NE 591: Advanced Reactor Materials

Fall 2021

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Last Time

- Wrapped up TRISO particle-based reactors
- Walked through a number of different fuel failure mechanisms
- Current state of TRISO fuel performance modeling
- Advanced concepts in TRISO

Questions

- tau is burnup
- k1d – fission products in solid solution
- k1p – fission product precipitates
- k2p – porosity
- k4r – irradiation damage

$$k_{\text{fuel}} = K_{1\text{d}} K_{1\text{p}} K_{2\text{p}} K_{4\text{r}} k_{0,\text{fuel}}$$

$$K_{1\text{d}} = \left(\frac{1.09}{\tau^{3.265}} + \frac{0.0643}{\sqrt{\tau}}\sqrt{T_{\text{kern}}}\right) \arctan\left(\frac{1}{\frac{1.09}{\tau^{3.265}} + \frac{0.0643}{\sqrt{\tau}}\sqrt{T_{\text{kern}}}}\right)$$

$$K_{1\text{p}} = 1 + \frac{0.019}{3 - 0.019\tau} \frac{1}{\left(1 + \exp\left(-\frac{T_{\text{kern}} - 1200}{100}\right)\right)}$$

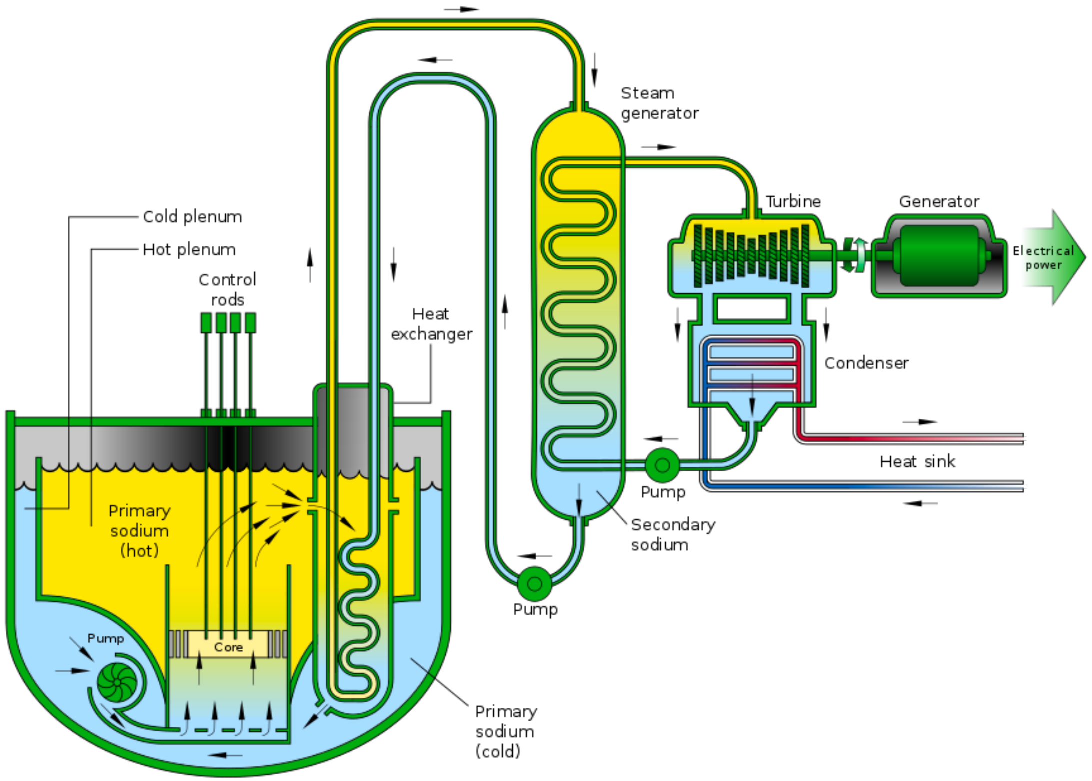
$$K_{2\text{p}} = \frac{1 - P}{1 + 2P}$$

$$K_{4\text{r}} = 1 - \frac{0.2}{1 + \exp\left(\frac{T_{\text{kern}} - 900}{80}\right)}(1 - \exp(-\tau))$$

SODIUM COOLED FAST REACTORS

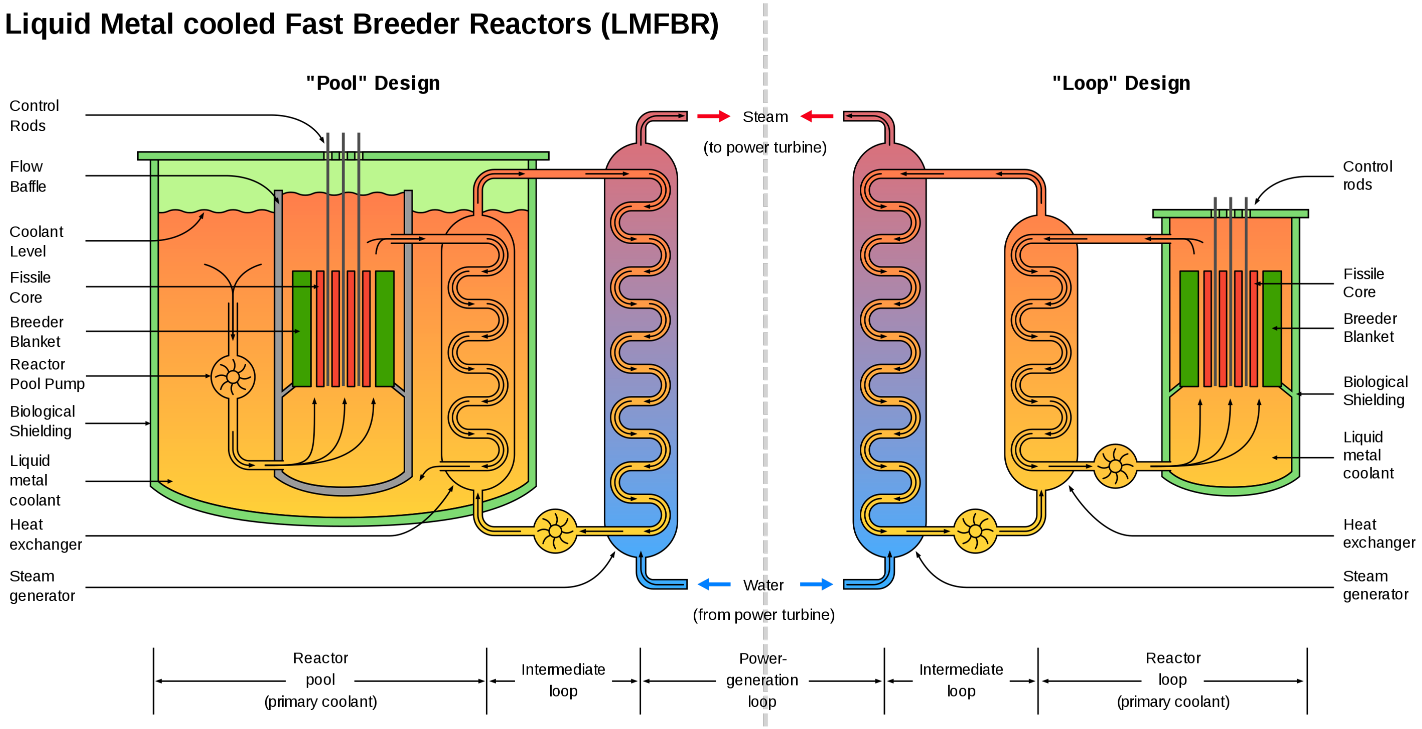
Sodium Cooled Fast Reactors (SFRs)

- Two main families:
  - Liquid metal cooled reactor, which utilizes MOX fuel
  - Integral fast reactor, which utilizes metallic fuel
- SFRs are designed for management of high-level wastes and management of plutonium and other actinides
- Important safety features of the system include a long thermal response time, a large margin to coolant boiling, a primary system that operates near atmospheric pressure, etc.



SFRs

- Pool vs Loop
  - In the pool type, the primary coolant is entirely contained in the main reactor vessel, which therefore includes not only the reactor core but also a heat exchanger (US EBR-2, French Phenix)
  - In the loop type, the heat exchangers are external to the reactor tank (French Rapsodie, BN-350)



