Nuclear Fuel Performance

NE-533

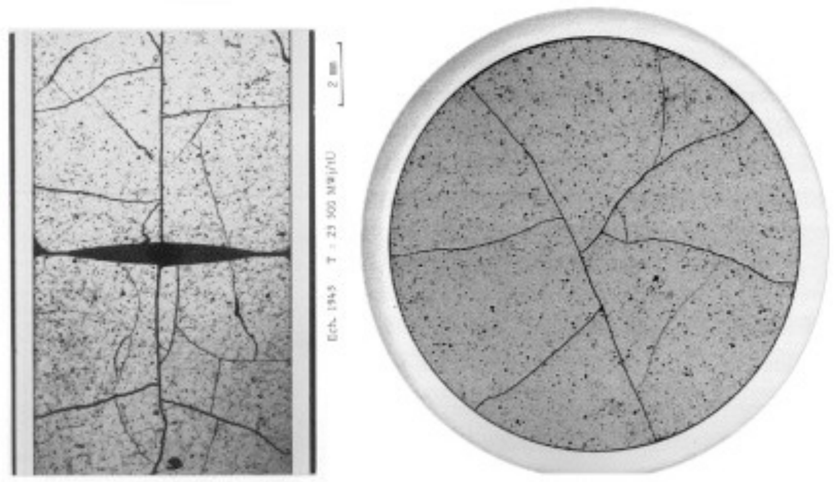
Spring 2023

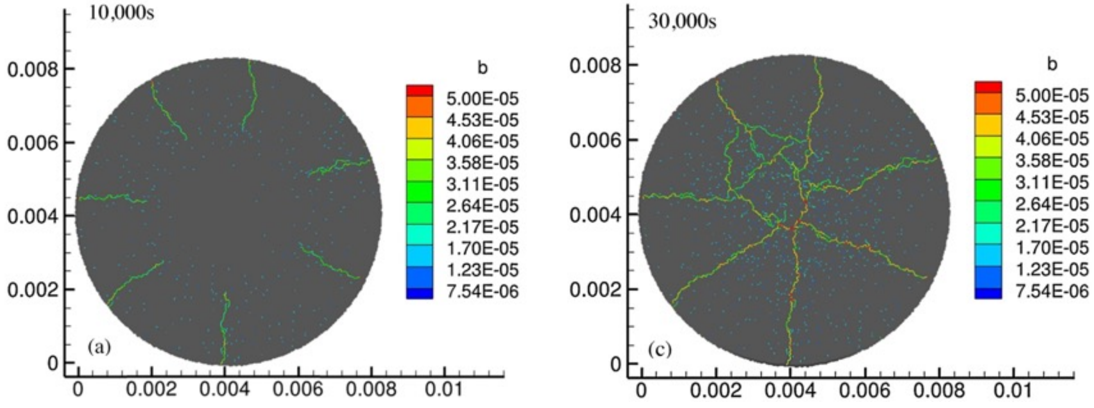
Last time

- Fission gas release models are used to understand fission gas experiments and to predict gas release for fuel performance codes
- Spherical grain models predict a fraction of gas release for post-irradiation annealing or for in-pile gas release
- Booth model takes Stage 1 into account; Forsberg-Massih model takes Stage 1 and 2 into account
- Fission gas diffusivity behavior changes with temperature and fission rate
- Many materials models for fuel are empirical and correlated to burnup
- Fuel pellets change shape due to
  - Thermal expansion (increase in volume); Densification (decrease in volume); Swelling (increase in volume); Creep (volume stays the same)

Fracture

- UO_2 pellets fracture during changes in temperature due to large thermal stresses
- Fracture results in:
  - Increased gap reduction
  - Reduced thermal conductivity
  - Increased avenues for fission gas release
- Fracture has been typically modeled in two ways:
  - Empirical relocation model that is a function of burnup
  - Semi-empirical smeared cracking model
- Modern methods provide means of modeling discrete cracks

