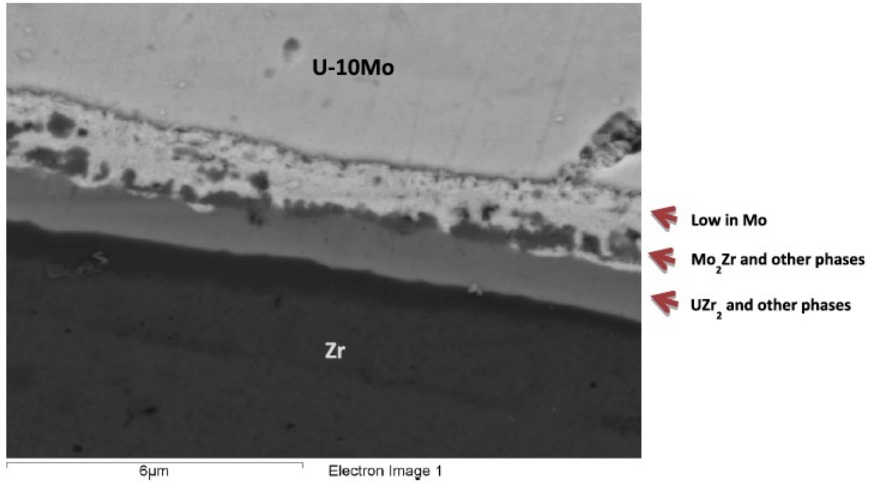




Interaction Layer

- The U-Mo/Zr interface contains multiple phases that develop during the interdiffusion that occurs between U-10Mo and Zr during the co-rolling and HIP processes
- Phases observed near the U-10Mo
- interface with the Zr diffusion barrier include: UZr2, gamma-UZr, Zr solid solution, Mo2Zr phases, and small amounts of alpha-U were also observed in the Mo depleted zone that forms





Zr/Al Interaction Layer

- The Zr/Al-6061 cladding interface must also be well-bonded and stable
- This multi-phase zone develops during HIP processing
- Ten different binary Al-Zr intermetallic compounds exist, however, not all form
- Si interacts with the Al-Zr system to form four distinctive layers AlSi4Zr5, (Al,Si)Zr3, (Al,Si)3Zr and (Al,Si)2Zr
- Interdiffusion layer can be limited by processing at lower temperatures

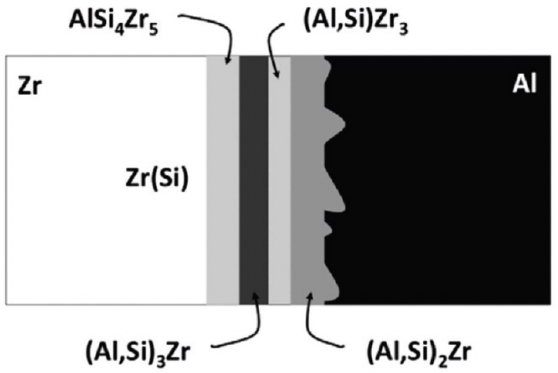
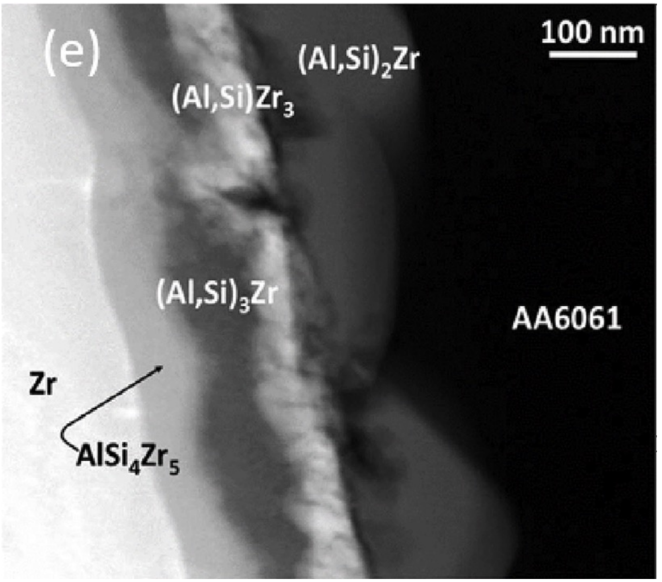
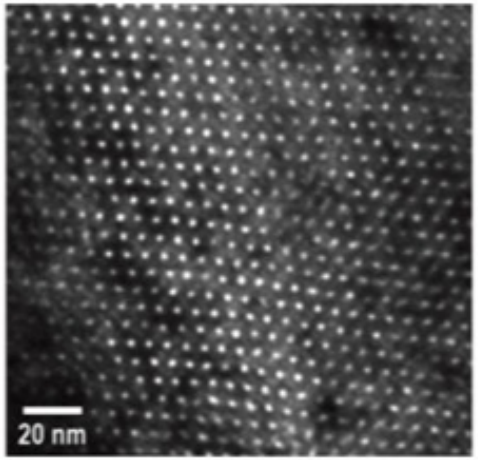


Fig. 9. Schematic Representation of Microstructure at the Interface between the Zr Diffusion Barrier and Al-6061.