SFRs

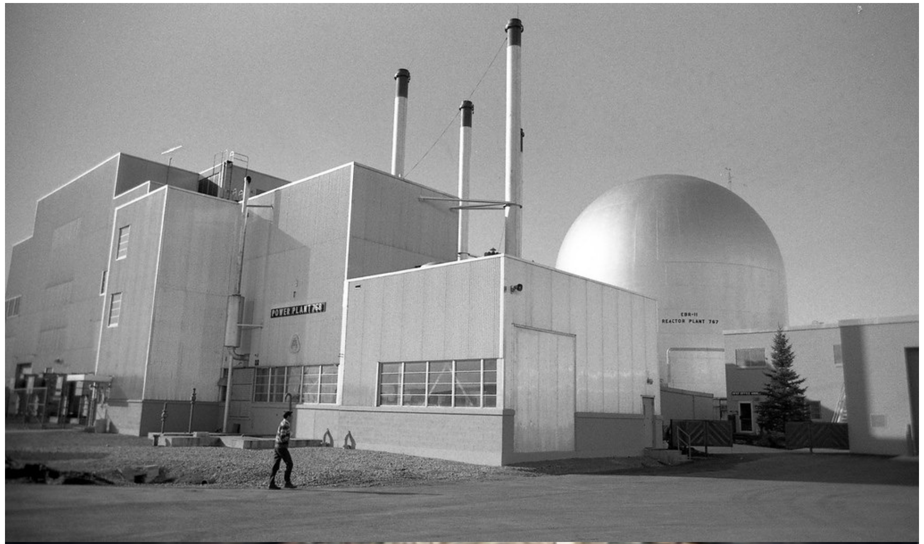
- Currently operating SFRs:
  - BN-600 (Russia), BN-800 (Russia), CEFR (China), FBTR (India), Joyo (Japan)
- Benefits of liquid metal coolants:
  - metals atoms are weak neutron moderators, allowing for fast neutron spectrum
  - sodium melts at 371K and boils at 1156K, a total temperature range of 785K between solid and gas states, whereas water is only 100K at atmospheric pressure
  - boiling temperature is well above operational temperatures
  - sodium is non-corrosive, compatible with metallic fuel
  - sodium has higher thermal conductivity than water
- Drawbacks:
  - sodium catches on fire in air, reacting with water to form hydrogen gas
  - requires special precautions to prevent and suppress fires and limit sodium leaks

History of SFRs

- Sodium-cooled fast reactors were among the first to be designed, built, and operated
- EBR-I
  - ANL-W, INL
  - Startup in Dec. 1950
  - The world's first breeder reactor
  - Demonstrated the ability to breed fuel
  - Utilized metallic fuel and NaK coolant

Key SFRs in the US

- EBR-II (1965-1994)
  - Purpose was to demonstrate a complete breeder-reactor power plant with on-site reprocessing of solid metallic fuel
  - It demonstrated its ability to self-cool its fuel through natural convection of the sodium coolant during the decay heat period following the shutdown
- FFTF (1980-1992)
  - Operated as a national research facility to test various aspects of commercial reactor design and operation, especially relating to breeder reactors
- Large portions of current knowledge on metal fuel and fast flux environment derive from these reactors



Increased Safety

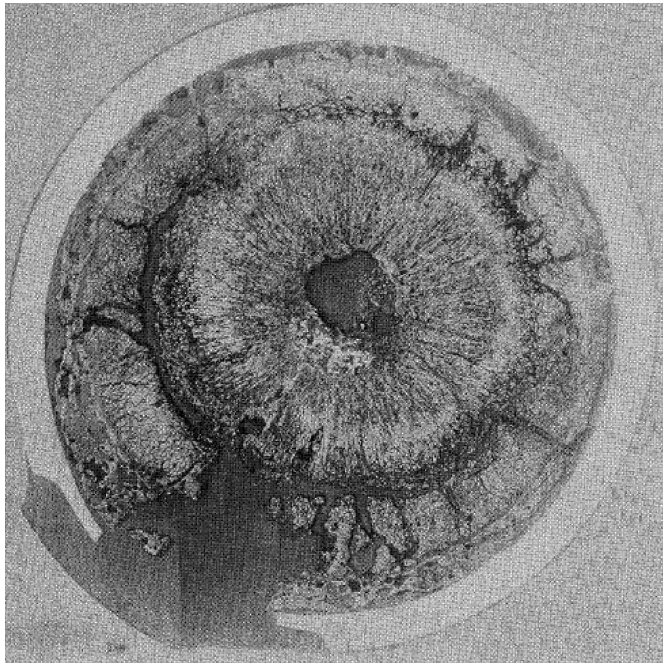
- Metal fuel has excellent transient capabilities
- The metal fuel itself does not impose any restrictions on transient operations or load-following capabilities
- The robustness of metal fuel is illustrated by the following sample history of a typical driver fuel irradiated during the EBR-II inherent passive safety tests conducted in 1986:

- 40 start-ups and shutdowns
- 5 15% overpower transients
- 3 60% overpower transients
- 45 loss-of-flow (LOF) and loss-of-heat-sink tests including a LOF test from 100% power without scram

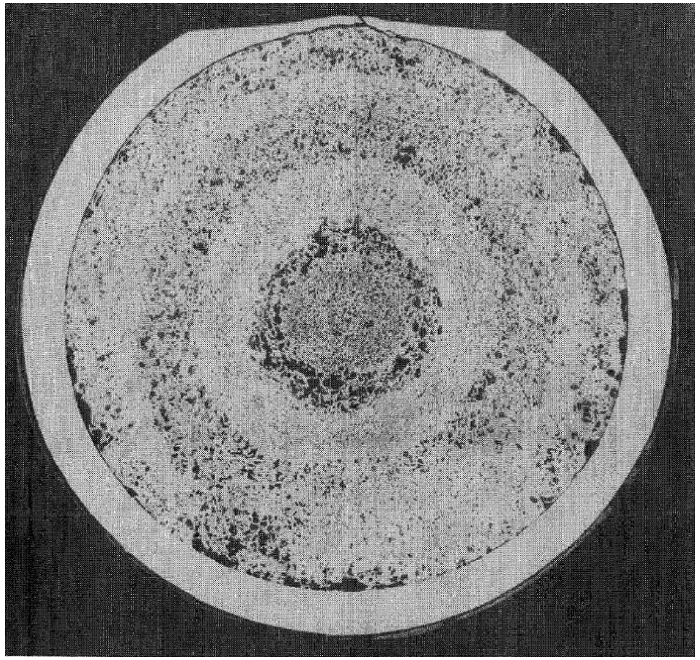
Increased Safety

- Metal fuel also has benign run beyond cladding breach (RBCB) performance characteristics
- After 150+ days of operation with a cladding breach, metallic fuel retained shaped and did not exacerbate the breach

oxide



metal



Inherent Safety

- Although the metal fuel melting temperature is much lower than that of oxide fuel, it is also much more difficult to raise the fuel temperature because of the 10X higher thermal conductivity
- Thus, operating margins in terms of power can, in fact, be greater for the metal core than for oxide
- The high thermal conductivity and the thermal inertia of the large sodium pool can shut the reactor down during very severe accident situations, without depending on human intervention
- One experiment in EBR II (LOFWS) simulated station blackout, combined with loss of emergency power supply, failure of primary and secondary shutdown systems, and no operator action – a worst case
- The rising coolant temperature during accident causes thermal expansion of the core components which enhances the neutron leakages and hence slowing down the chain reaction
- Due to this negative reactivity feedback, the reactor power is shut down all by itself and the coolant temperature rise stops

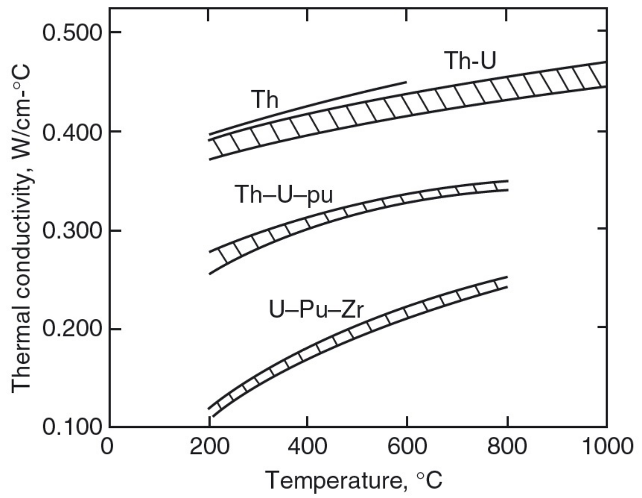
Summary of SFR Usage

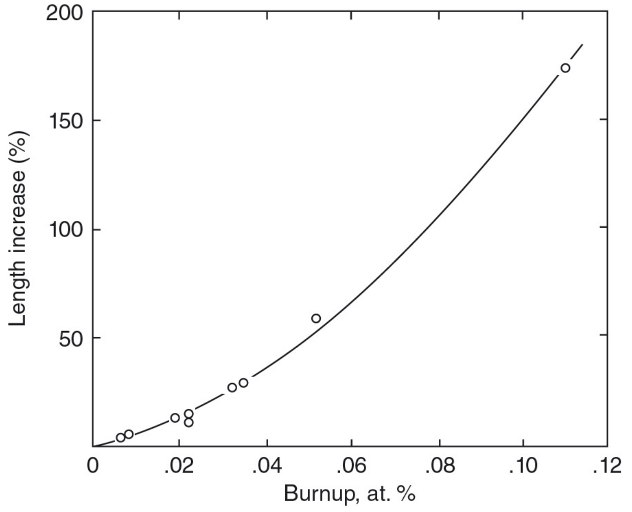
Model	Country	Thermal power (MW)	Electric power (MW)	Year of commission	Year of decommission	Notes
BN-350	Soviet Union		135	1973	1999	Was used to power a water de-salination plant.
BN-600	Soviet Union		600	1980	Operational	Together with the BN-800, one of only two commercial fast reactors in the world.
BN-800	Soviet Union/ Russia	2100	880	2015	Operational	Together with the BN-600, one of only two commercial fast reactors in the world.
BN-1200	Russia	2900	1220	*2036*	Not yet constructed	In development. Will be followed by BN-1200M as a model for export.
CEFR	China	65	20	2012	Operational	
CRBRP	United States	1000	350	Never built	Never built	
EBR-1	United States	1.4	0.2	1950	1964	
EBR-2	United States	62.5	20	1965	1994	
Fermi 1	United States	200	69	1963	1975	
Sodium Reactor Experiment	United States	20	65	1957	1964	
S1G	United States					United States naval reactors
S2G	United States					United States naval reactors
Fast Flux Test Facility	United States					
PFR	United Kingdom	500	250	1974	1994	
FBTR	India	40	13.2	1985	Operational	
PFBR	India		500	*2020*	Under construction	Under construction
Monju	Japan	714	280	1995/2010	2010	Suspended for 15 years. Reactivated in 2010, then permanently closed
Jōyō	Japan	150		1971	Operational	
SNR-300	Germany		327	1985	1991	
Rapsodie	France	40	24	1967	1983	
Phénix	France	590	250	1973	2010	
Superphénix	France	3000	1242	1986	1997	Largest SFR ever built. Suffered a terrorist attack during its construction.