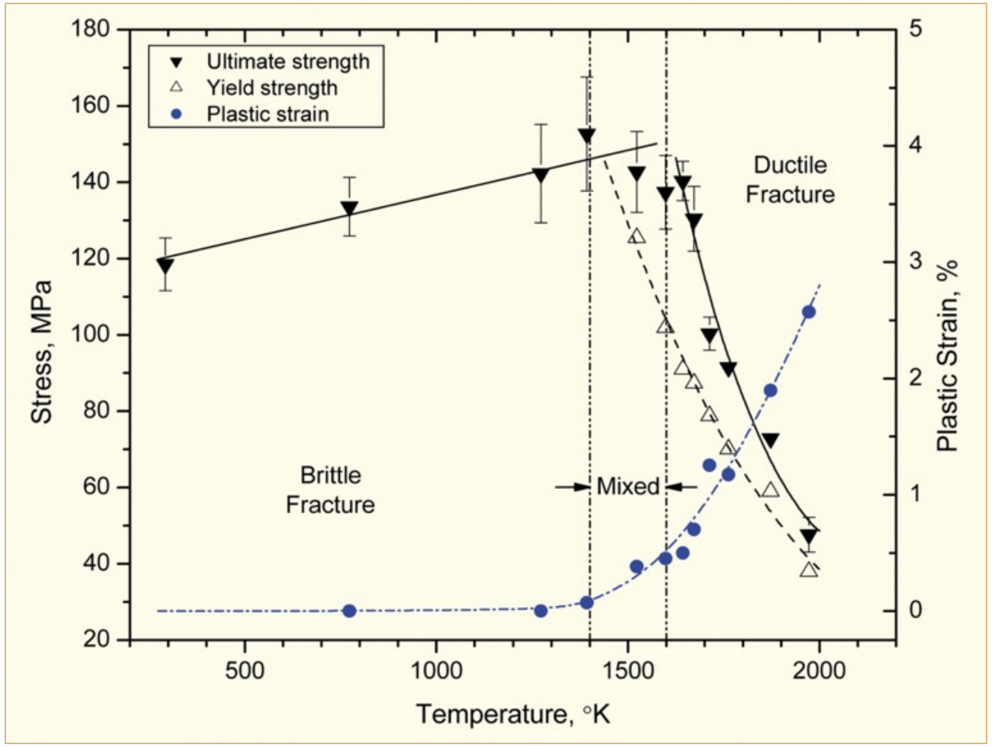




- Radial cracks partially penetrate the pellet during temperature increase
- Full cracking occurs when the temperature decreases

Fracture

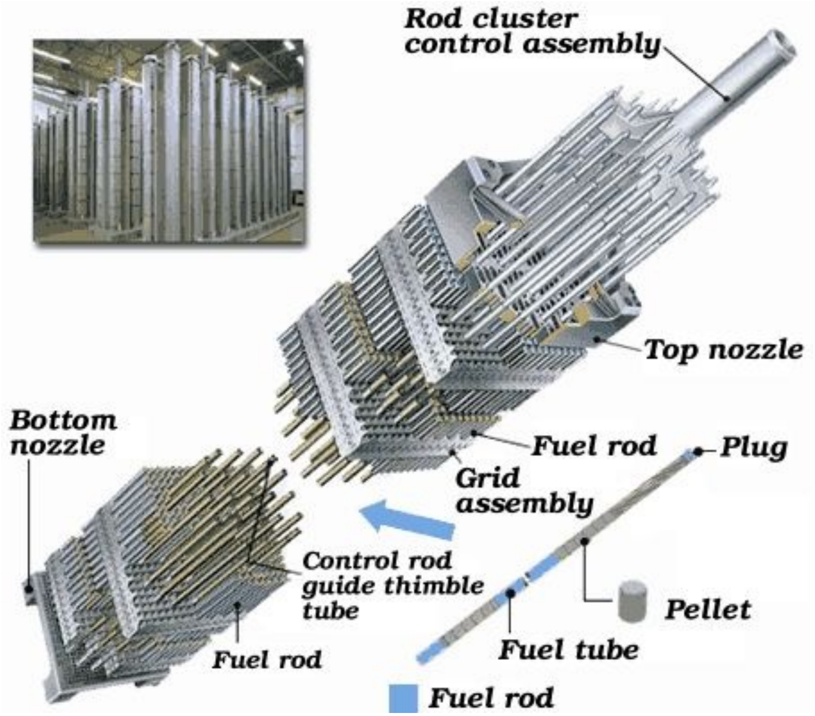
- The fracture behavior of the fuel is fairly complicated
- Fracture strength varies with grain size (G)
  - $\sigma_{frac} = G^{-m}\sigma_{frac,\, ref}$, m = 0.04 – 0.05 (vs. m ~ 0.5 for metal)
  - Increasing grain size from 10 μm to 100 μm reduces σ_{frac} by ~10%
- Ductility transition temperature is lower in-reactor than in thermal tests
- Fracture strength is ~10 × higher in compression than in tension
- Load-deformation behavior strongly affected by creep under in-reactor conditions

