Nuclear Fuel Performance

NE-591-010

Spring 2021

Last time

- Corrosion is the environmental degradation of materials
- Cladding oxidizes, forming ZrO_2
- The limiting step for oxidation is the oxygen transport through the oxide layer
  - It begins being controlled by diffusion
  - Then, a protective layer forms that slows oxidation
- Hydrogen released by oxidation enters the cladding
- Due to low solubility (that is a function of temperature), hydrides form
- Hydrides are brittle, and so reduce the ductility of the cladding

METALLIC FUEL MICROSTRUCTURE AND PERFORMANCE

Why metallic fuel?

- It has demonstrated a high burnup capability beyond 20%
- Metal fuel also has superior off-normal performance characteristics, in particular for the run-beyond-cladding-breach conditions due to the compatibility of metal fuel with sodium coolant
- The inherent passive safety potential where metal fuel is far superior to other fuel types (less stored Doppler reactivity)
- Metal fuel also allows a very simple injection-casting fabrication technique and electrorefining-based fuel cycle closure, which promises simpler waste management, proliferation-resistance, and much improved economics

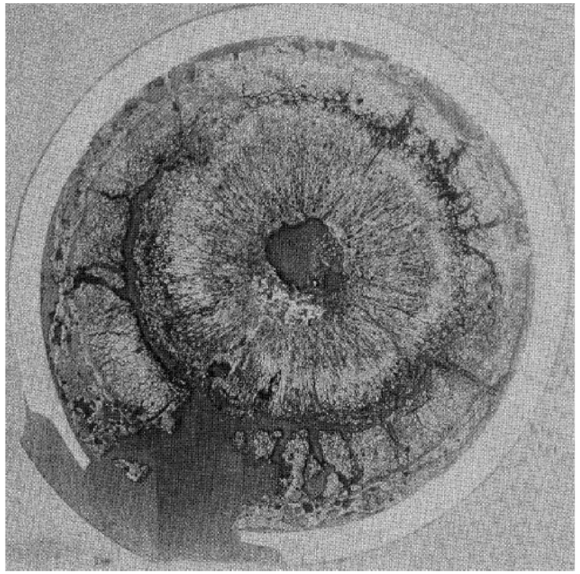


Fig. 3 Oxide Fuel (9% burnup) RBCB Test

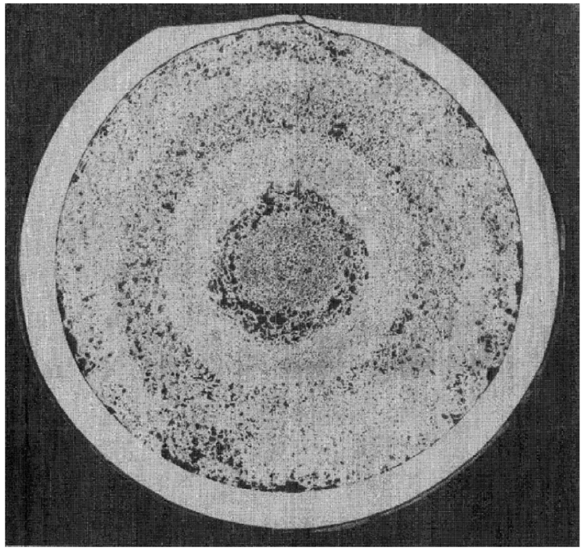
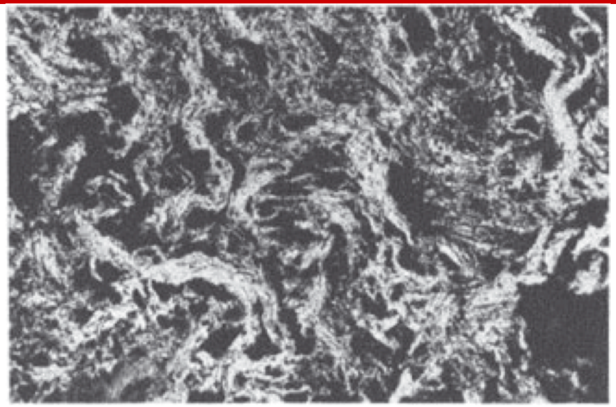
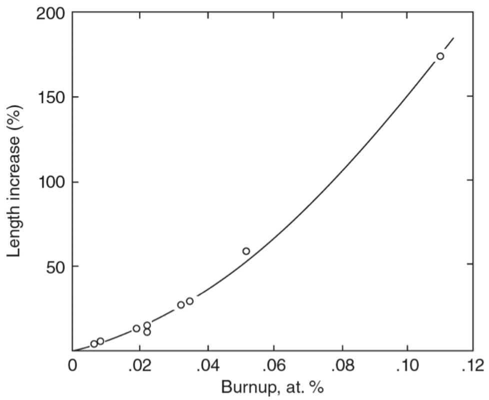


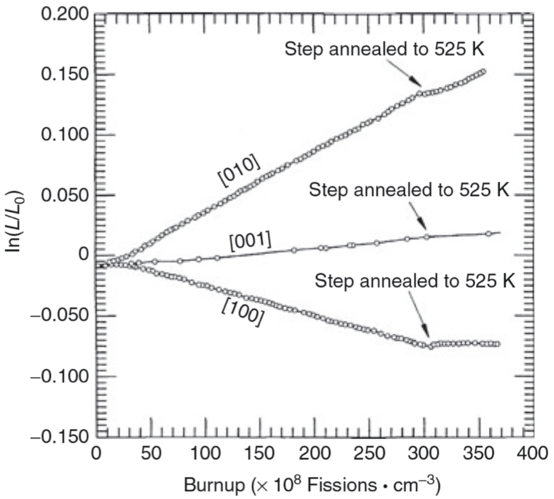
Fig. 4 Metal Fuel (12% burnup) RBCB Test

Why do we alloy?



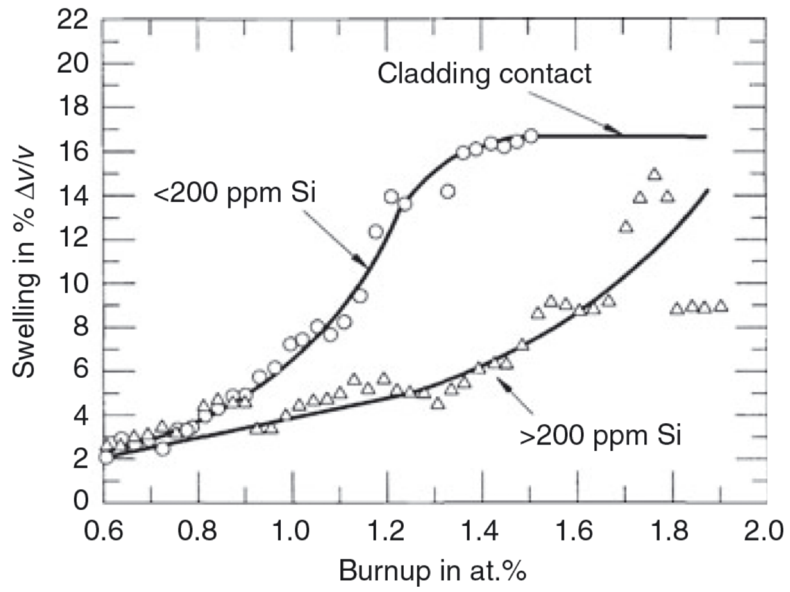
- Unalloyed swelling can reach >150% at target burnups
- Anisotropic swelling of alpha U
  - thermal expansion
  - irradiation growth
- Cavitation swelling is characterized by large irregular cavities that form by mechanical tearing at grain and sub-grain boundaries, resulting in a very deformed “swirled” microstructure