METAL FUEL

First Metal Fuels

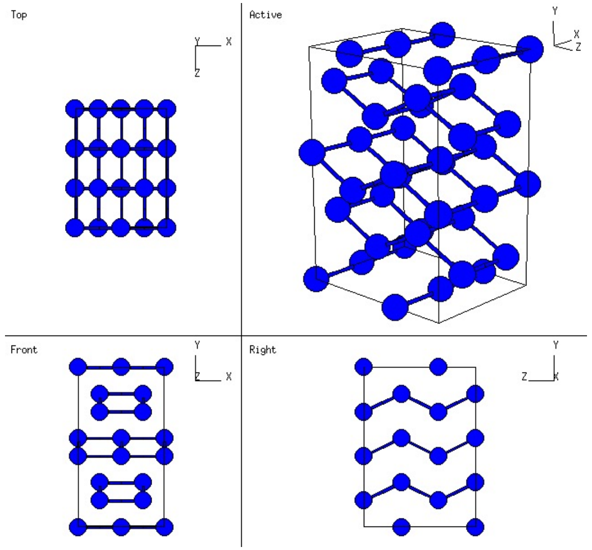
- Starting in the late 1940s, metallic fuels dominated the early development of nuclear reactors because of their key attributes of high fissile-atom density and good thermal conductivity
- Alloys of U, Pu were of interest, and alloys with Th were pursued to increase thermal conductivity
- Initial experiments with pure U metal met with spectacular swelling and growth, as observed in the mid 1950s

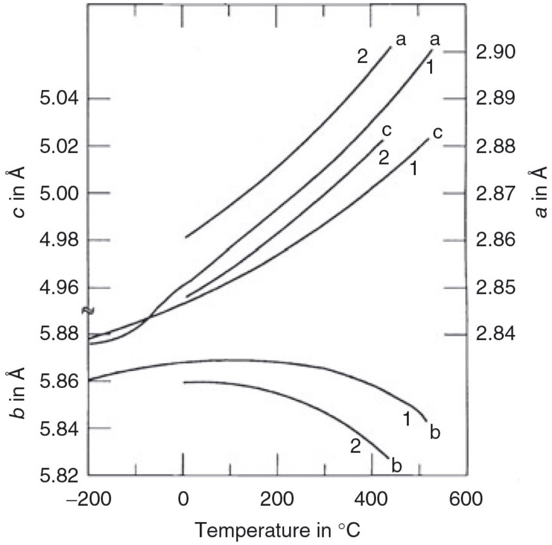


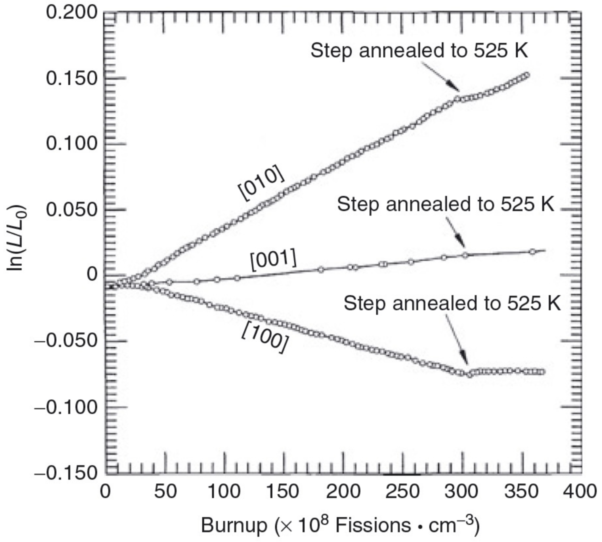


Alpha U

- The most important irradiation characteristic of α-U is its dimensional instability in the form of anisotropic growth and swelling, which results from the anisotropy of the orthorhombic crystal structure
- In addition to anisotropic thermal expansion, irradiation growth occurs anisotropically
- The current theory of this phenomenon is that fission-generated interstitials and vacancies form loops on {010} and {110} planes, respectively, as a result of the anisotropic thermal expansion induced in α-U by thermal spikes in displacement cascades

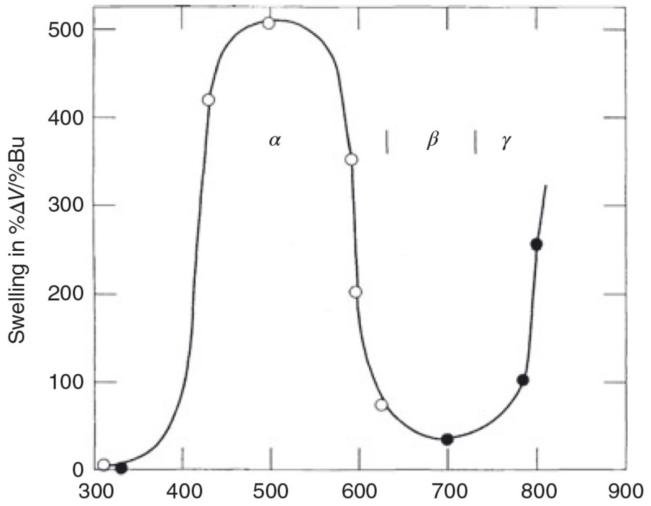


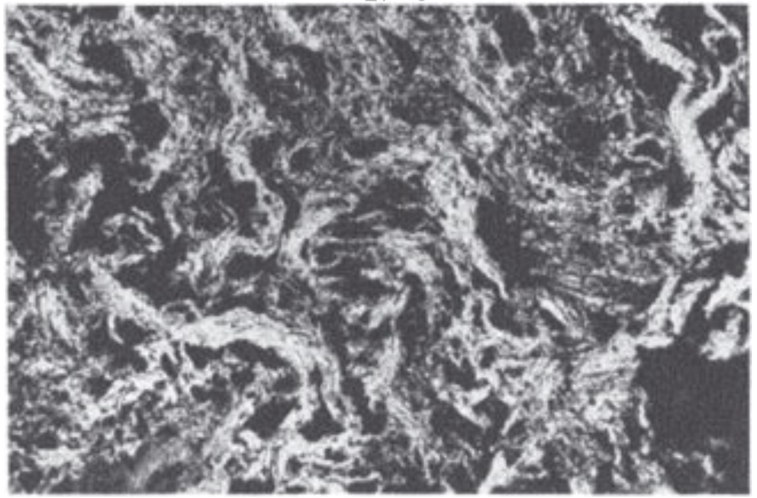




Alpha U Swelling

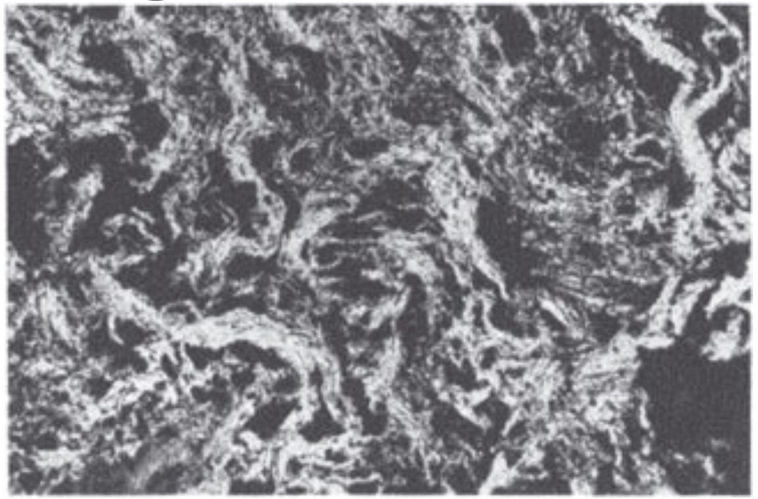
- The temperature dependence of the irradiation growth process takes on a bell-shaped form, due to recombination of point defects
- In a polycrystalline sample, irradiation growth in individual grains results in shape changes and mismatched strains between the individual grains
- The stress developed due to these mismatched strains can be released by plastic deformation at the grain boundaries; this is commonly referred to as tearing or cavitation
- Cavitation swelling dominates between 400 and 600 C