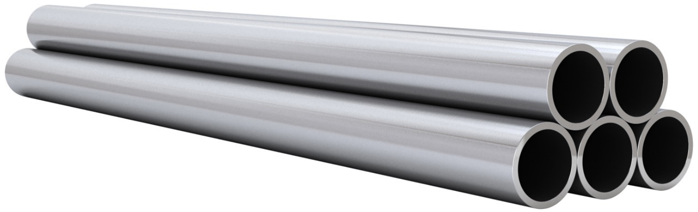


ZIRCONIUM CLADDING

Cladding

- The purpose of the cladding is to:
  - Hold the pellets together so that coolant can freely flow past
  - Transport heat from fuel to the coolant
  - Contain fission products
  - Contain fuel fragments
- We would also prefer if the cladding had little to no impact on the neutron transport in the reactor





Why Zirconium alloys?

Benefits

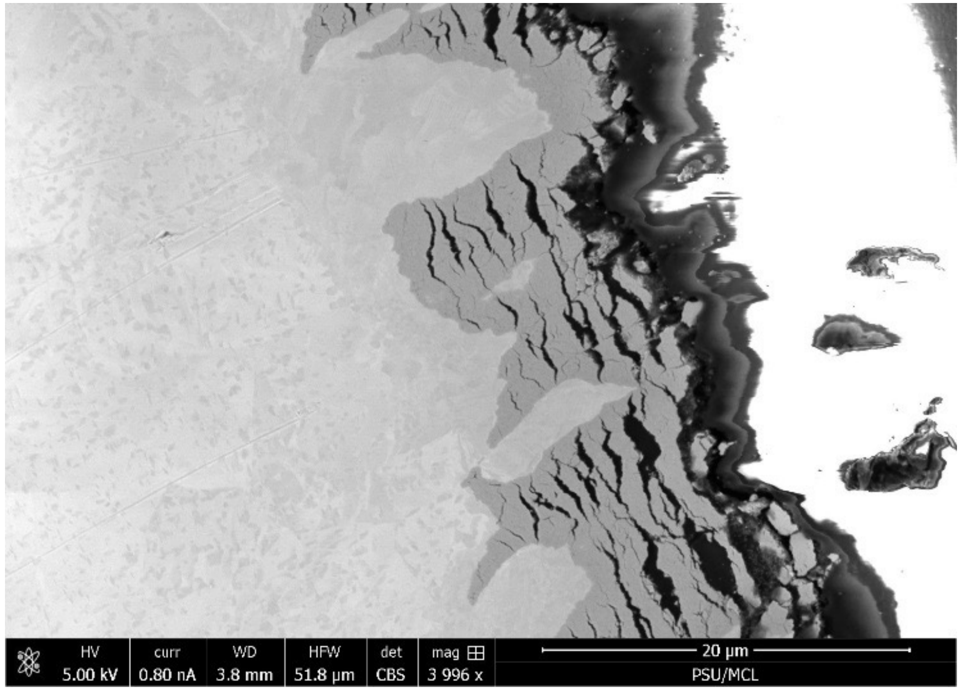
- Low neutron cross section
- Corrosion resistance in 300˚C water
- Resistance to void swelling
- Adequate mechanical properties
- Good thermal conductivity
- Affordable cost
- Available in large quantities

Problems?

- Corrosion under high temperature steam
- Hydride embrittlement
- Anisotropic characteristics lead to creep and growth

Zirconium

- Pure zirconium was not acceptable due to its oxidation behavior
- Oxygen with water reacted to form an oxide layer
- The oxide layer is brittle and flakes off, allowing more oxidation to occur
- Zr alloys were developed to reduce corrosion resistance