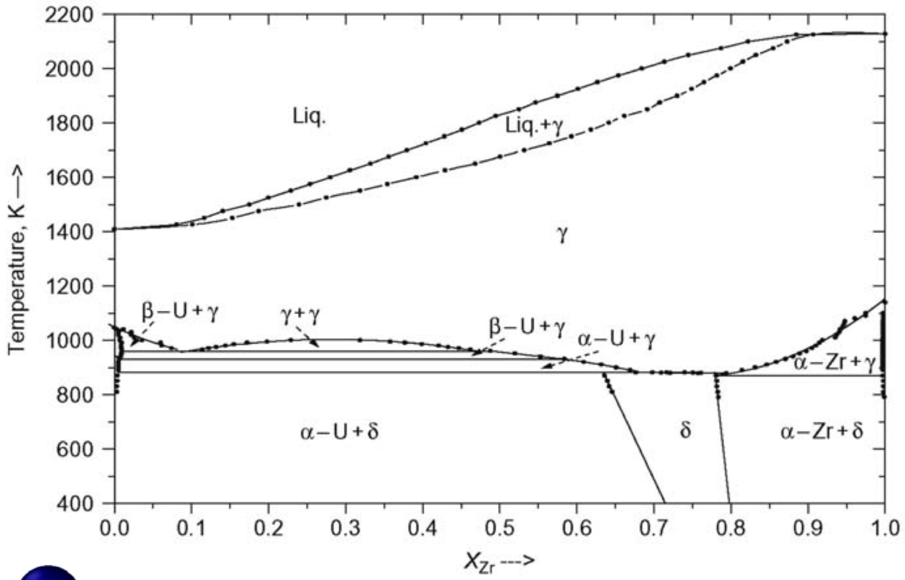
Alloying attempts

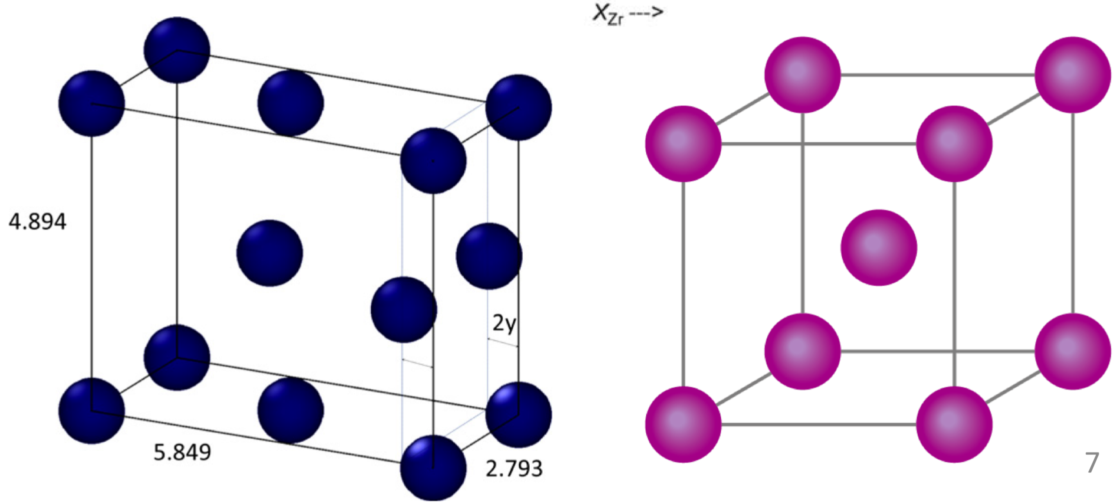
- Adjusted U
  - Small amounts of Al, Fe, or Si
- U-Fissium (U-Fs)
  - Fissium is an artificial mixture of the natural elements represented among the fission products to simulate the chemical composition of the material resulting from fission
- Further studies on U alloys have centered on elements that form extensive solid solutions with U in the high-temperature γ-phase, specifically Mo, Zr, Ti, and Nb
- U-Nb and U-Ti show excessive phase decomposition to alpha-dominated systems
- U-Mo is utilized in research reactors which operate at low T



U-Zr (U-Pu-Zr)

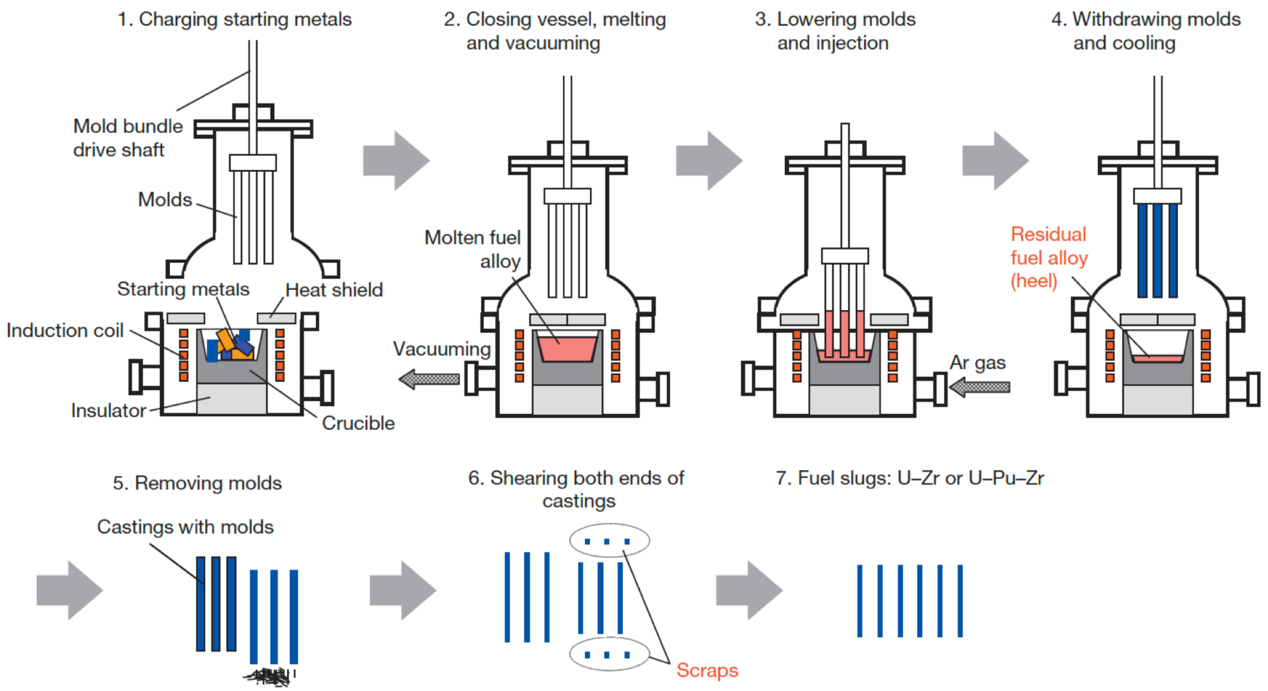
- Alloying with Zr increases melting point and stabilizes the high temperature bcc phase
- Temperature range for fuel is ~800-1100 K
- Typically 10 weight percent Zr (23 atomic percent)
- Directly in multiphase region
  – find bcc, beta, alpha U, delta UZr2, alternate bcc phases