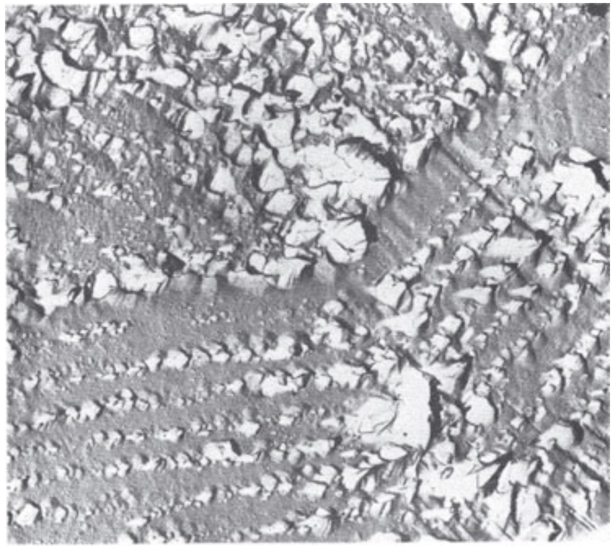




Alpha U Swelling

- Lower temperature regime cavitation swelling results in a swirled microstructure, with irregular cavities formed by tearing of grain boundaries
- At higher temperatures (500-600 C), the cavities along the grain boundaries can be crystallographically aligned, and are potentially related to twinning
- Cavitation can be reduced by minor additions of alloying elements, which can serve to affect the defect evolution in the system





Swelling in Gamma U

- Swelling in the higher temperature phases occurs primarily by growth of fission gas bubbles, and is isotropic
- All phases will also have solid fission product swelling
- Fission gas is effectively insoluble in metallic U, and thus prefers to precipitate
- Sufficiently large bubbles can maintain an equilibrium gas pressure by balancing the surface tension and the external pressure
- This growth is described by the Young-Laplace equation

$$P_i = P_{ex} + \frac{2\gamma}{r}$$

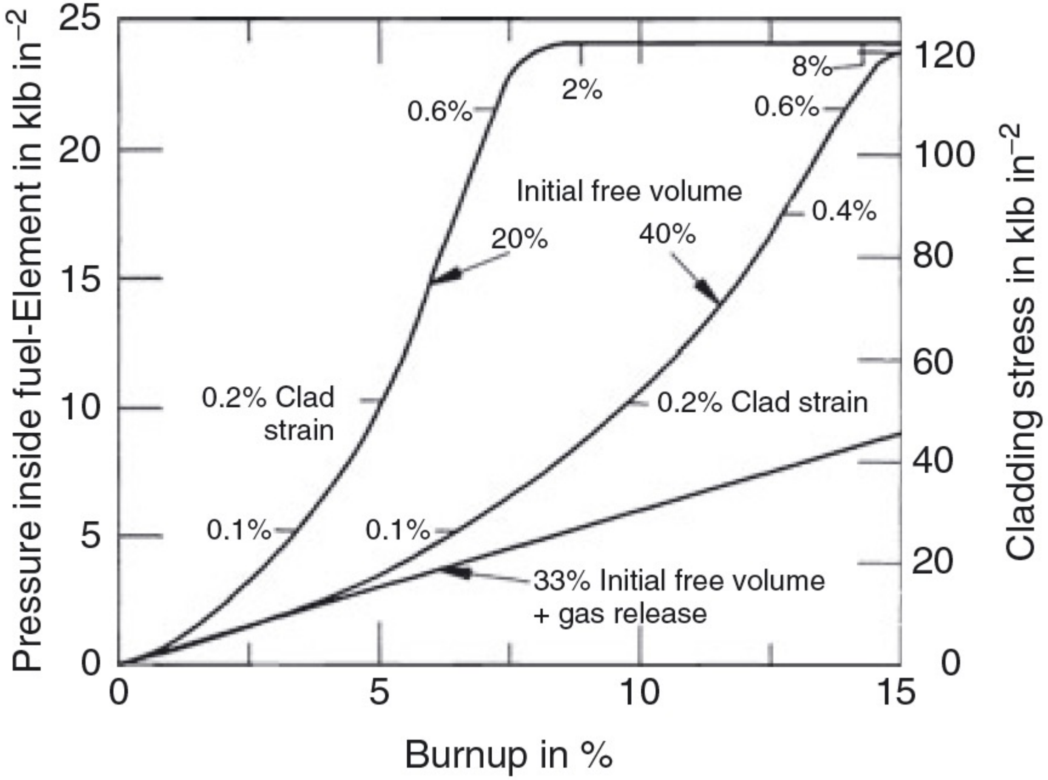
- The external pressure is determined by the plastic flow characteristics of the fuel surrounding the bubble and the mechanical restraint of the cladding
- Fission gas swelling can be substantial, and must be accounted for

Alloying to modify properties

- As with alpha U, small alloying additions were explored to limit the fission gas swelling behavior
- Adjusted U – U alloyed with small amounts of Al, Fe, or Si
- Additions form precipitates and act as point defect sinks, limiting swelling for low burnups
- U-Fissium contained Mo-Ru, with lesser amounts of Rh, Pd, Zr, and Nb, and was the early EBR II fuel
- Higher content alloys have focused on elements that form extensive solid solutions with U in the high-temperature γ-phase, specifically Mo, Zr, Ti, and Nb
- These elements lower the temperature of the cubic γ-phase boundary, allowing more of the fuel to operate in the isotropic γ-phase; they also increase the solidus temperature, raising the margins at which the fuel melts
- The addition of Zr and Mo seem to most substantially reduce the swelling of metallic U and low T and low burnup

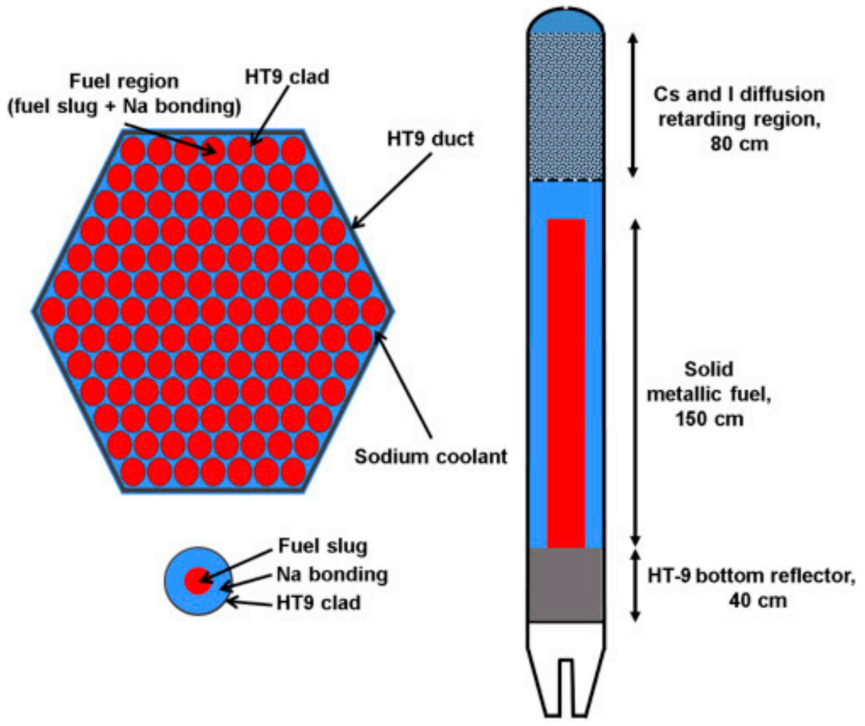
Accommodation of Swelling

- With high-content alloys, calculations of fuel swelling and gas release showed that if 30-40% free volume was afforded inside the cladding, fuel could swell, and when it contacted the cladding, allowing for mechanical strength of cladding to counteract further radial swelling
- It was found that the majority of fission gas is released by about 30% swelling, and by including a large plenum, stresses on cladding could be reduced



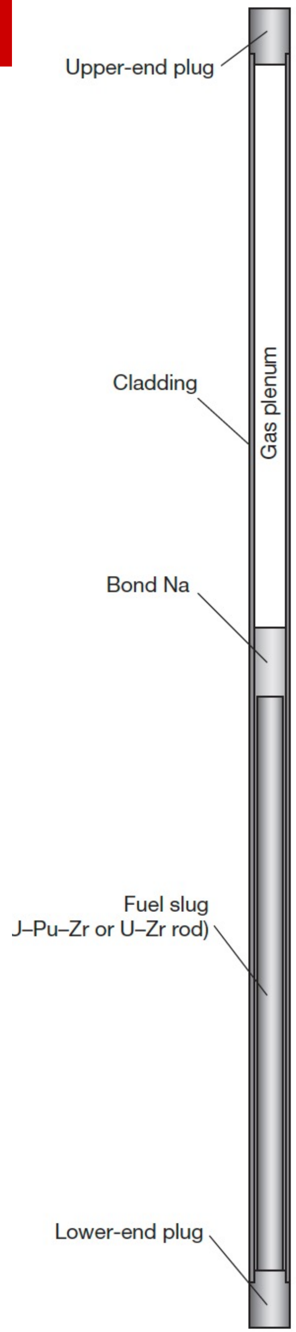
Metal Fuel

- Metal fuels are ideal for fast reactors because they have higher densities of fissile and fertile materials and provide higher breeding ratio
- The burnup of metal fuel in the early days was limited to a few atom percent (at.%) because of the increase in the fuel–cladding mechanical interaction (FCMI) caused by gas swelling of fuel alloys
- Reducing the fuel smear density to about 75% is effective in promoting fission gas release before fuel-cladding contact and in suppressing FCMI



Base Metal Fuel System

- The typical metallic fuel consists of a U(Pu)–10 wt% Zr fuel slug with ~75% smear density
- The cylindrical fuel alloy rod is called a ‘fuel slug
- The annular gap between the fuel slug and the cladding is filled with sodium (bond sodium)
- A relatively large gas plenum (1-1.5x fuel length), which is a space above the fuel slug, is provided to mitigate the pressure of the fission gas accumulating during irradiation