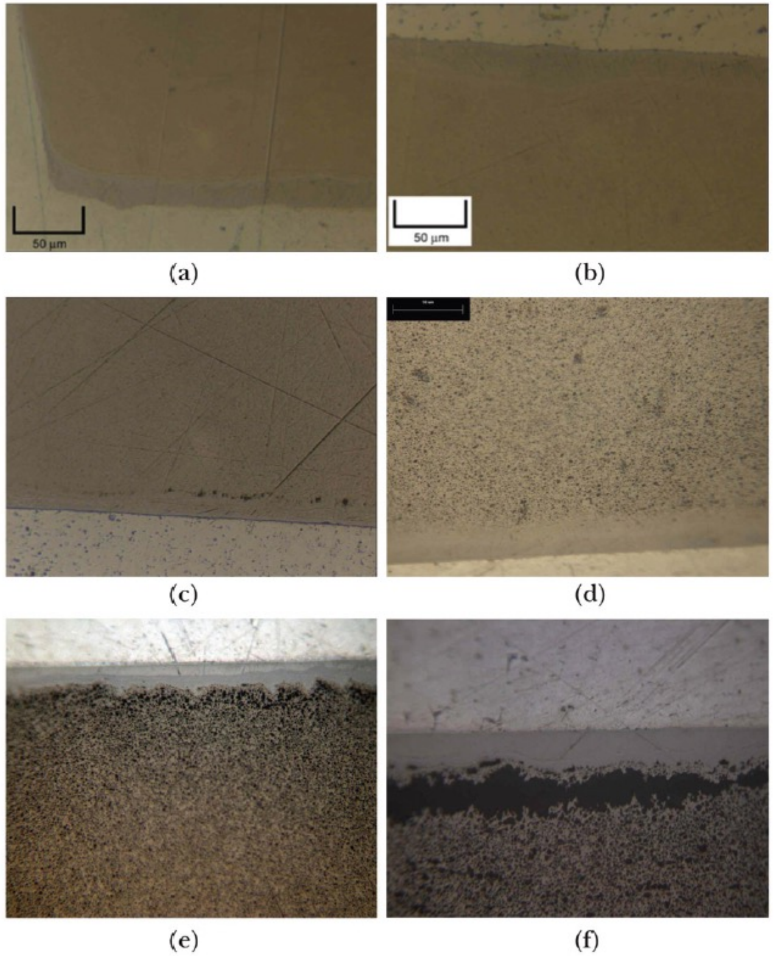
Monolithic Irradiation Behavior

- The key phenomena that affect the dimensional stability of a monolithic fuel plate during irradiation are fuel swelling, phase transformation under irradiation, radiation-enhanced diffusion resulting in the formation of unstable phases, creep of materials, and mechanical-property degradation
- These are similar to dispersion fuels, with the difference of a single connected fuel system and the potential for macroscale changes
- Fuel swelling undergoes the fission gas superlattice formation, and subsequent recrystallization (and FGS destruction) leading to accelerated swelling
- Solid fission product swelling occurs in the same manner as with other intermetallic fuels



Microstructure Evolution

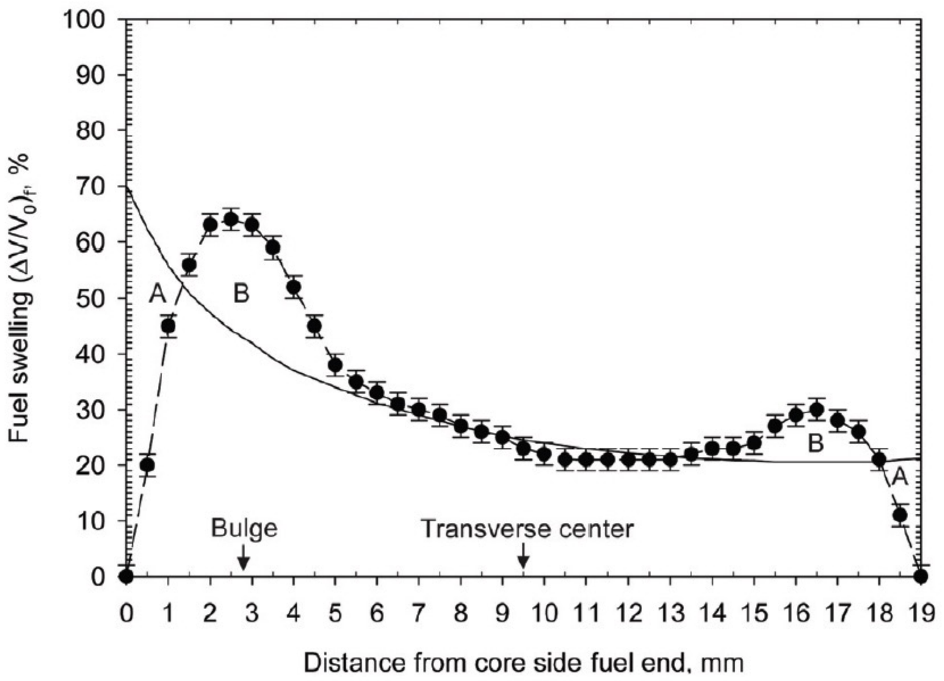
- At fission densities in the range of 2.2E27 to 4.0E27 f/m3, little microstructural change is apparent using OM
- As fission density increases from 4.0E27 f/m3 to 6.2E27 f/m3, grain refinement occurs, and formation of micron-scale porosity is visible in some regions near the U-Mo/Zr interface
- At a fission density of 7.2E27 f/m3, grain refinement has occurred throughout the majority of the microstructure
- Delamination of the fuel near the U-10Mo/Zr interface occurs at a local fission density of 9.5E27 f/m3