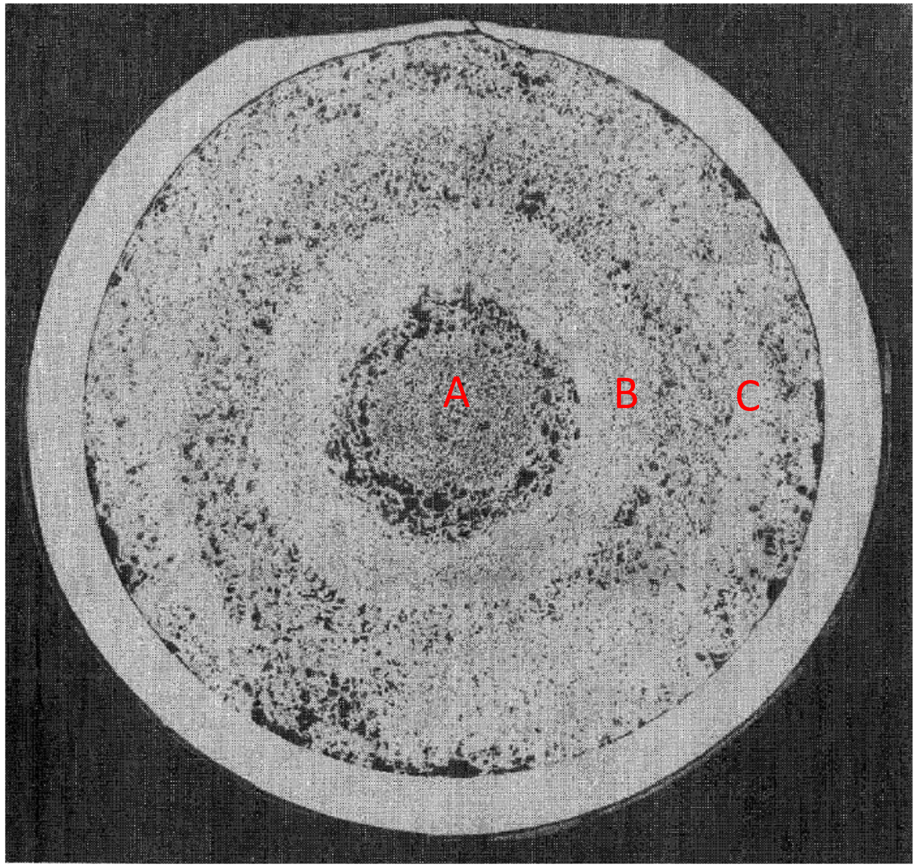
Fuel Fabrication

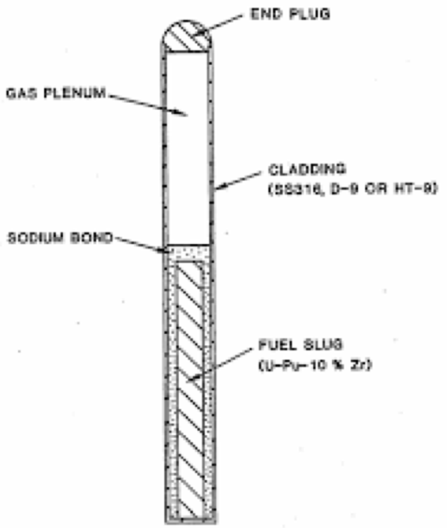
- Injection casting
  - starting materials are charged into the graphite crucible in the injection casting furnace
  - The crucible's interior is coated with yttria and the mold's interior is coated by zirconia for protection against reaction with molten uranium alloy
  - Silica tube molds with the top ends closed are set above the crucible
  - After melting, furnace is made a vacuum, molds lowered, then Ar gas refills the furnace, with pressure difference serving to inject the liquid metal into the molds



Fuel Redistribution

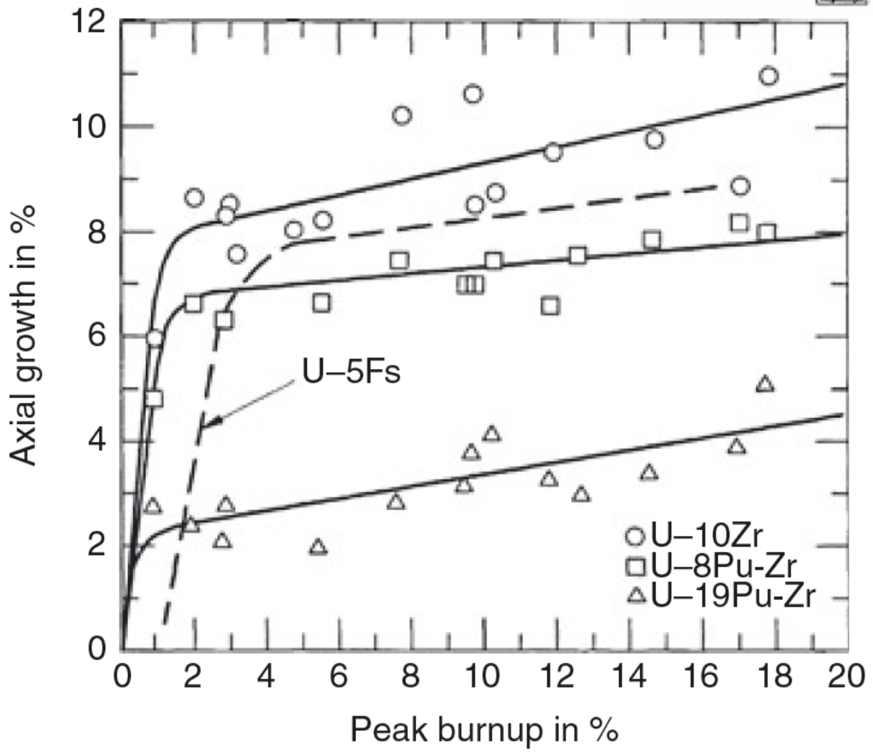
- Zr diffuses via Soret diffusion up the temperature gradient
- Zr also possesses different solubilities in each phase of U
- This leads to distinct zones of Zr content in radial rings
- A) gamma phase, high Zr content, B) beta phase-ish, low Zr content, C) alpha/delta phase, as-fabrication Zr content
- Lower Zr content = lower melting temperature
- Also, different elastic and thermal properties for each phase and each Zr content





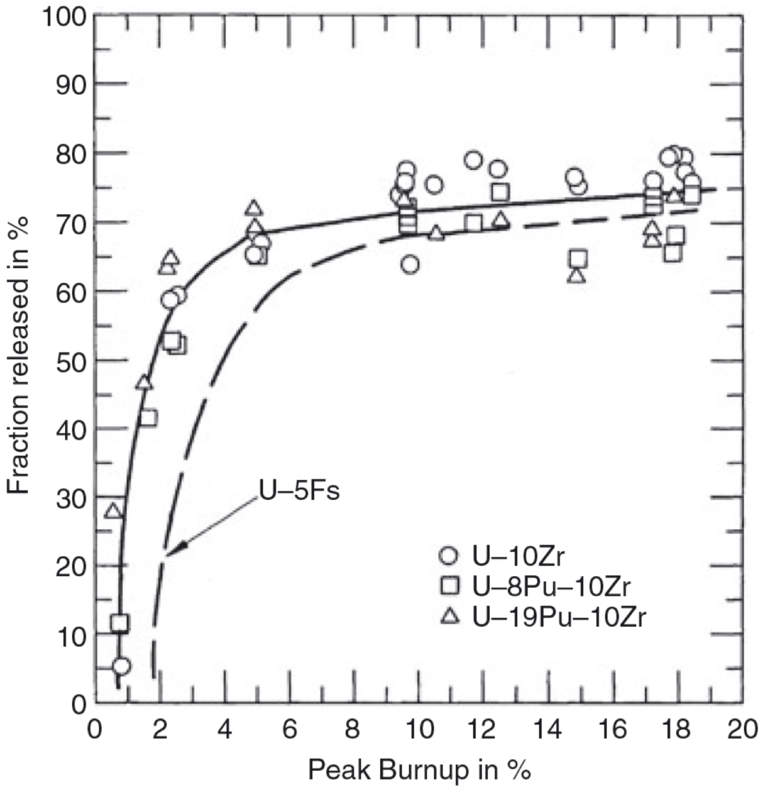
Swelling of Metallic Fuel

- While oxide fuels well a few percent, metallic fuels swell by as much as 30 %
- This swelling occurs very rapidly, e.g. in the first few percent burnup, then displays a linear increase
- Fuel swells anisotropically: more radially than axially
- Swelling is accounted for by low smear density
- Swelling rate decreases due to fission gas bubble interconnection and release
- Low slope of swelling vs burnup for burnup > 5% is largely due to solid fission product generation