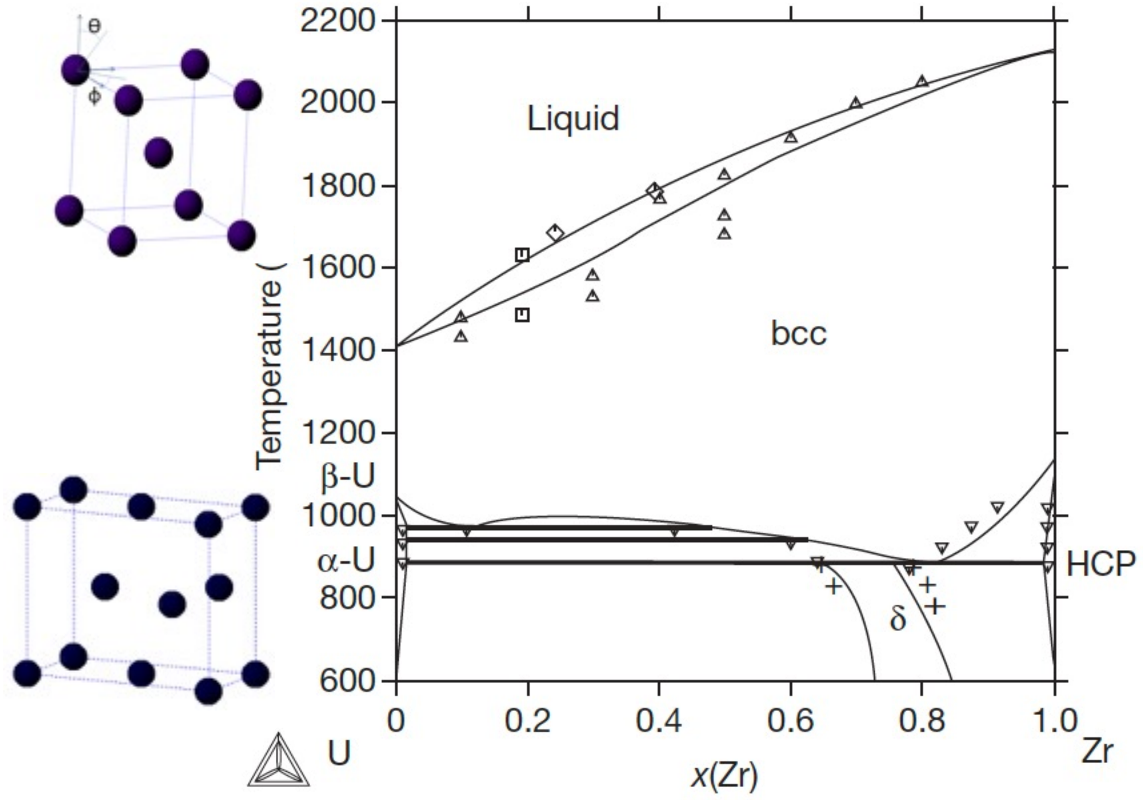


Closing the Fuel Cycle

- An important factor in selecting a fuel form for fast reactors is ease of fuel recycling, including reprocessing and fuel refabrication
- Pyrometallurgical processing, called ‘melt refining,’ can be combined with refining techniques to recycle spent metallic fuel
- This recycling process has been demonstrated for uranium alloy fuels in the 1960s, where fuel alloys were recycled as many as four times, and the fuel was returned to the reactor within 4–6 weeks of its removal from the reactor core
- Will talk more about this later

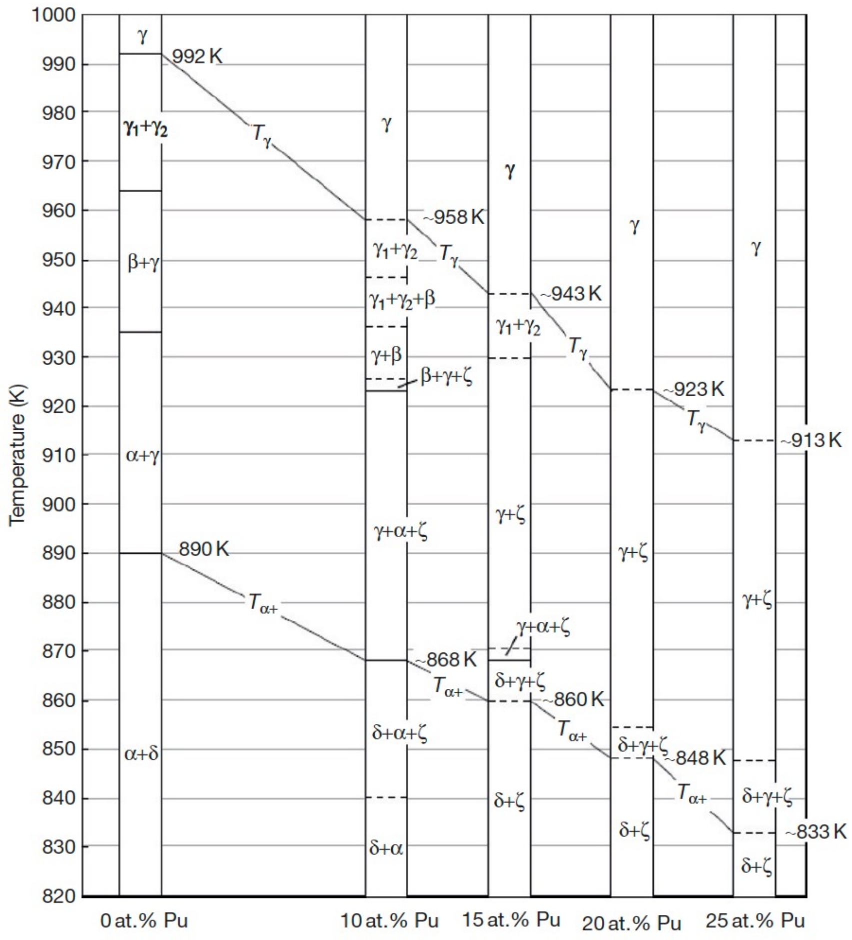
The U-Zr System

- Metallic uranium is alloyed with Zr to stabilize the bcc phase, increase the melting point, improve mechanical properties, and reduce swelling
- Typical composition is 10 wt.%, or approximately 23 atom %
- At relevant temperature ranges in SFRs, multiple phases will be present



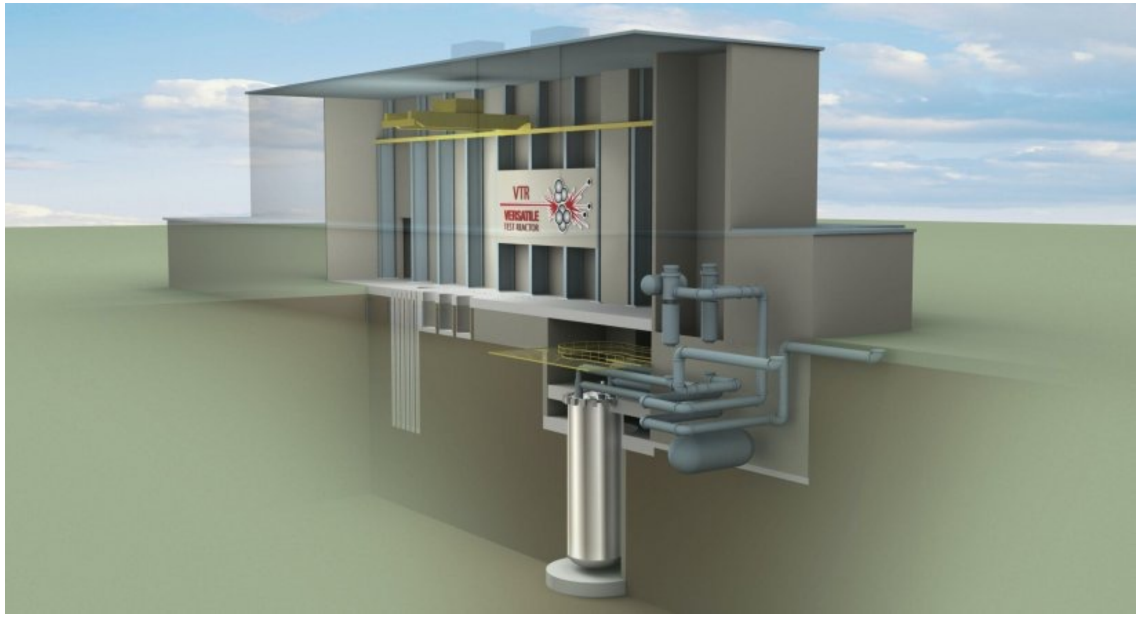
U-Pu-Zr

- Ternary phase diagram isotherms
- γ: Body-centered cubic (bcc), has complete solid solubility for bcc ε-plutonium and bcc β-zirconium
- α: Orthorhombic allotropic modification of uranium that dissolves up to 15 at.% of plutonium, but has limited solubility for zirconium
- β: Tetragonal allotropic modification of uranium that dissolves up to 20 at.% of plutonium, but has limited solubility for zirconium
- η: A high-temperature intermediate phase in the U–Pu binary system that is believed to be tetragonal and has limited solubility for zirconium
- ζ: A complex cubic U–Pu intermediate phase that dissolves up to 5 at.% zirconium
- δ: A hexagonal intermediate phase in the U–Zr system that occurs approximately at the composition UZr2 and has extensive solid solubility for plutonium