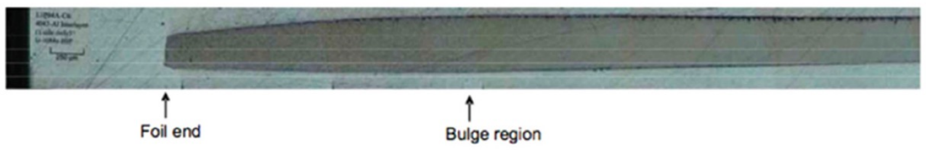
Fission Density
(a) 2.2,
(b) 4.0,
(c) 6.2,
(d) 7.2,
(e) 8.4,
(f) 9.5

Creep and Peaking

- Irradiation enhanced creep is a key phenomenon that makes the monolithic fuel system viable
- Fuel foils are constrained from swelling in the plane of the fuel plate by the cladding rails, and examination of irradiated fuel plates shows that little or no dimensional change is measured in this plane
- Plate failures occur in the regions of the fuel plates with the highest fission density
- Peaks in fission density are caused by power peaking at the ends and edges of fuel plates in this test configuration

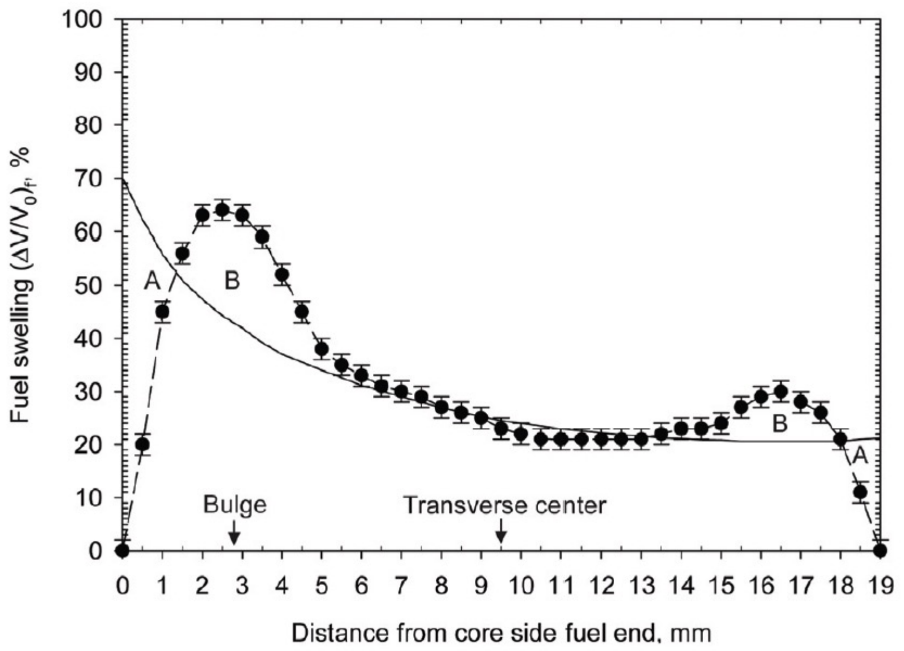


(a)



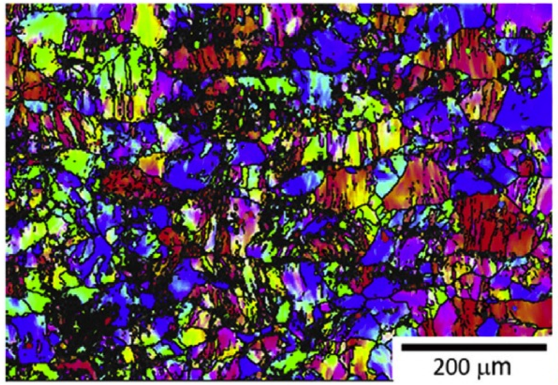
Creep Forming Bulges

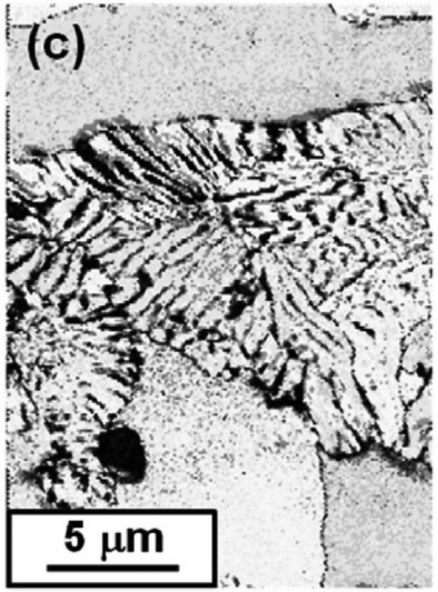
- The formation of this thick region can be explained by irradiation enhanced creep of the fuel driven by the stress that builds up at the rail constraint caused by fission product swelling
- Swelling at the plate edges is lower than expected, due to the constraint of the plate rail
- Fuel moves inward by irradiation-enhanced creep to form the thick region approximately 3 mm from the edge of the foil
- While there is a creep correlation to predict this dimensional change, very little is known about UMo creep