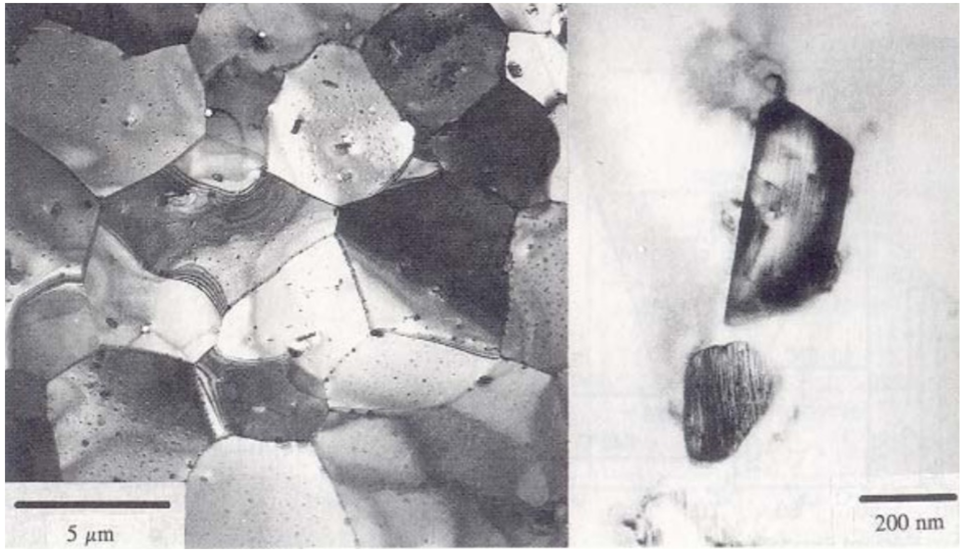
Commercial Zr Alloys in PWRs

Alloy	Sn %	Nb %	Fe %	Cr %	Ni %	O %
PWRs (structural components and fuel rods)						
Zircaloy-4 (SRA)	1.2-1.7	-	0.18-0.24	0.07-0.13	-	0.1-0.14
ZIRLO (SRA)	1	1	0.1	-	-	0.12
Optimized ZIRLO (pRXA)	0.7	1	0.1	-	-	0.12
M5 (RXA)	-	0.8-1.2	0.015-0.06	-	-	0.09-0.12
HPA-45 (SRA/RXA)	0.6	-	Fe+V	-	-	0.12
NDA (SRA)	1	0.1	0.3	0.2		0.12
MDA (SRA)	0.8	0.5	0.2	0.1		0.12

PRXA Partial Recrystallization Anneal
RXA Recrystallization Anneal
SRA Stress-Relief Anneal

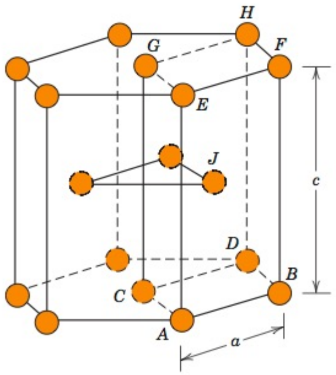
Alloying Elements

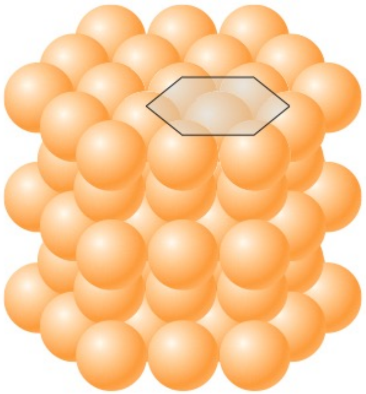
- The alloying elements impact the microstructure of the material
- The intermetallic precipitates significantly improve the oxidation behavior
- In Zircaloy 2, the precipitates are
  - $Zr(Cr, Fe)_2$
  - $Zr_2(Ni, Fe)$
- In Zircaloy 4, the precipitates are
  - $Zr(Cr, Fe)_2$
- Phosphides (Zr_3P) and silicides (Zr_3Si) are also occasionally found
- The precipitate distribution, size, morphology, and composition all impact the properties of the material