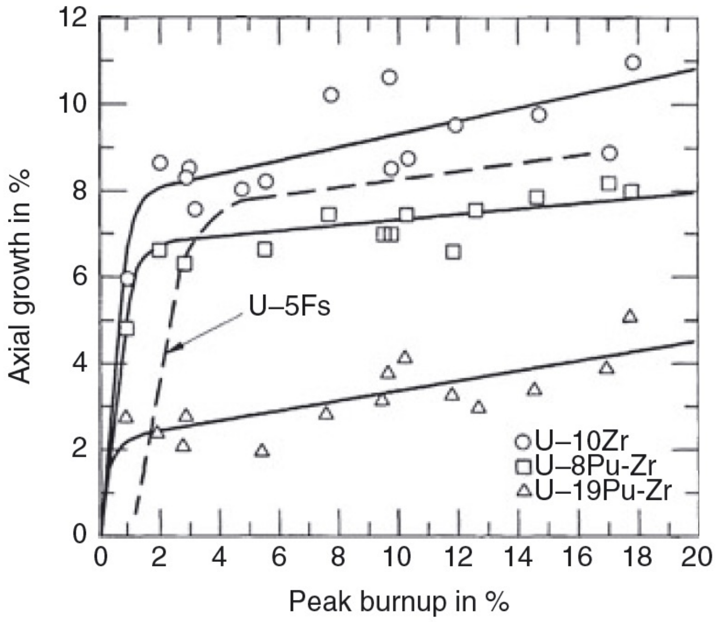
U-Pu-Zr

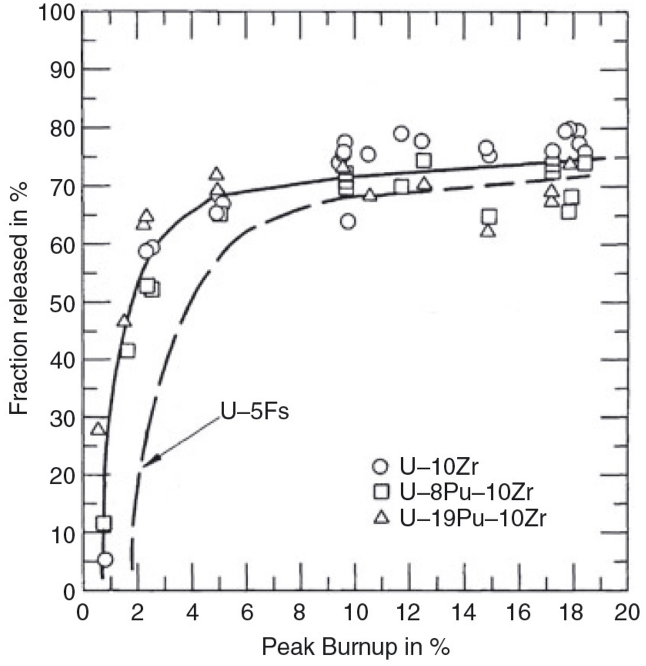
- The U-Pu-Zr system was the fuel choice for the Integral Fast Reactor design of the 1980s, and the current Versatile Test Reactor design
- U-Pu-Zr alloys are the most recent alloys under active exploration, and thus have the most recent experimental data



U-Pu-Zr Swelling

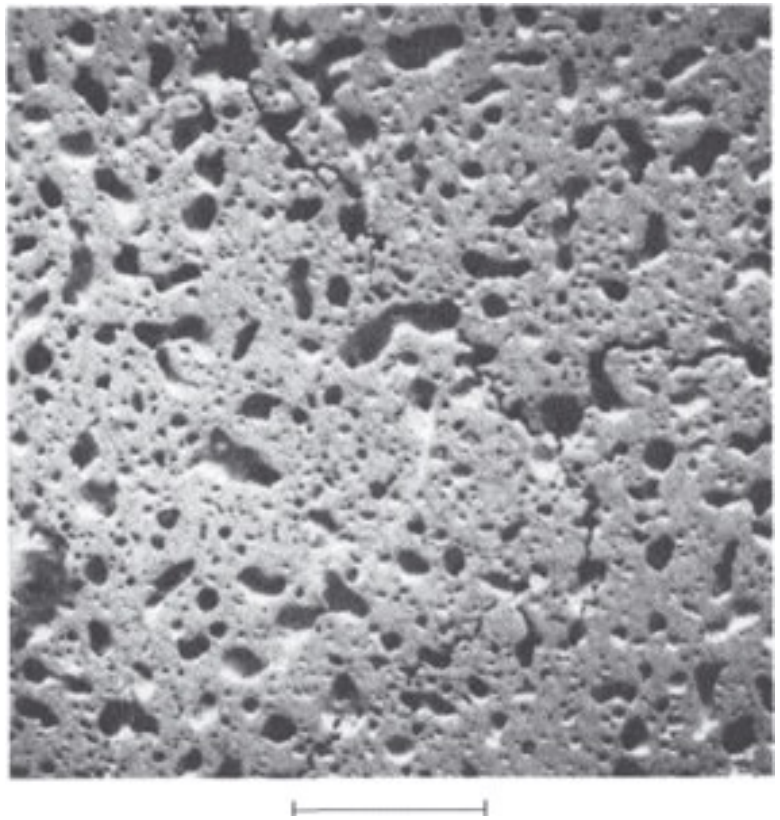
- Swelling is typical of metallic fuels, in that it occurs rapidly, leveling off once in contact with the cladding
- Fission gas release occurs within the first 5% burnup, corresponding with the rapid swelling
- These fuels swell less in the axial direction than in the radial direction, if isotropic swelling is assumed





U-Pu-Zr Swelling

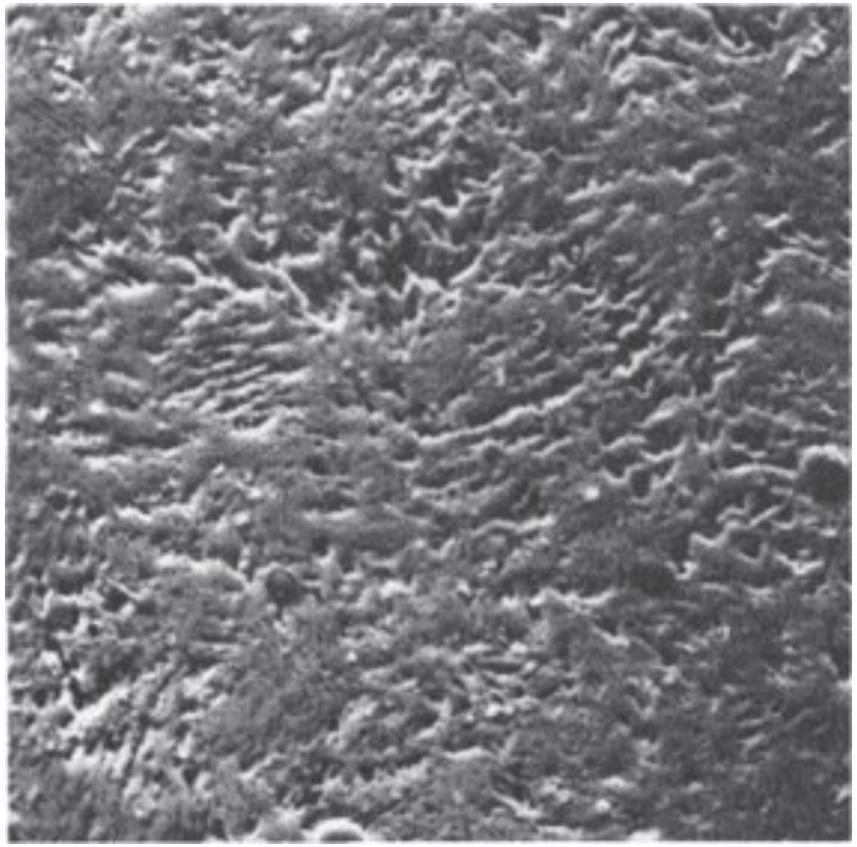
- The is believed to be due to the difference in swelling behavior between the hotter center of the fuel pin and the colder periphery
- The gamma phase dominates in the center of the fuel pin at high temperatures
- This phase has large gas bubbles, indicative of a higher plasticity of the fuel and, therefore, a lower capability to sustain shear stresses