Effect of Fabrication on Microstructure

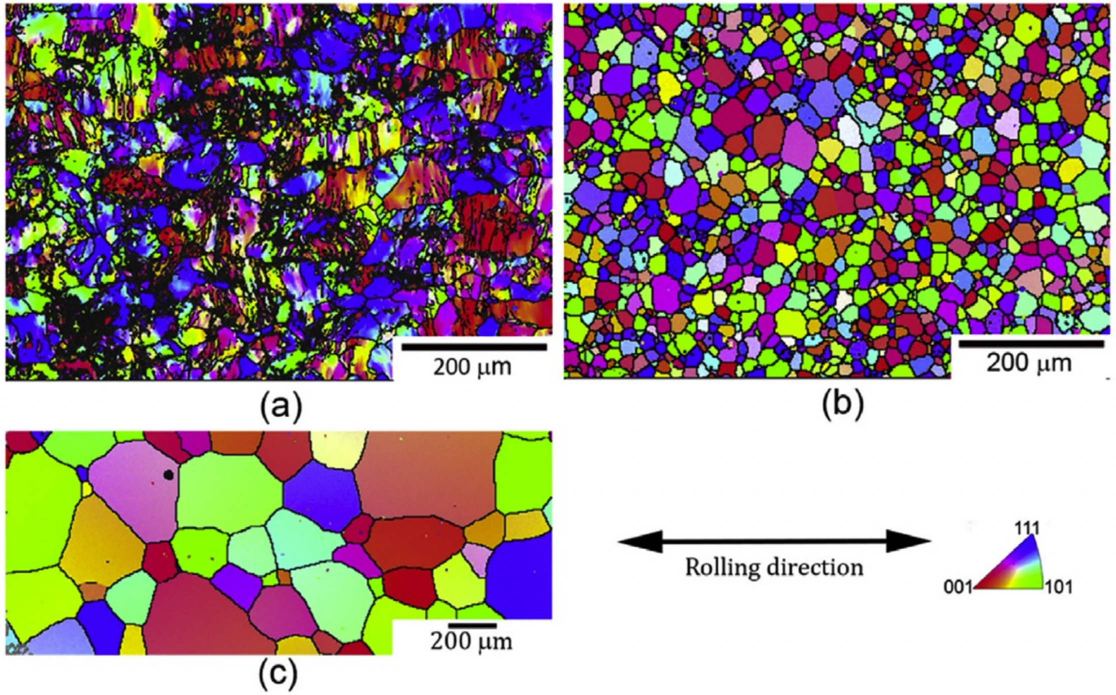
- The rolling process can degrade the gamma phase, inducing phase decomposition
- The eutectoid decomposition initiates when alternate lamellar structures, consisting of alpha-U and gamma-U with higher Mo content than the matrix (or potentially U2Mo), forms in U-Mo fuel
- The lamellar structure tends to nucleate primarily along the prior gamma-UMo grain boundaries, but nucleation on deformation bands inside of the grains has been noted as well





Effect of Fabrication on Microstructure

- When the as-rolled material is subjected to annealing (heat treatment), the grains relax internal stresses
- Grain size distribution of U-10Mo fuel widens with an increase in annealing temperature, likely due to pinning from precipitates
- Homogenization of the microstructure must be weighed against cost of high temperature fabrication processes



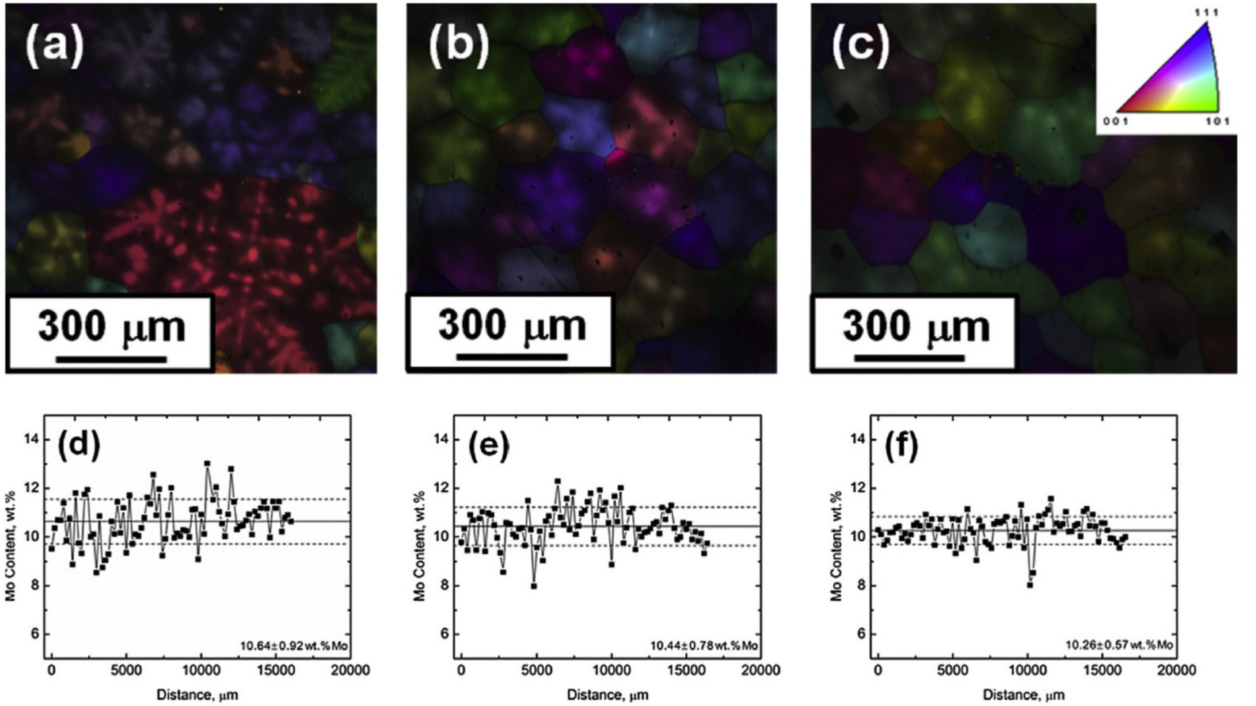
(a): as-rolled (b) rolled and annealed at 700C and (c) rolled and annealed at 1000C