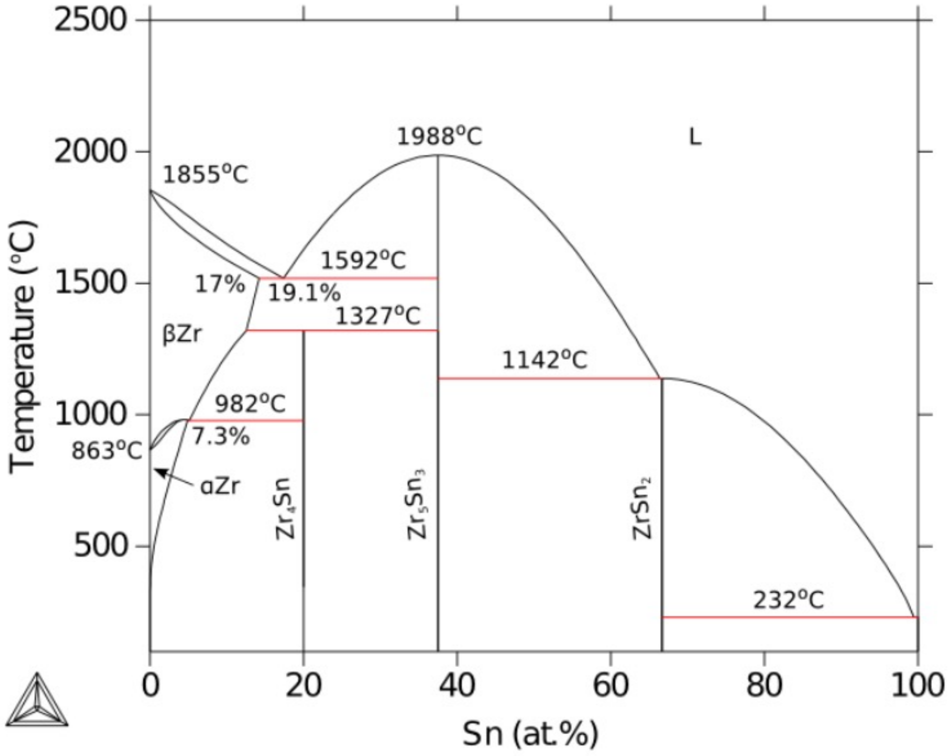


Zirconium Phases

- The α-Zr phase has a hexagonal-close-packed (HCP) structure
  - At temperatures below about 863ºC
  - Has the most desirable properties
- The β-Zr phase has a body-centered cubic (BCC) structure
  - We try to avoid this phase







Zr Tube Fabrication

- The cladding tubes are fabricated using various processes that SEVERELY deform the material
- The severe plastic deformation causes significant dislocation hardening, that can make the tube material too brittle
- Plastic deformation along slip planes results in reorientation of the grains
- The properties of the material are controlled by heat treatment
- Raising the temperature, but below 863˚C, anneals the sample to reduce cold work (stress-relieved)-SRA
- Raising the temperature above 863˚C changes to the β phase. They then quench the sample to create a random texture in the α phase-RXA or pRXA

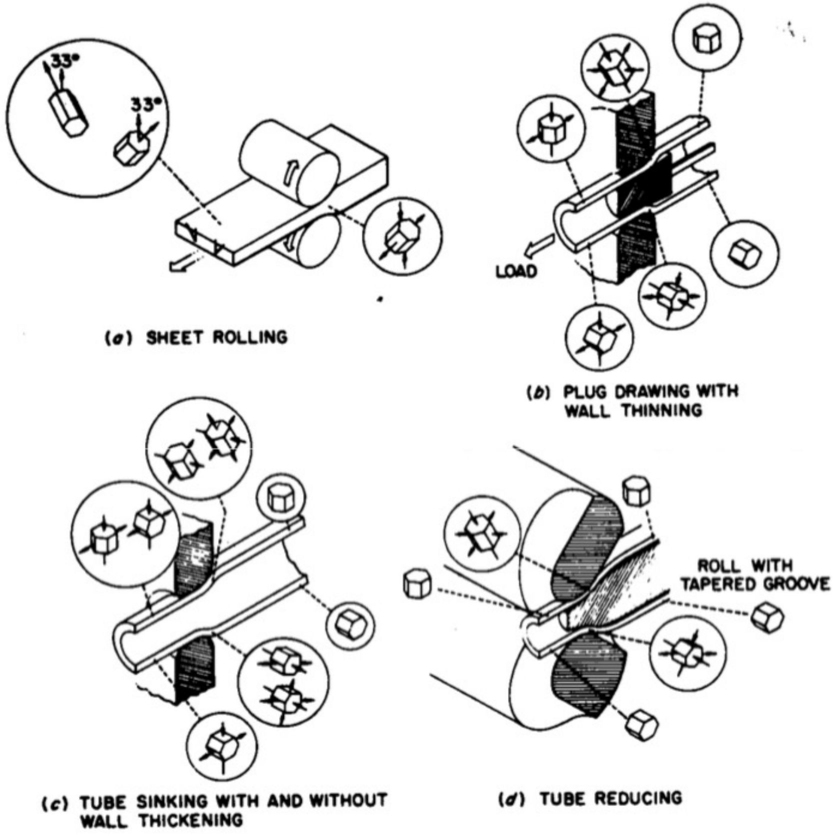


FIG. 48. Strain states for various types of fabrication of Zircaloy-2 [109].