gamma

U-Pu-Zr Swelling

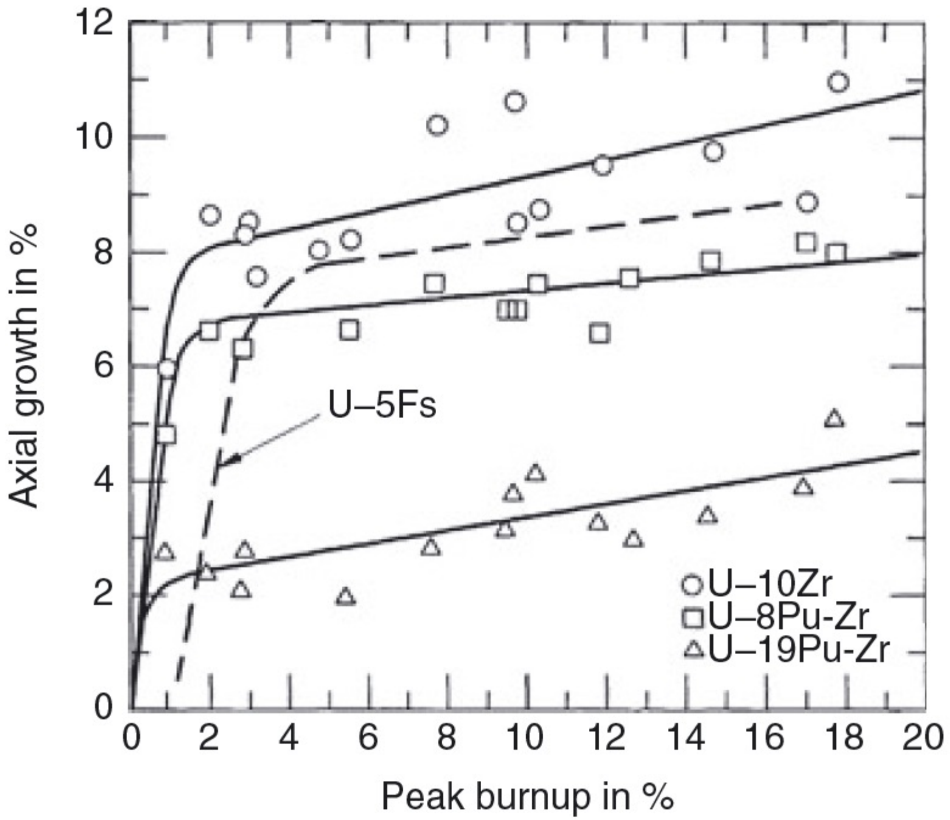
- The alpha phase dominates on the colder periphery of the fuel pin, and has smaller, textured porosity
- The alpha phase is known to have higher strength, and is assumed to be less plastic than gamma
- The fission gas pressure in the center would can potentially result in a near-biaxial loading of the peripheral shell, the radial stress component being twice the axial component
- A stress effect on swelling in the peripheral fuel zone would result in a larger radial strain, and hence anisotropic swelling



alpha

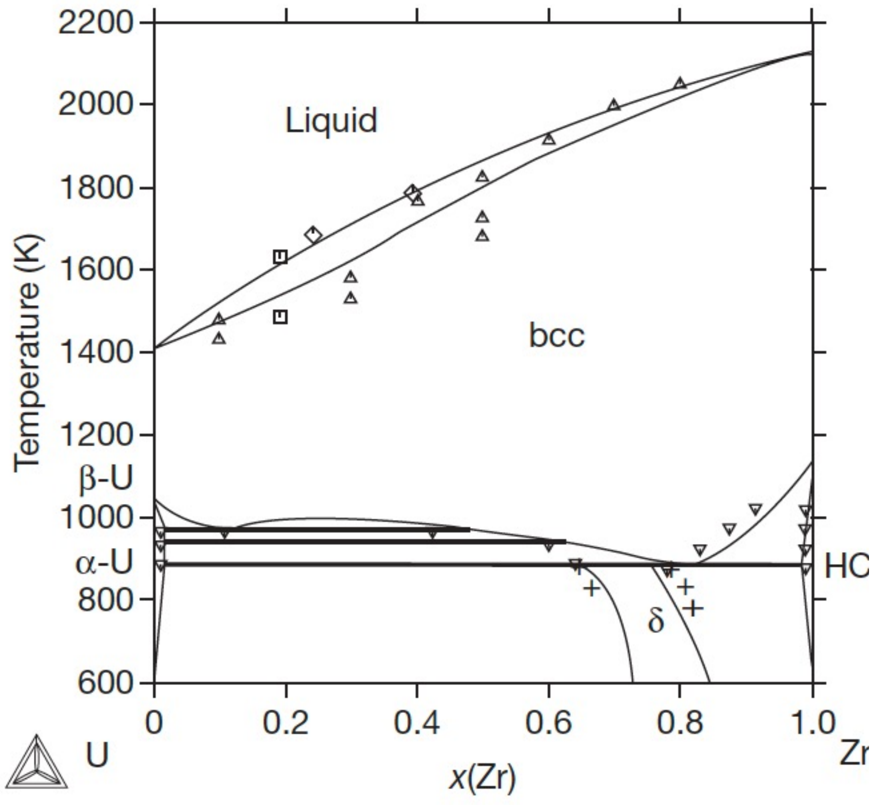
U-Pu-Zr

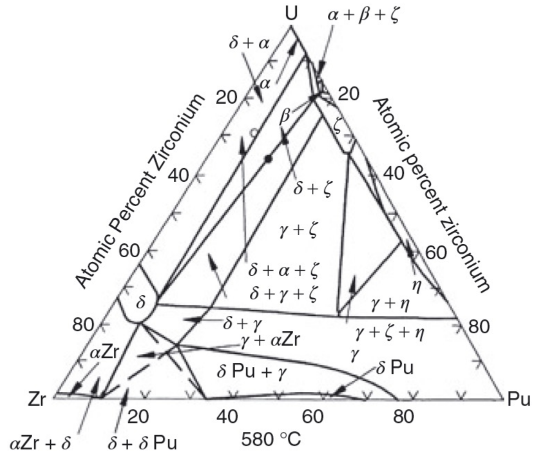
- An increase in the Pu content exacerbates the anisotropic swelling
- This is believed to be driven by the redistribution of Zr along the temperature gradient of the fuel

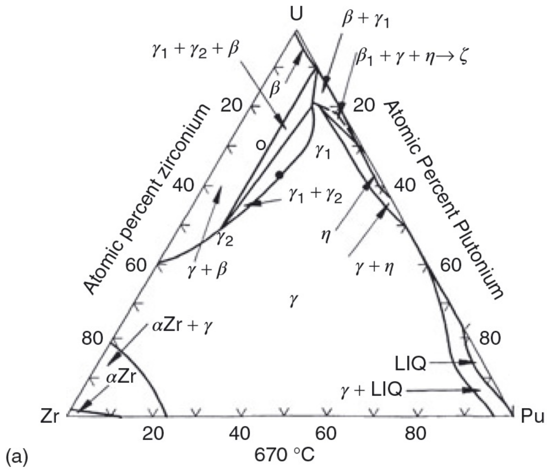


U-Pu-Zr

- Various phase boundaries exist radially across the fuel
- The solubility of Zr in these phases varies with temperature; thus, a driving force for diffusion can be created both by the gradient in chemical potential, and by the temperature gradient