Fission Gas Release

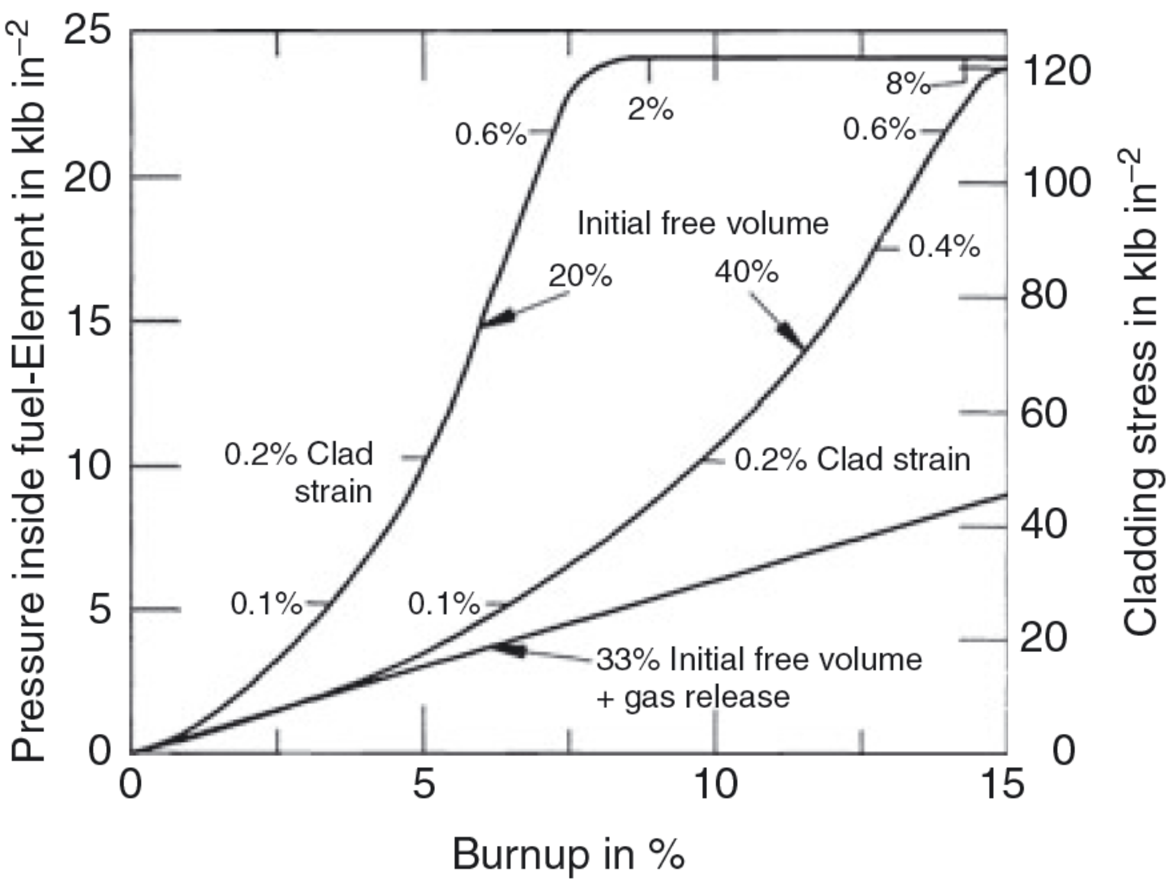
- Gas release is not wholly understood, but largely presumed to be intragranular bubble growth and interconnection
- Bubble interconnection, and thus fission gas release, typically occurs within the first 3-5 percent burnup
- There exists limited experimental data, but this is a key phenomenon affecting fuel swelling and performance
- Gas released into the plenum leads to internal pressure increase



$$\bar{\sigma}_\theta = \frac{pR}{\delta} \qquad \bar{\sigma}_z = \frac{pR}{2\delta} \qquad \bar{\sigma}_r = -\frac{1}{2}p$$

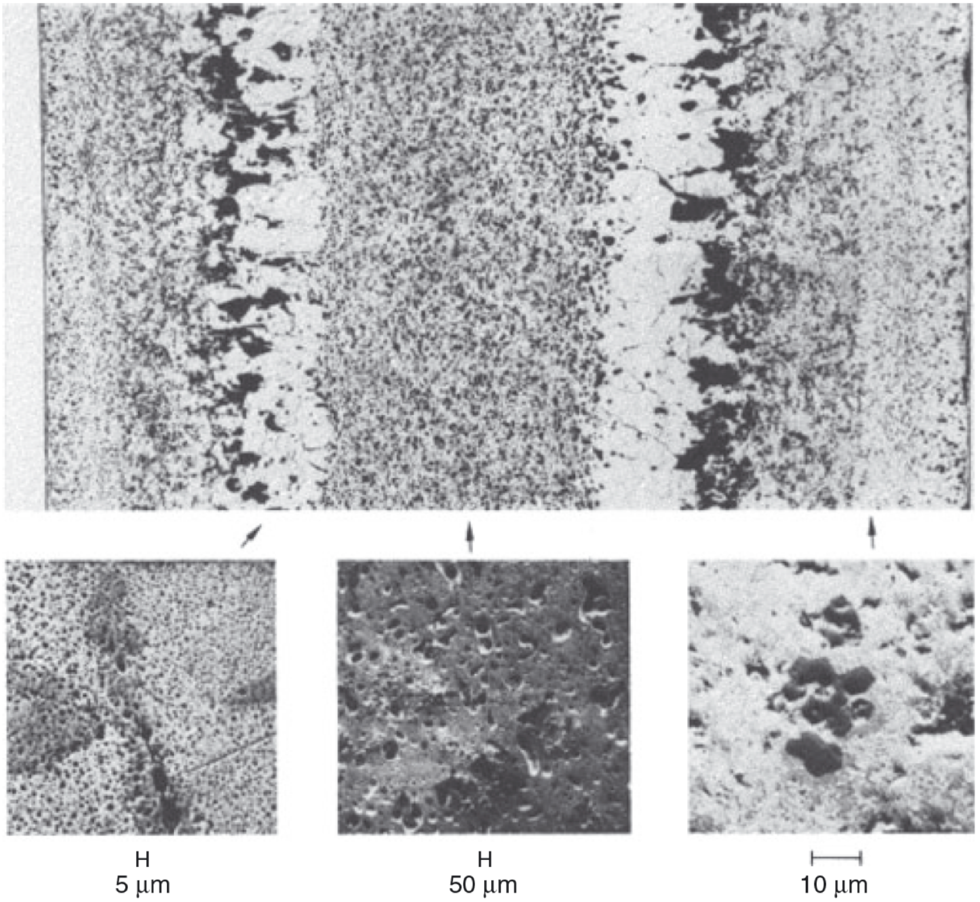
Cladding Strain

- Since most of the fission gas was released from the fuel at ~30% swelling, it was clear that the pressure inside the fuel element could be decreased by providing a plenum for this gas
- Thus, with smear density ~75% and a large plenum, allow for both swelling and gas release without excessive stress on cladding