Microstructure Effects on Performance

- When the decomposed regions revert back to gamma-UMo, a perfect phase transformation is not possible, and a network of dislocations is left in the gamma-UMo matrix
- This increased dislocation density has been experimentally associated with grain refinement zones
- Thus, with an increase in the decomposed regions in the as-fabricated fuel, there are additional nuclei for grain refinement

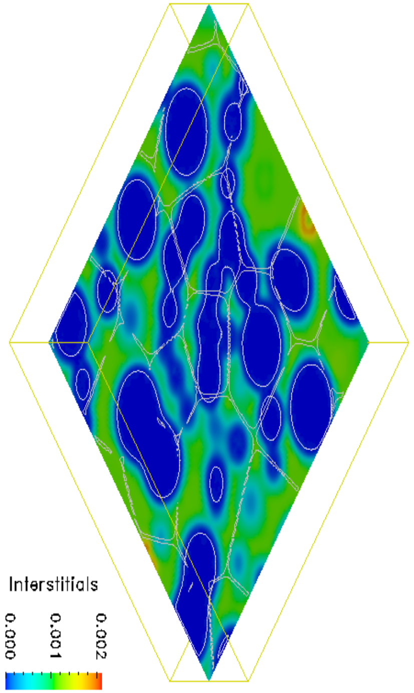
Chemical Homogeneity

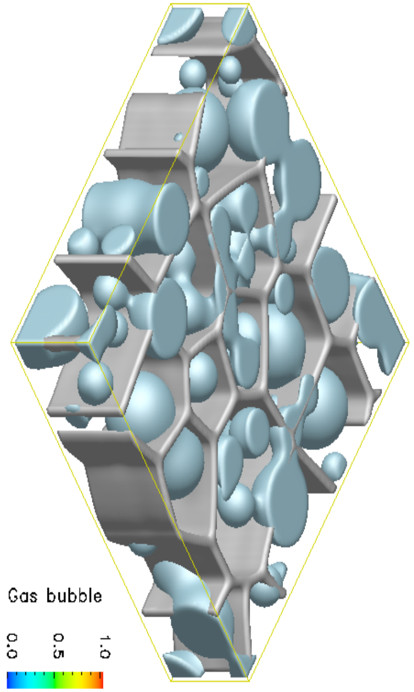
- Due to the solidification process, similar to atomization, Mo rich regions will solidify first, and Mo depleted grain edges/boundaries will form
- Decomposition is more likely in Mo-lean areas, due to the phase diagram
- Homogenization processes can be performed (high T annealing) to reduce the chemical gradients
- Mo concentration gradients change with the homogenization temperature