Zr alloy microstructures

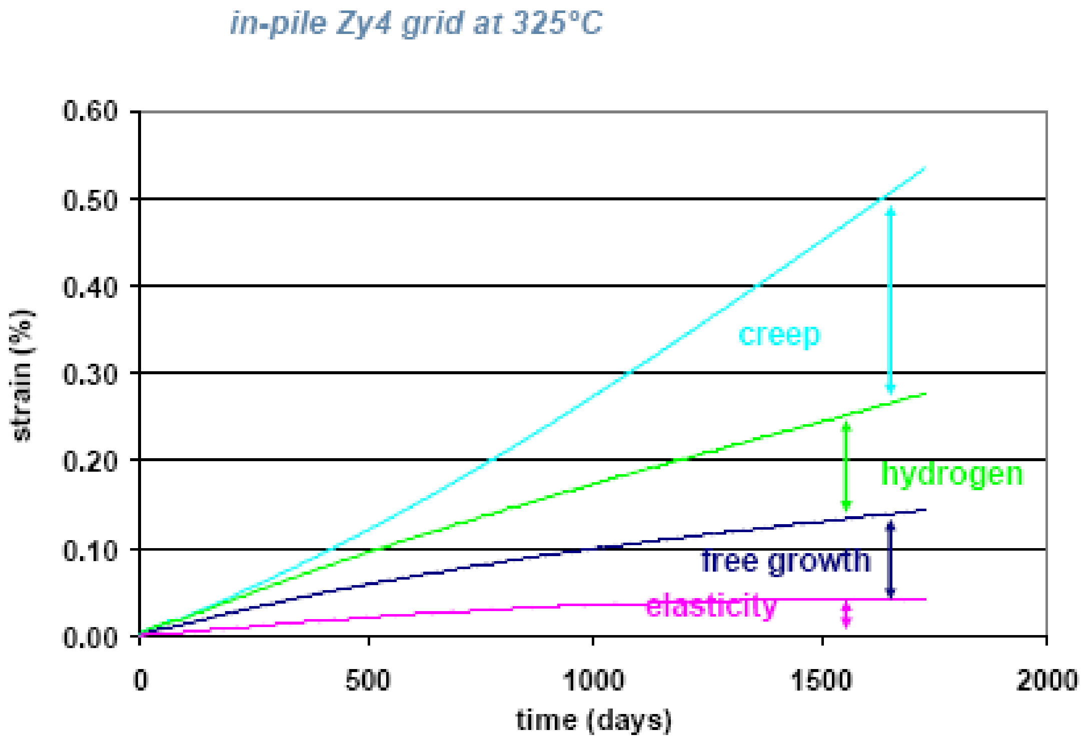
Fully recrystallized after quench

Stress-relieved microstructure

Zirconium Creep and Growth

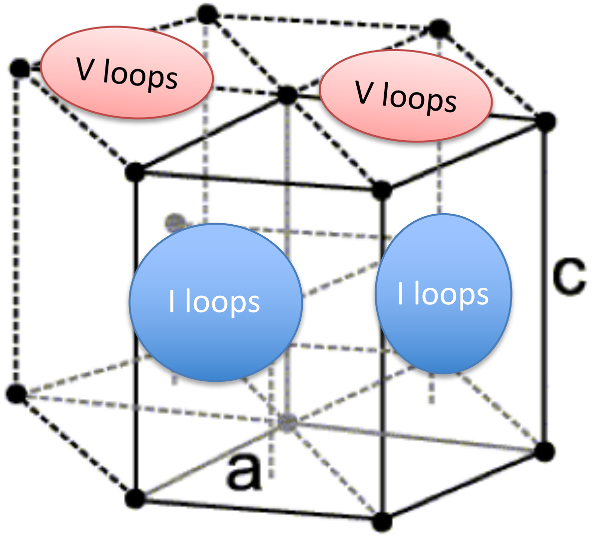
- The cladding undergoes a very large amount of irradiation during its life, the main effects are:
  - Interstitial loops form on prism planes
  - Later in the reactor exposure, vacancy loops form on the basal plane
- The overall number of displacements to the cladding can be calculated with the NRT model to be ~ 8 x 10^{-8} dpa/s, which, over 3 years exposure gives a total of ~8 dpa (every atom in the solid is displaced on the average eight times)