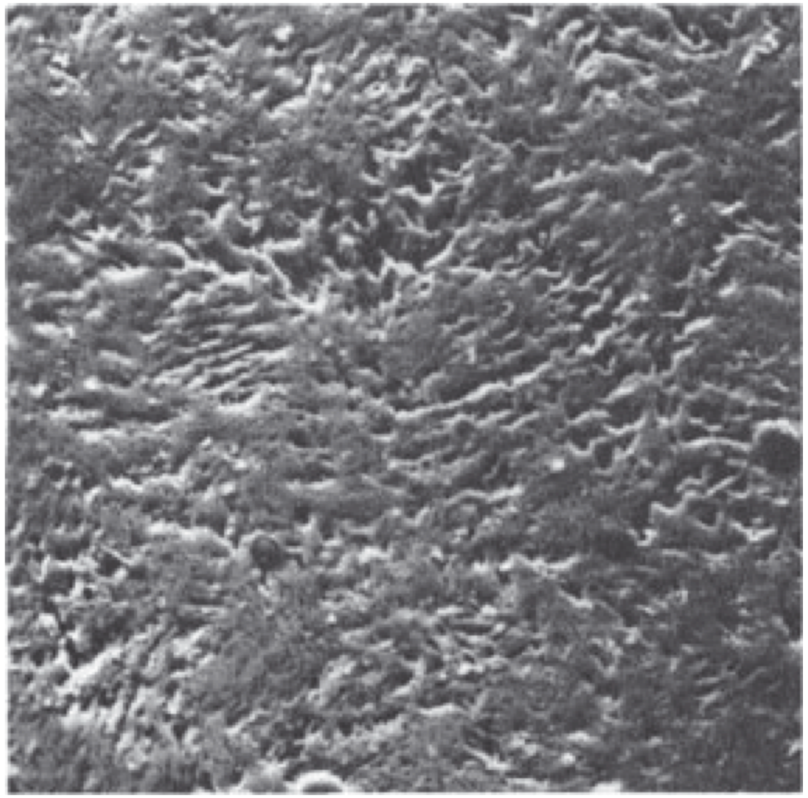


Radially Varying Bubble Morphologies

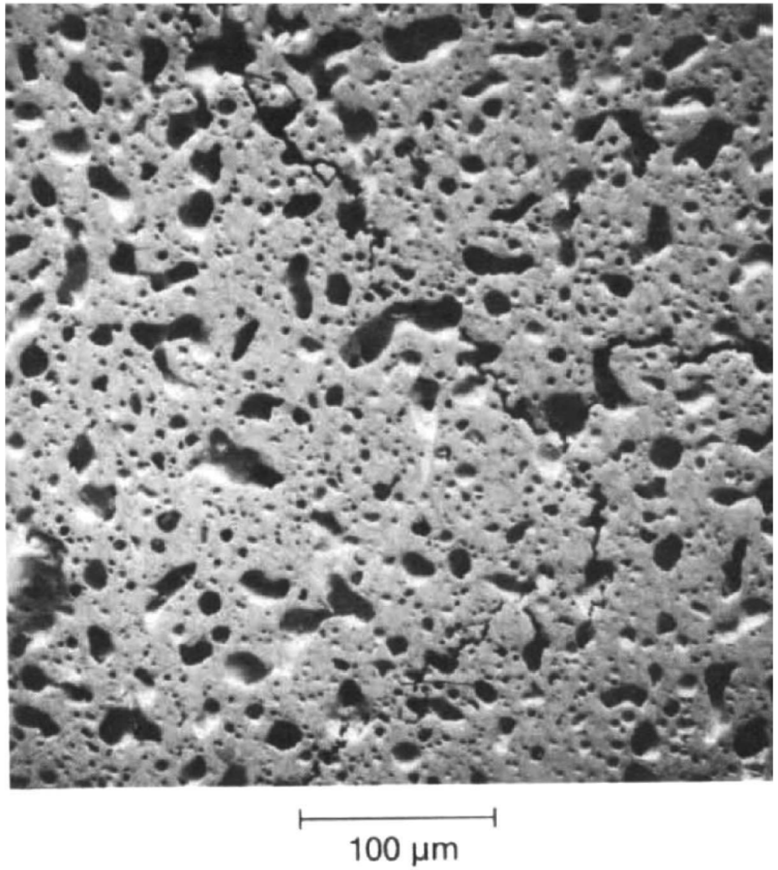
- In the center part of the fuel pin, where the γ-phase predominates, large gas bubbles form; this is indicative of a higher plasticity of the fuel
- Whereas at lower temperatures, where the U α-phase predominates, the characteristic tearing-type porosity is evident
- The fission gas pressure in the center may result in a near-biaxial loading of the peripheral shell, the radial stress component being twice the axial component
- This stress effect results in a larger diametral than axial strain, and hence anisotropic swelling