Microstructural Modeling of UMo

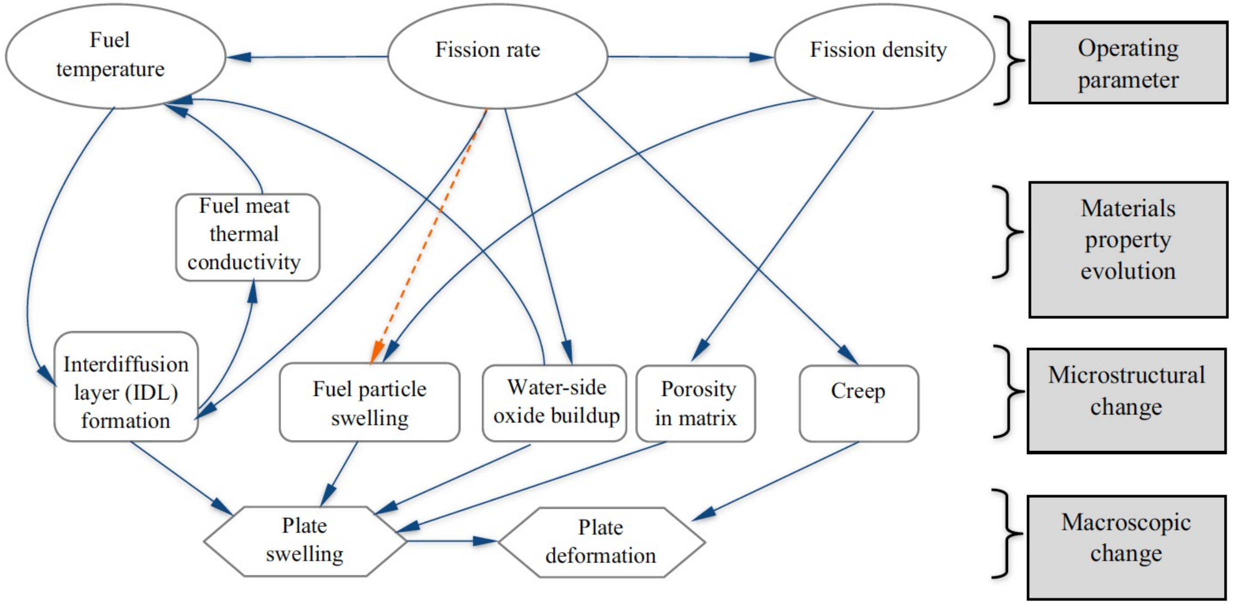
- Lower length scale modeling in the USHPRR program was initiated in 2015 to mitigate risks associated with unknown behaviors of UMo fuel
- The AFIP6-MkII experiment exhibited early onset breakaway swelling which did not fit with previous experimental efforts
- LLS modeling can explore fundamental phenomena to obtain mechanistic predictions of fuel behavior





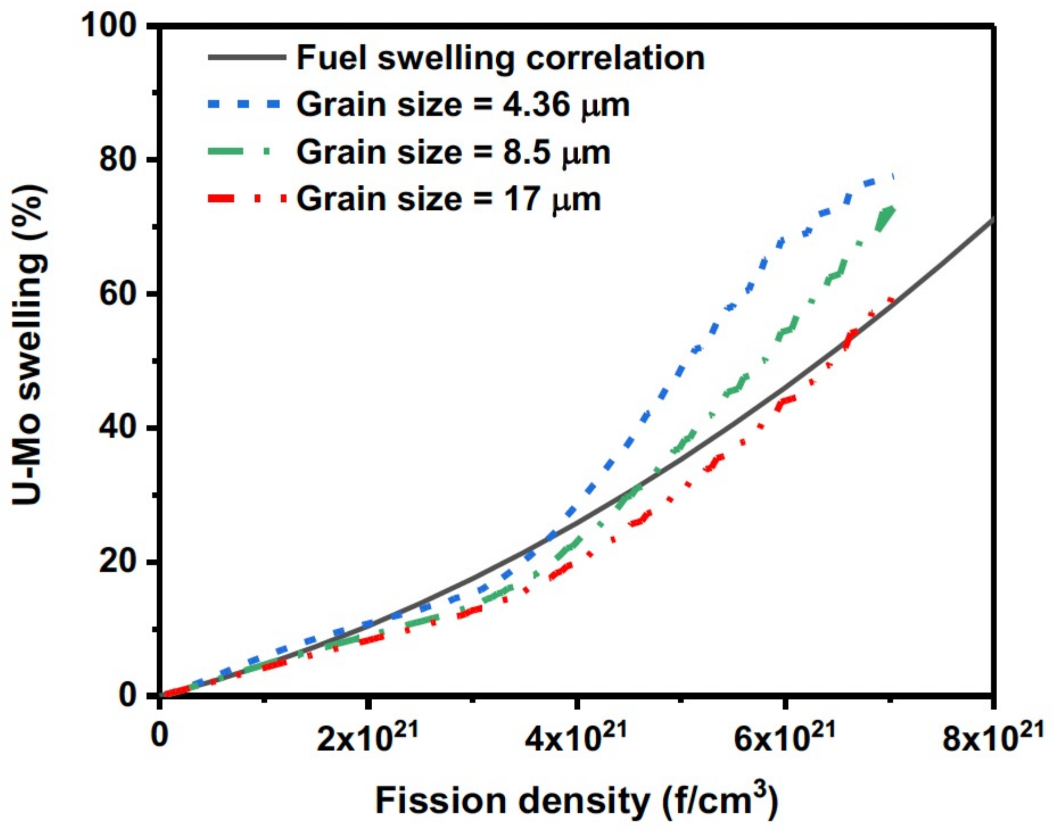
Microstructural Modeling of UMo

- The major physical processes impacting RTR fuel performance are intricately related
- The key four classes of physical processes: operating parameters, fuel microstructural and materials property changes, and dimensional changes of the fuel plate
- Microstructural change is the nexus of the irradiation behavior of fuel plates



Grain Size Effects on Swelling

- Grain size can affect the rate and magnitude of swelling
- Initial grain size impacts the rate at which the FGS is destroyed
- The initial conditions for which intergranular bubbles and new grains can nucleate are define by as fabricated grain size
- The continuing rate of grain refinement is also governed by initial grain size



Aluminum in Research Reactors

- Aluminum alloys are generally too weak or have temperature limitations that preclude their use in commercial reactors
- In most research reactors where bulk water coolant temperatures are usually <100C, aluminum alloys are quite comfortable and are universally employed