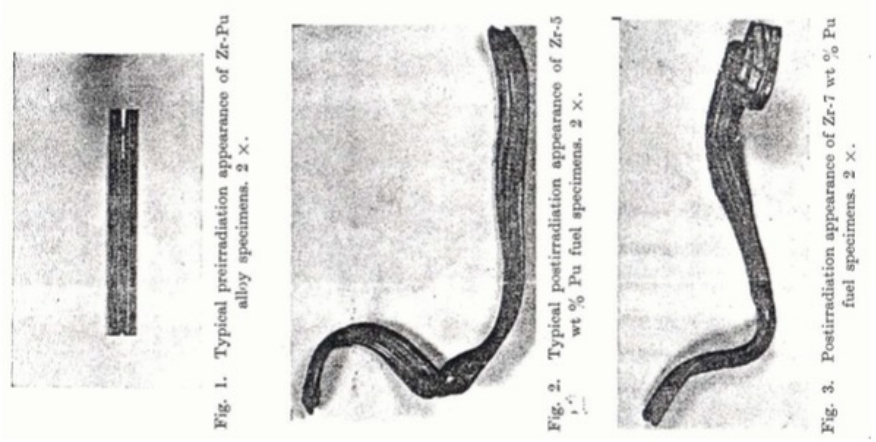
$$\nu(T) = \frac{\kappa E_D}{2E_d} = \frac{\kappa(T-\eta)}{2E_d},$$



Irradiation Growth

- Irradiation growth results from material anisotropy
- There must be anisotropy in the defect behavior within the unit cell
- There also must be a texture formed in the grain orientations
- Interstitial loops form on pyramidal planes, causing shrinkage along the center axis
- Later, vacancy loops form on basal planes, making it even worse
- The result is a change in shape, shrinking in the 0001 direction and expanding in the pyramidal direction





Irradiation Growth

- Three stages of irradiation growth
  - Initial rapid growth to small strains
  - Slow growth, gets skipped in cold worked material
  - Accelerated or breakaway growth, caused by formation of vacancy loops on the basal plane
- Irradiation growth is affected by:
  - Fluence, Cold work, Texture, Irradiation temperature, Material chemistry, Hydrogen effects