Fission Gas Bubble Morphology in alpha vs gamma U

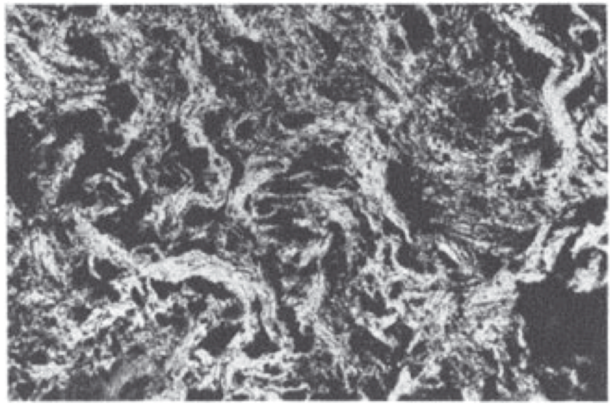


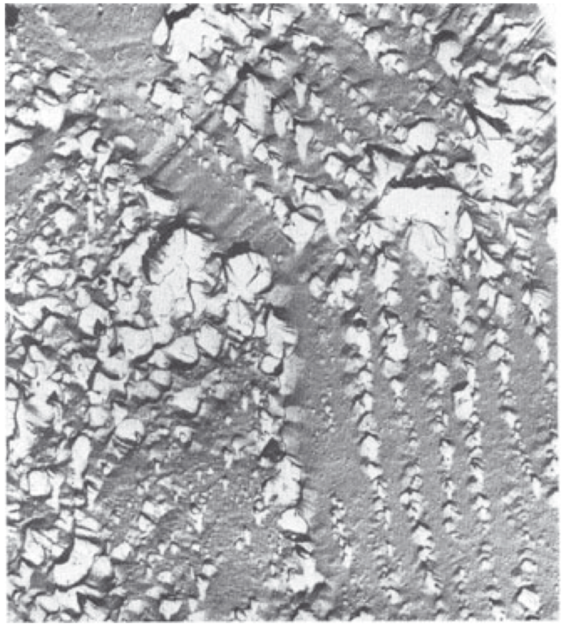
alpha



gamma

alpha U Tearing

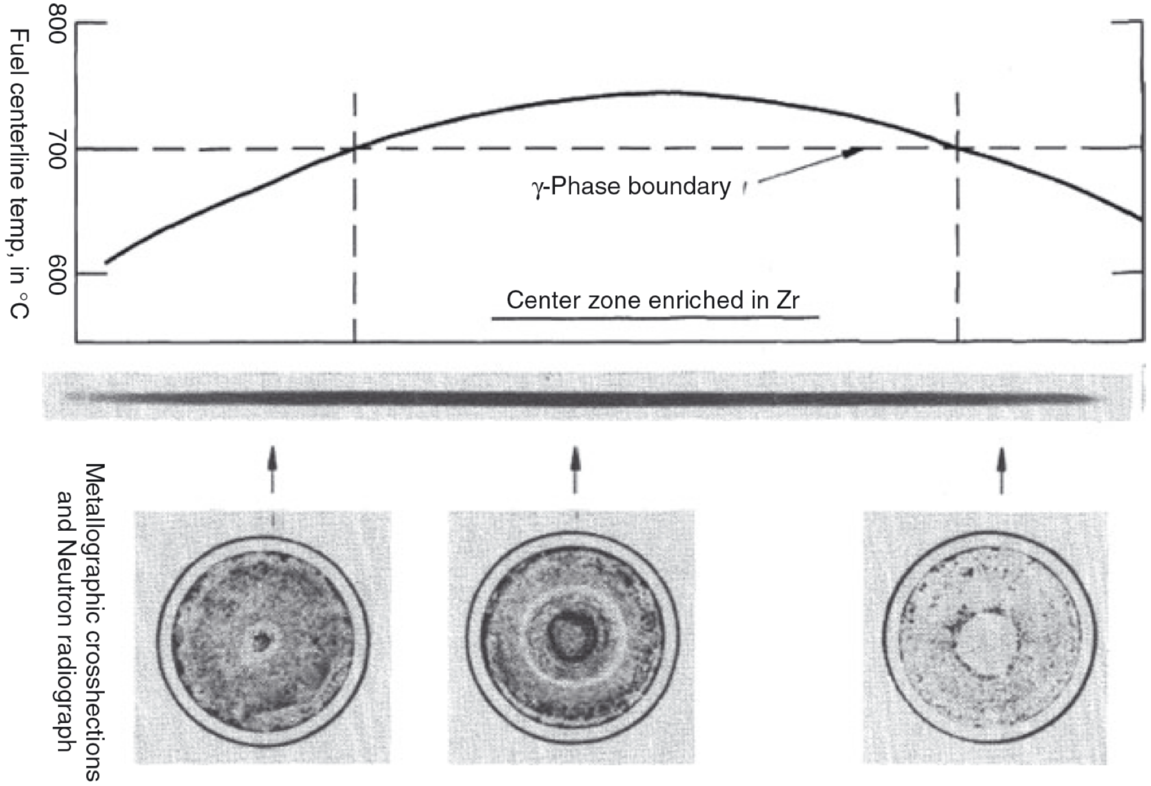




- Between 400 and 600 C, alpha U swelling is overwhelmingly dominated by cavitation
- Cavitation swelling is characterized by large irregular cavities that form by mechanical tearing at grain and sub-grain boundaries, resulting in a very deformed “swirled” microstructure
- In addition to these large cavities, many small cavities, or tears, develop within the α grains, particularly in the 500–600 C range
- These intergranular cavities are crystallographically aligned and appear to be related to twin boundaries

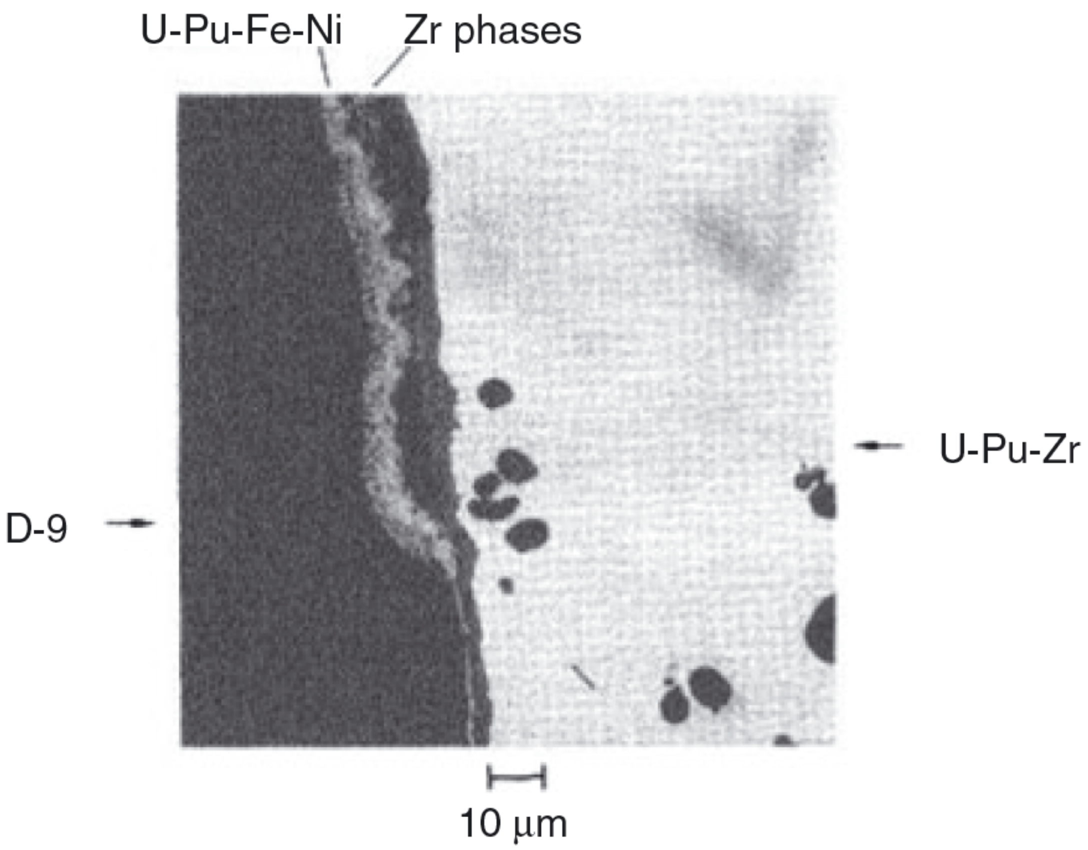
Axial Varying Microstructure

- The location of the radial zones essentially follows isotherms in the fuel which are determined by the various phase boundaries of the alloy
- In the usual situation of upward coolant flow and a cosine-shaped axial power profile in the fuel, the peak fuel temperature occurs between the center and top of the fuel column



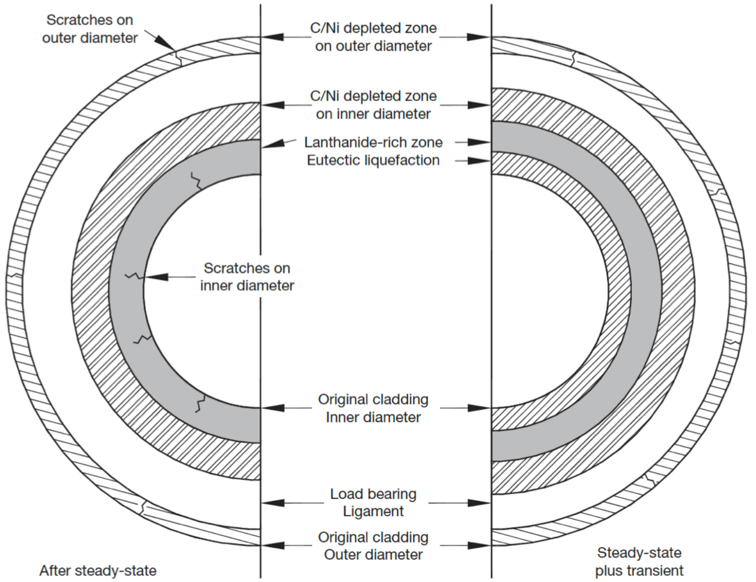
Fuel-Clad Chemical Interaction

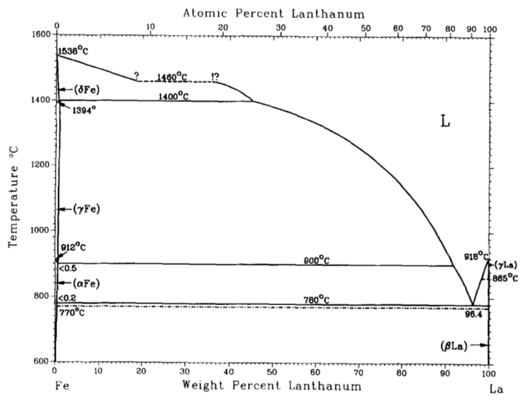
- FCCI in an all-metallic fuel element is in essence a complex multicomponent diffusion problem
- At least five major constituents participate in the diffusion process, in addition to minor alloy components such as C, N and O, as well as fission products
- The potential problem of interdiffusion of fuel and cladding components is essentially twofold: (i) a weakening of the cladding's mechanical properties; and (ii) the formation of relatively low melting point compositions in the fuel