Material	*Density ($kg\ m^{-3}$)*	*Specific heat ($J\ kg^{-1}\ K^{-1}$)*	*Thermal conductivity ($W\ m^{-1}\ K^{-1}$)*	*Melting point (°C)*	*E_{mod} (GPa)*	*CTE, lin. ($\times 10^{-6}\ K^{-1}$)*	*Nuclear cross-section (barns)*	
Aluminum	2700	887–963	160–230	660	70	23	0.23	1.5
Zirconium	6490	254–285	8–40	1852	88–98	5.7	0.19	6.4
Austenitic steel	~8000	377–565	11–21	~1425	190–201	~16	~3.0	~10
Ferritic steel	~7900	440–494	17–42	~1525	200–210	~12	2.5	11
Uranium	1900	111–167	11–28	1132	176–208	13.9	7.6	8.9

Aluminum Benefits

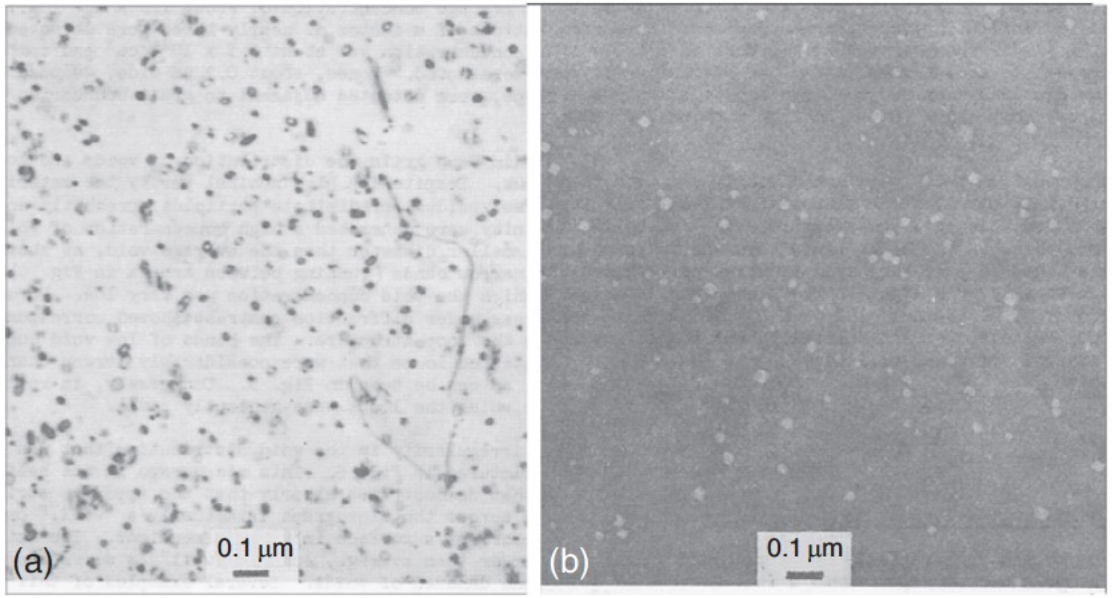
- Al is ductile, plentiful, cheap, and light weight
- It is castable, machineable, and weldable, and it can be shaped readily by conventional processes
- It has good aqueous corrosion resistance due to near-insolubility in water and formation of a passive, self-restoring surface film of hydrated aluminum oxide
- It has an fcc crystal structure and no crystallographic phase changes, with near isotropic properties
- It can be strengthened by cold work, solid solution hardening, and precipitation treatments
- At low temperatures, it has no ductile-to-brittle transition
- Its resistant to hydriding

Aluminum Drawbacks

- Al has a low elastic modulus and low melting temperature
- The low melting temperature of 660C imposes operating temperature limits of 100–150C
- The strength condition of prehardened alloys can become compromised at temperatures above 150C and Al alloys become substantially more susceptible to creep above 150C
- Difficulties may be encountered in obtaining leak-tight fusion welded joints for hi-tech applications
- It is prone to liquid metal embrittlement