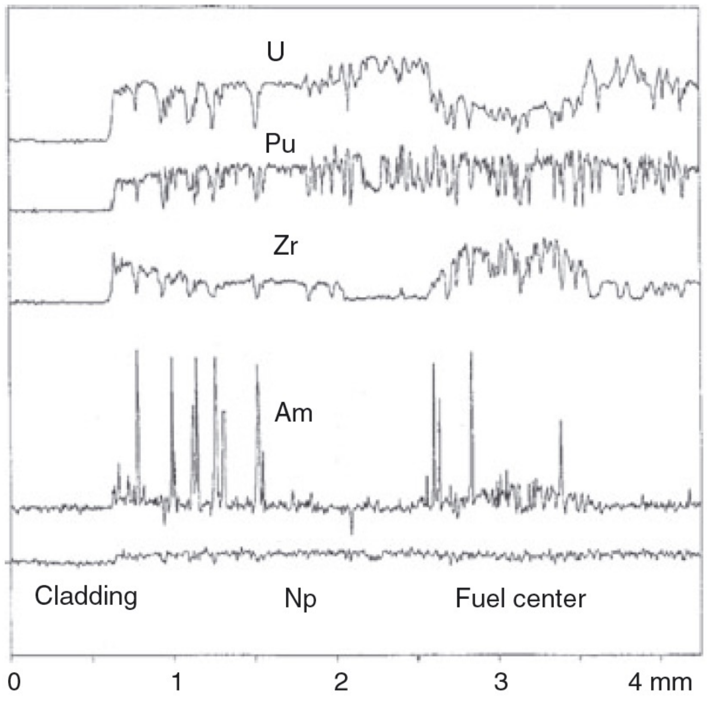






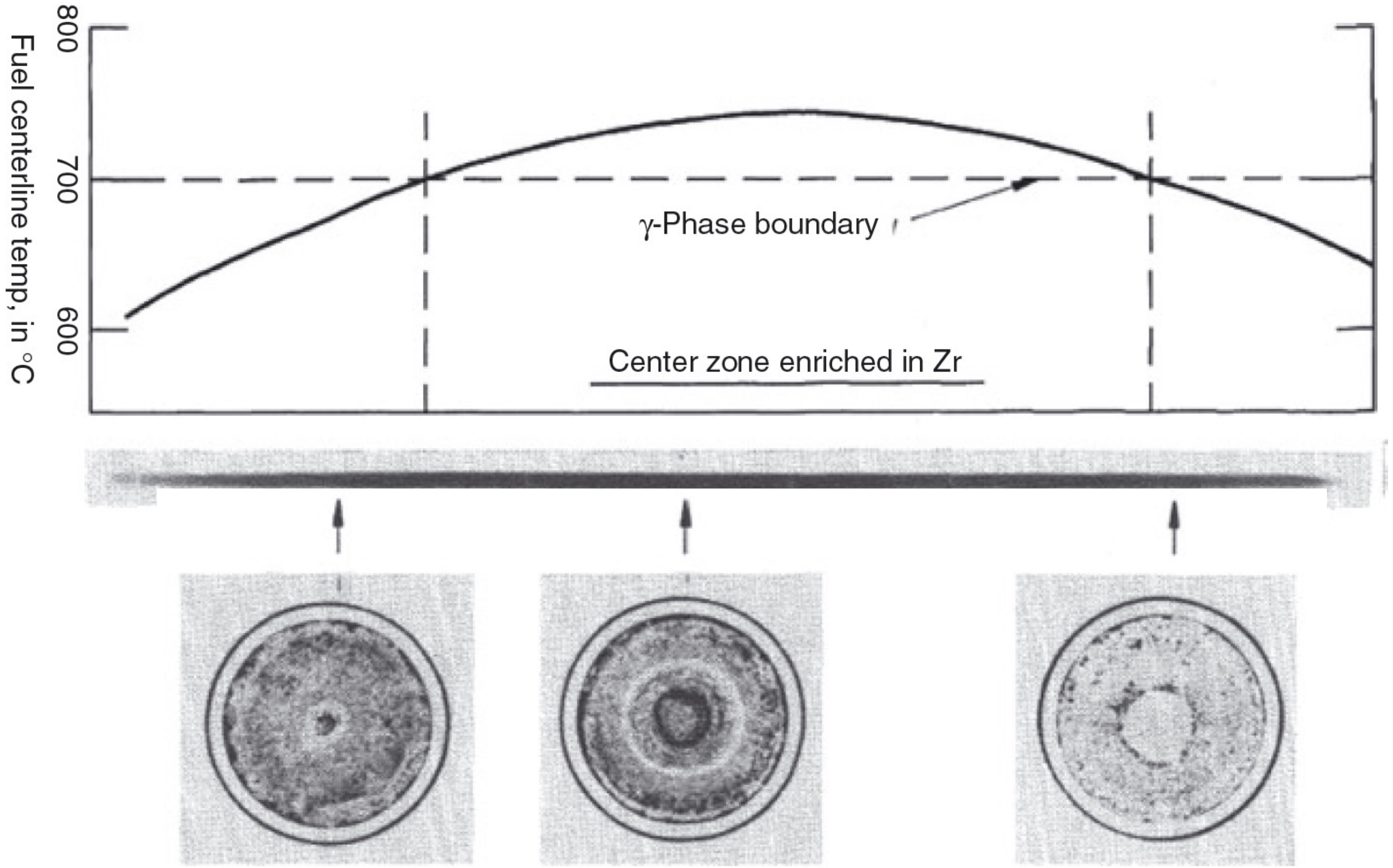
Redistribution

- There can exist three compositional regions: 1) enriched Zr, 2) depleted Zr, 3) as-fabricated Zr
- Zirconium diffuses up the temperature gradient to the high temperature gamma phase, leaving the intermediate temperature beta U phase
- Uranium compensates by diffusing to the intermediate region
- At lower T, Zr is enriched in the periphery in the delta phase



Redistribution

- The location of the radial zones essentially follows isotherms in the fuel which are determined by the various phase boundaries of the alloy
- The extent of redistribution, and the radial location of the individual compositional regions, varies axially with temperature
- Both chemical potential and Soret effect diffusion contribute



Soret Diffusion

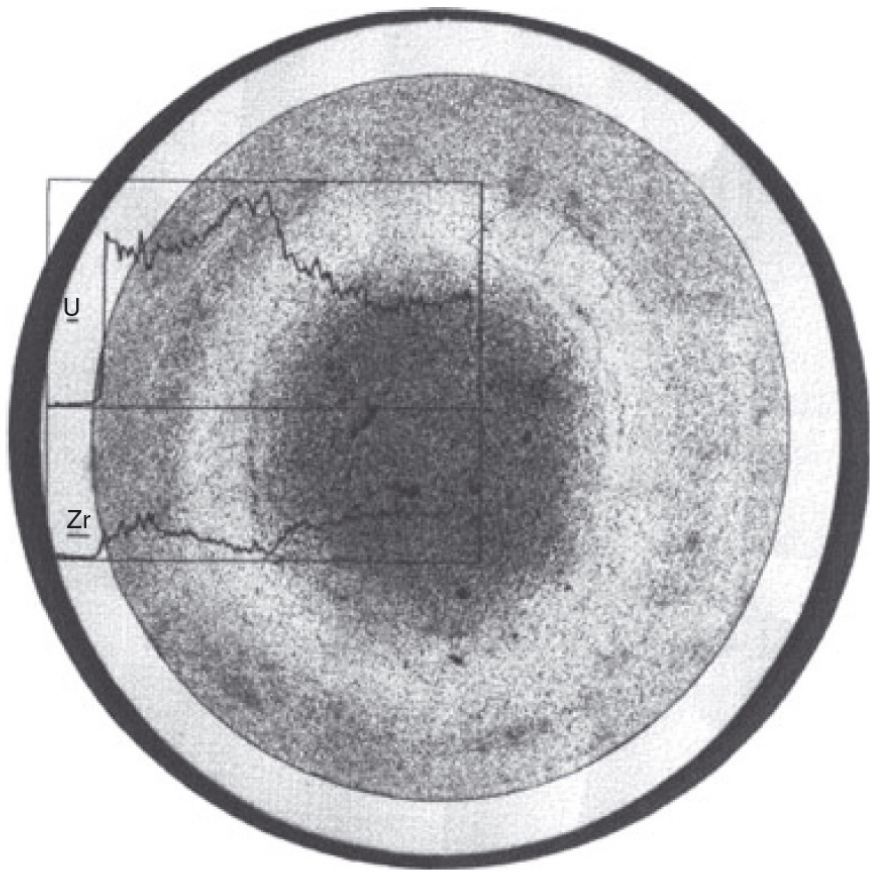
- Soret effect is a phenomenon observed in mixtures of mobile particles where the different particle types exhibit different responses to the force of a temperature gradient
- The solute flux in a temperature gradient is characterized by the heat of transport, Q*, of the migrating solute according to Fick's law:

$$J_i = \frac{-D_i N_i}{RT}\left(\frac{RT\partial \ln N_i}{\partial x} + \frac{Q^*}{T}\frac{\mathrm{d}T}{\mathrm{d}x}\right)$$

- where Di and Ni are the diffusivity and concentration of the migrating solute
- The heat of transport can be either positive or negative, depending on the sign and magnitude of its components

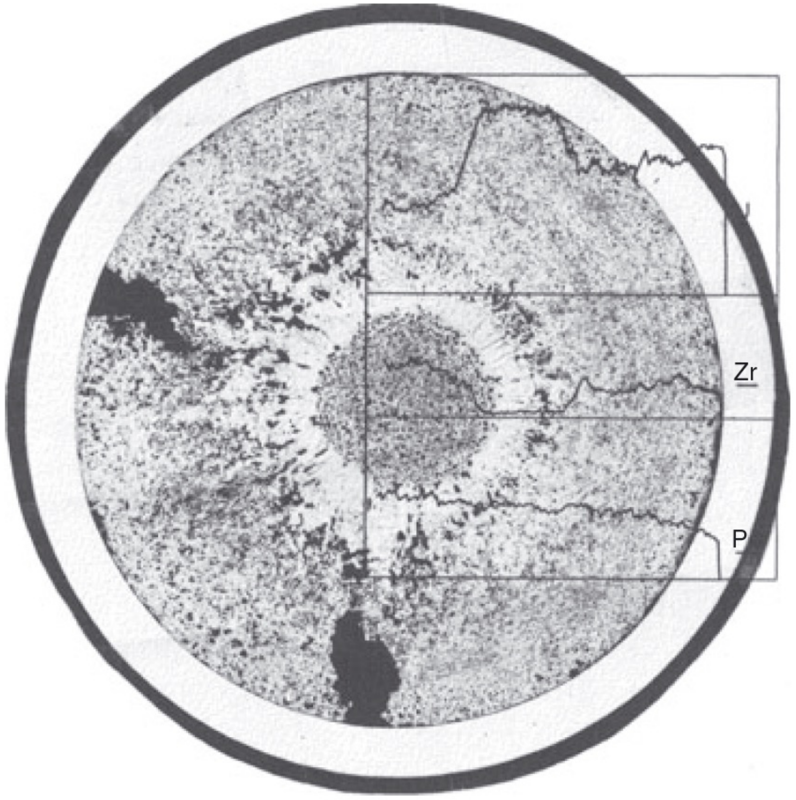
Soret Diffusion in UZr

- The value of Q* for pure U was measured to be +5 kcal/mol and to be −34 kcal/mol for Zr
- Thus, in their pure states, U moves down a temperature gradient while Zr moves up, but it is unknown if this behavior directly translated to U-Zr alloys
- For low Pu content alloys, the rate of redistribution is similar to UZr



Soret Diffusion in UZr

- For high Pu content alloys (>19%), redistribution is much more rapid
- At the right is a 3% burnup fuel slug at a similar power density to the U-Zr slug in the previous slide
- There is either a higher chemical driving force, or more rapid Zr diffusion present in these alloys
- This more rapid redistribution can lead to greater anisotropic swelling, and thus more rapid radial swelling



Summary

- SFRs and Metallic Fuel
- Inherent safety associated with high thermal conductivity, large margin to boiling for Na, negative reactivity
- Alpha U undergoes anisotropic expansion and irradiation growth
- Alloy to stabilize gamma phase and improve properties
- Swelling is inevitable, can accommodate with smear density
- Include large plenum to accommodate fission gas release
- Radial redistribution of chemical species driven by thermodynamics and Soret diffusion