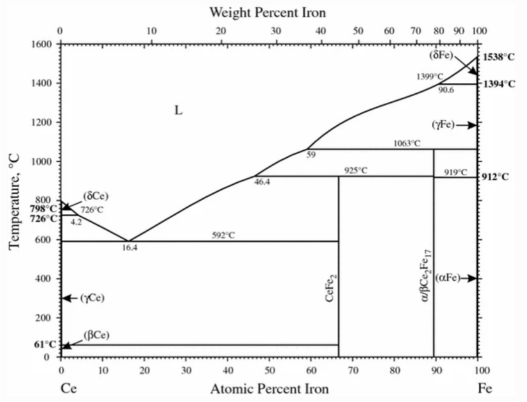


Cladding wastage mechanisms

- Prior to accumulation of lanthanide FPs at the fuel–cladding interface (due to FPs migration), FCCI is characterized by a ferritic layer formation, which is a result of Ni depletion in austenitic cladding or decarburization of the martensitic cladding
- Severe wastage is due to interaction of cladding with FPs, generating eutectic low melting phases
- Fe-96Ln has a melting point of 770˚C, Fe-92Ce has a melting point of 590˚C

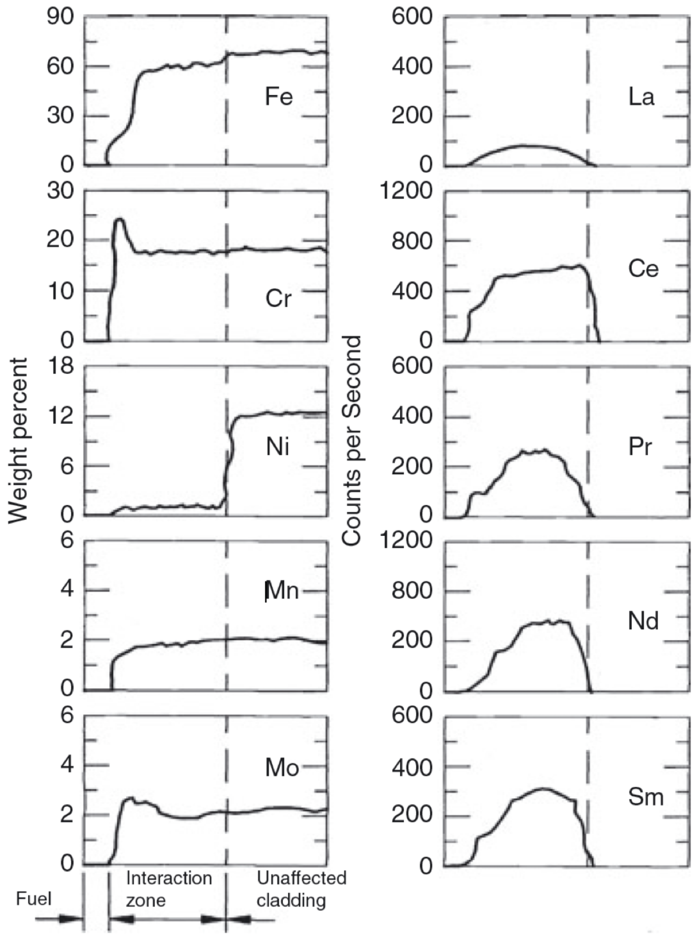






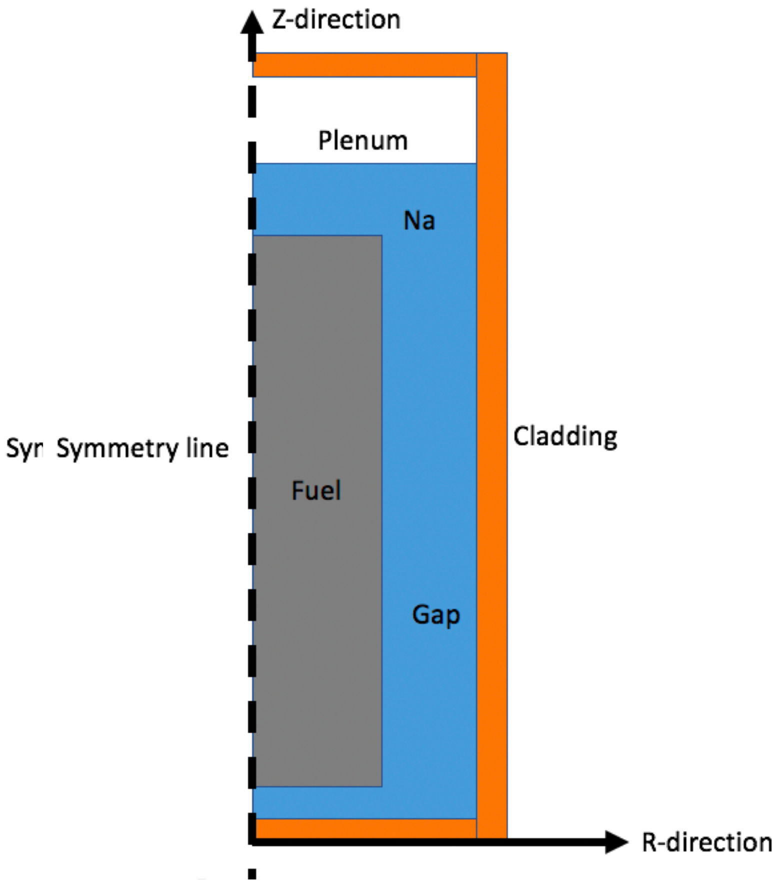
Chemical Species Diffusion and Interaction

- Lanthanides (Ln) ultimately control FCCI, and their presence at the internal cladding surface depends not only on burnup but also very strongly on their radial migration in the fuel
- Nd and Ce are two of the most commonly found Ln in FCCI regions
- Fission products diffuse into the Fe-based cladding and segregate along grain boundaries, further weakening the cladding
- It is key to develop transport models for Ln species through the fuel, in order to develop a source term for FCCI, but very little is known about Ln transport in metallic fuel



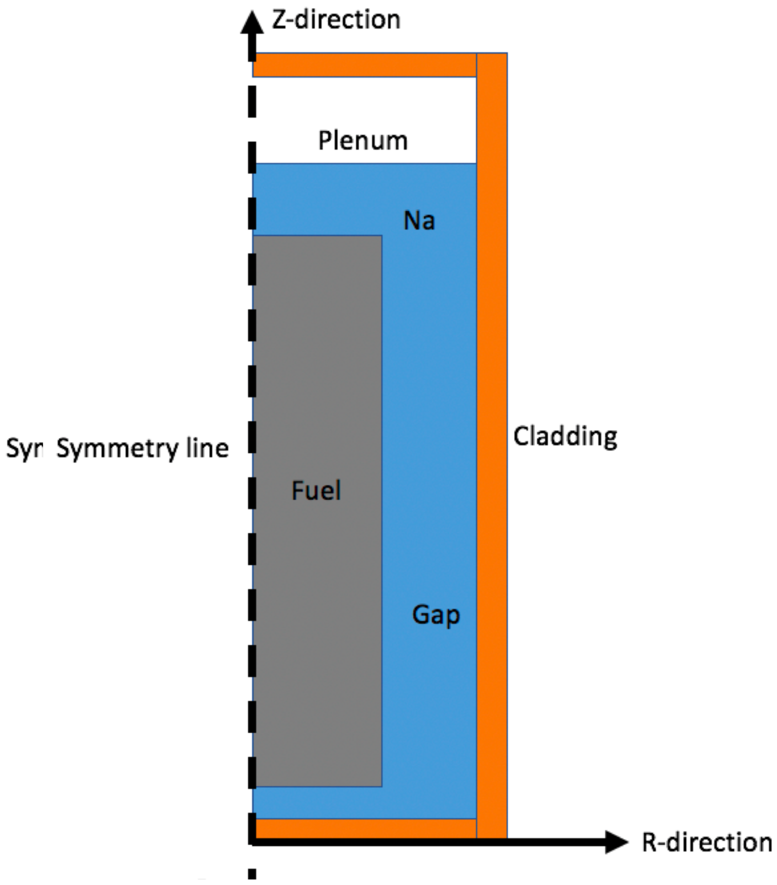
Metallic Fuel Performance Modeling - BISON