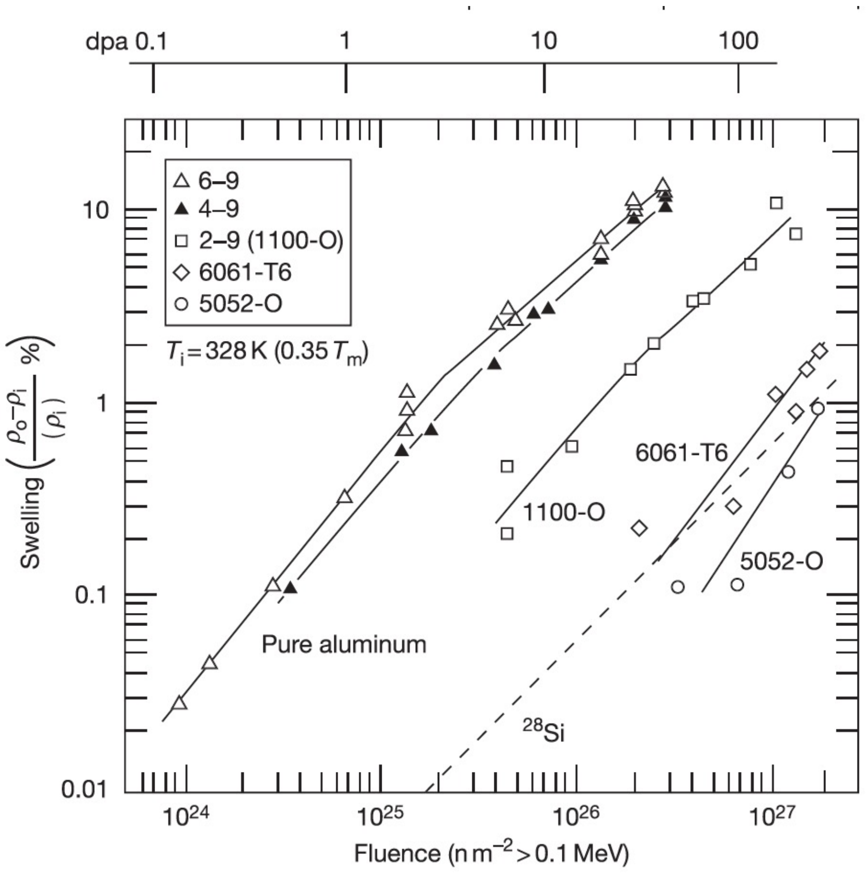
Damage Microstructure

- Dislocation loops (a) and voids (b) in high-purity aluminum after irradiation at 50C to a fluence of 3.5E24 n/m2
- The loops and voids are of order 30 nm diameter
- In typical nuclear cladding materials, under similar temperature and neutron fluence conditions, the radiation damage microstructure is resolvable as 1–2 nm black dots



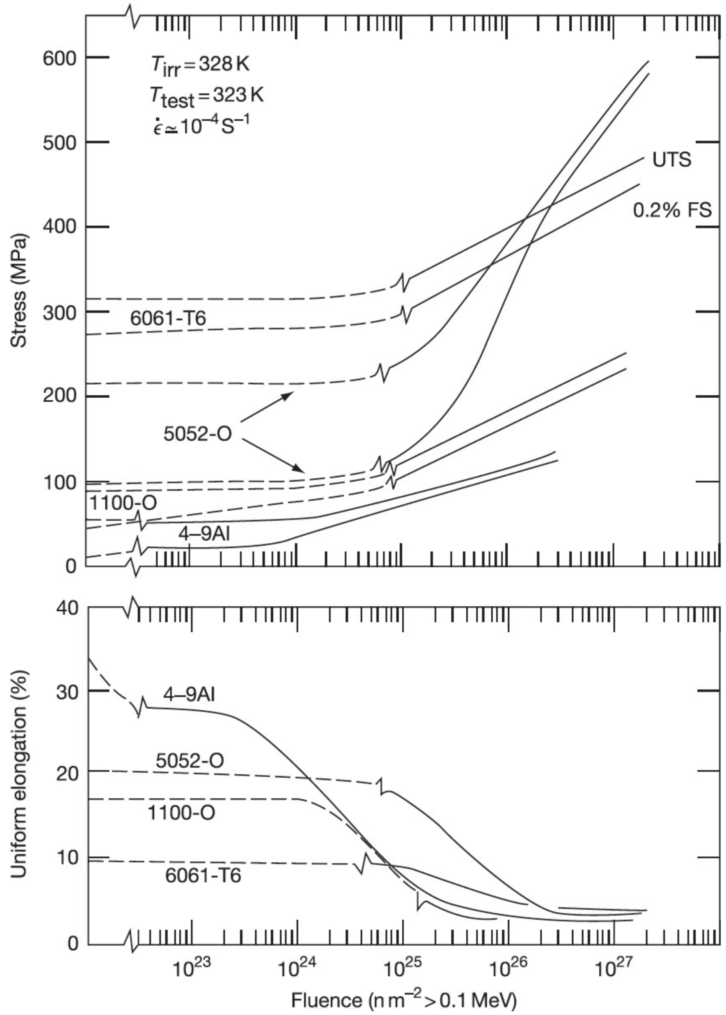
Swelling

- Swelling is due to the accumulation of voids from excess vacancies
- Gas bubble swelling is generally not an issue for Al in RRs because the temperature is too low
- The 6061 alloy, with its inherent Mg2Si phase, starts swelling appreciatively later in dose than the 1100-O or pure Al
- Swelling is limited to less than 1% for the higher levels of fluence expected in RRs



Hardening

- The major consequences of radiation damage structures on the mechanical properties of Al alloys are radiation hardening and associated loss in ductility
- Al6061 is generally a stronger alloy than comparable Al alloys
- Hardening and reduction in elongation occur at approximately 1E25 n/m2, with as much as a 2X (1/2X) effect on the associated property



Summary

- To remove this (U,Mo)Al phase, we can: 1) coat the particles; 2) modify chemistry (such as by adding Si); 3) substitute the Al matrix with another materials; 4) remove the matrix
- Fuel fabrication specifications can dramatically affect the microstructure and thus the performance of the fuel – this is more critical for monolithic fuels
- The Zr interdiffusion barrier prevents UMo-Al interaction, but creates new interaction layers with a series of phases present
- UMo fuel creeps, creating a bulge just inside the edge of the fuel
- Decomposed regions can accelerate grain refinement and swelling and can be reduced through chemical homogenization and annealing
- Al is ideally suited for the research reactor environment with its excellent corrosion and swelling resistance at applicable temperatures
- Microstructural modeling is attempting to describe the variety of governing phenomena in UMo fuel