- The mechanical performance of the fuel is governed by a model that represents the elastic moduli as functions of temperature, porosity, and zirconium and plutonium content
- A creep model that is a function of fission rate, temperature, and porosity is also used
- Eigen strains (stress free strains) are modeled by constant thermal expansion along with gaseous and solid fission product swelling models
- The solid swelling model is a straight-forward model directly proportional to fission rate
- The gaseous swelling model comes from a force balance applied to a bubble with the metallic fuel and generates porosity at all material points (quadrature points) in the domain



Metallic Fuel Performance Modeling

- The thermal conductivity model in the fuel is a function of temperature, porosity, and zirconium and plutonium content
- The heat capacity models are functions of temperature and alloy phase
- The fission gas model is simply based on fission rate for gas generation and the fission gas released is governed by the value of porosity
- Cladding mechanical response is governed by constant elastic moduli and thermal expansion, with thermal and irradiation creep
- Thermal conductivity and heat capacity models are functions of temperature
- The redistribution of zirconium is governed by Fickian and Soret diffusion



Metallic Fuel Performance Modeling

- Compare BISON to EBR-II experiments

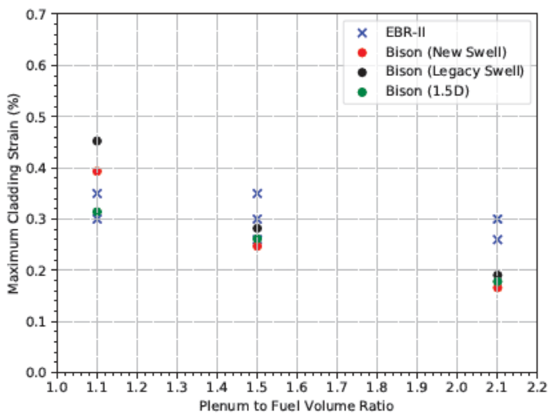


Figure 2: Comparison of BISON results and EBR-II experimental data for clad hoop strain vs. plenum/fuel volume ratio.

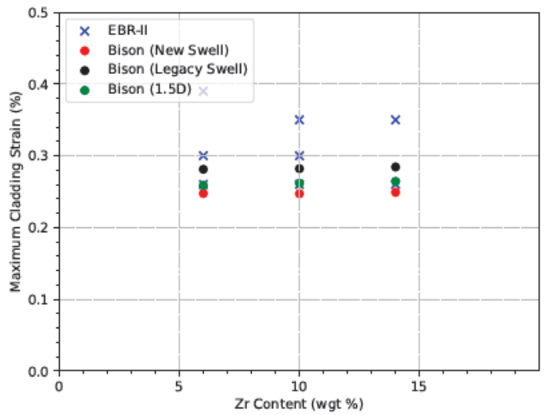


Figure 3: Comparison of BISON results and EBR-II experimental data for clad hoop strain vs. Zr content.

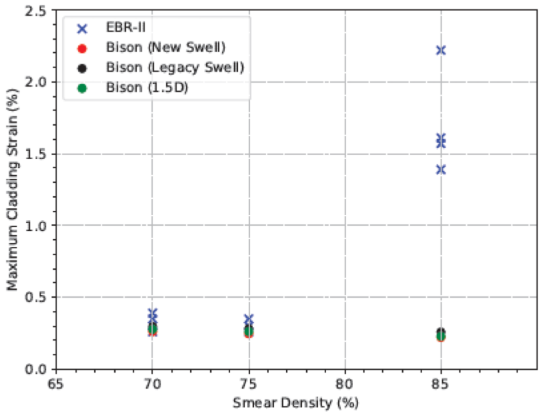


Figure 4: Comparison of BISON results and EBR-II experimental data for clad hoop strain vs. smear density.

Other Metallic Fuel Codes

- LIFEMETAL
  - ANL code that has evolved from the LIFE series of codes that perform steady-state and transient analyses for the thermal, mechanical, and irradiation behavior of nuclear fuel, and was originally developed for UO2
- ALFUS
  - ALloyed Fuel Unified Simulator, Japanese development (CRIEPI), is an irradiation behavior analysis code for metallic fast reactor fuel
- FAST
  - the current NRC thermal-mechanical fuel performance code that is the next evolution of FRAPCON

Incorporation of lower length scale information

- Surface energy
- Porosity interconnection threshold
- Fission gas bubble number density
- Alpha tearing/initial porosity

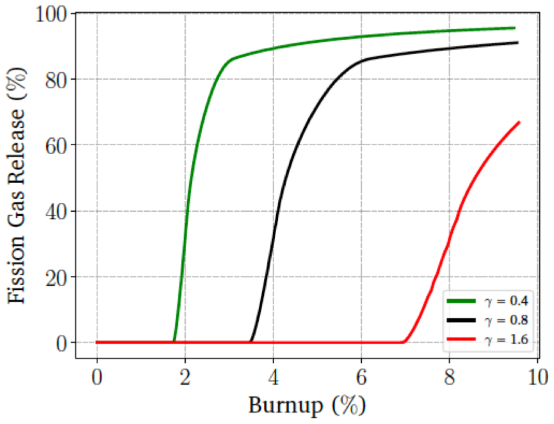


Figure 5: Comparison of fission gas release as a function of fuel surface tension.

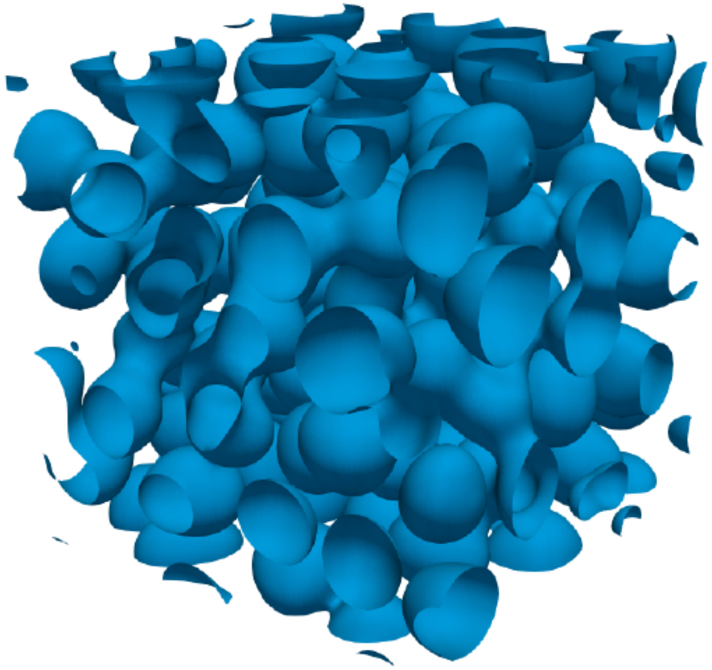


Figure 7: Final microstructure of interconnected bubbles.

Summary

- Metal fuel can go to high burnup, good accident performance, inherent safety, easy to make
- Complex phase and composition environment
- Rapid, substantial swelling, followed by fission gas release
- Fission gas bubble behavior is very phase dependent
- Observe constituent redistribution radially, in addition to axially varying microstructure
- FCCI is a major factor in cladding failure, dependent upon thermodynamics and kinetics of a number of lanthanides, actinides and transition metals
- We are trying to model all of this!