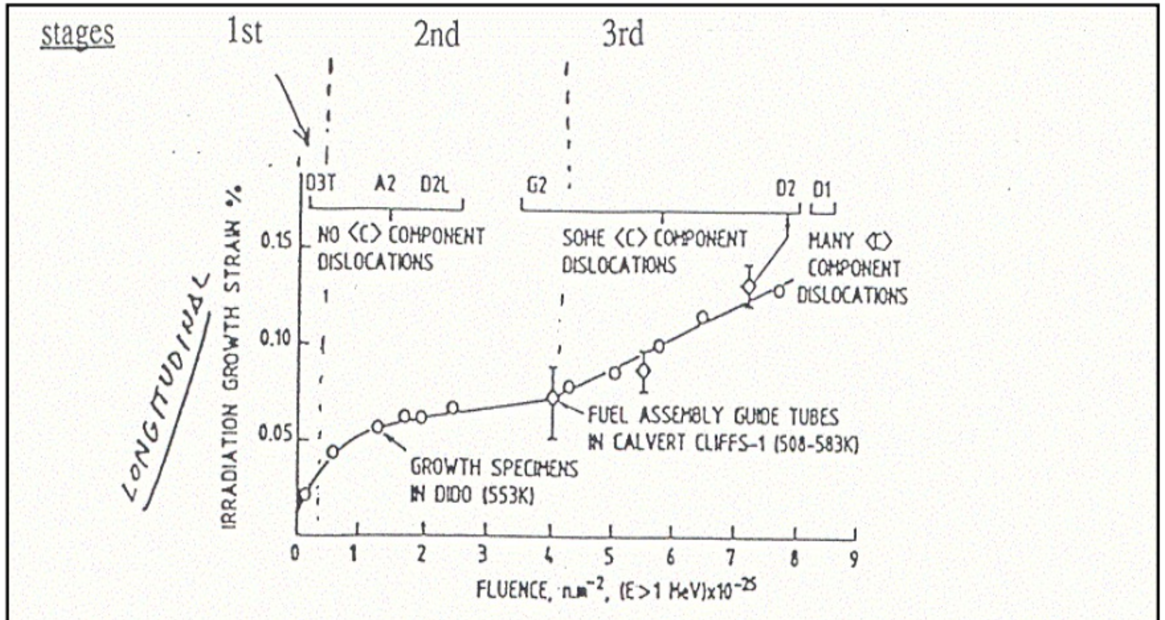
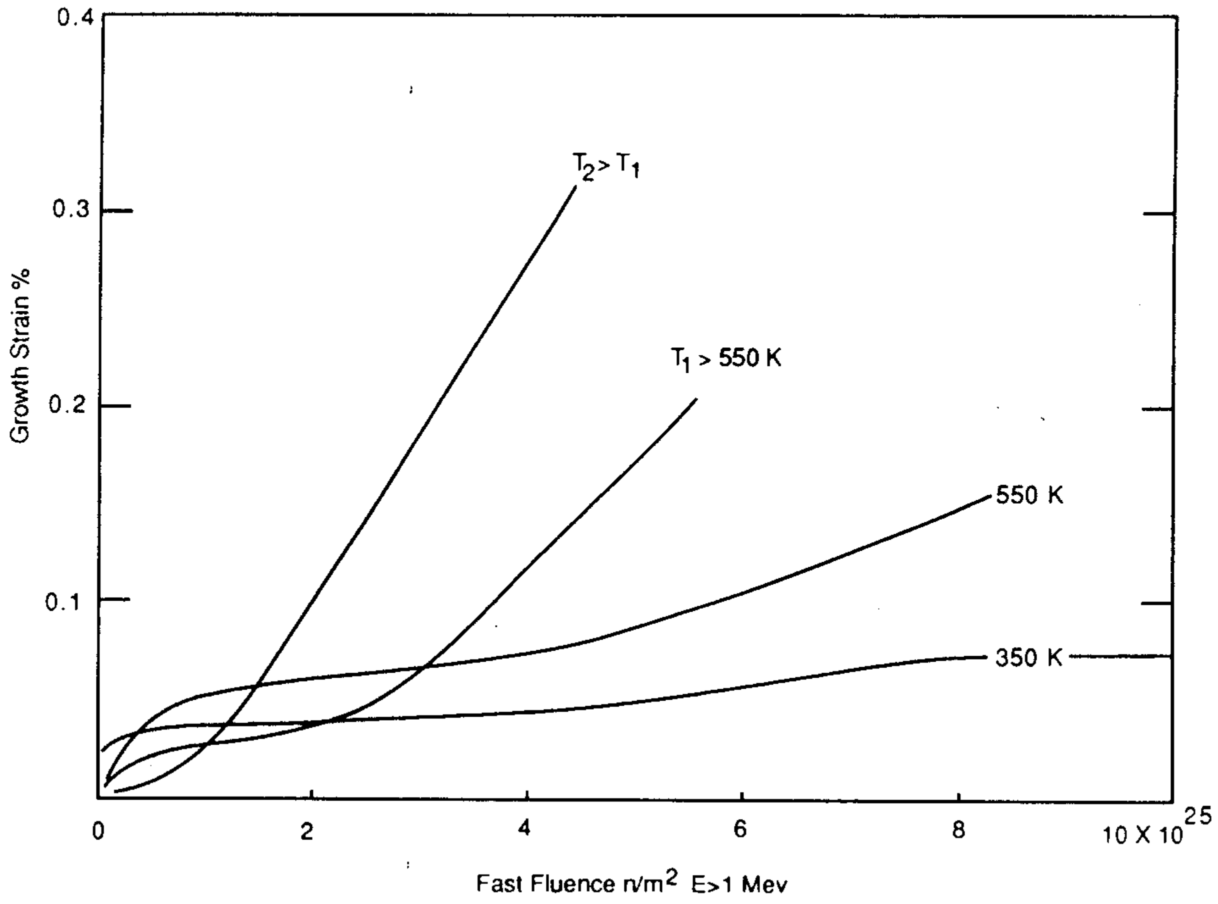


FIG. 11—*Irradiation growth in annealed Zircaloy at 550 to 580 K, showing accelerating growth at* 4 × 10^{25} *n/m*^{2}, E > *1 MeV* [61].

Irradiation Growth

- Higher temperature leads to earlier breakaway and higher growth
- Higher temperature adds mobility to defects, allowing them to diffuse faster, form clusters, and exacerbate the defect cluster induced anisotropic growth



Creep

- Empirical models have been developed for thermal and irradiation creep of Zircaloy
- Both based on the Von Mises stress

$$\sigma_m = \sqrt{\frac{1}{2}\left((\sigma_{11}-\sigma_{22})^2 + (\sigma_{22}-\sigma_{33})^2 + (\sigma_{22}-\sigma_{11})^2 + 6\left(\sigma_{12}^2+\sigma_{23}^2+\sigma_{31}^2\right)\right)}$$

- Thermal Creep $\dot{\epsilon}_{ss} = A_0\left(\frac{\sigma_m}{G}\right)^n e^{\left(\frac{-Q}{RT}\right)}$
  - With A_0 = 3.14 × 10^{24} (1/s); shear modulus G = 4.2519 × 10^{10} − 2.2185 × $10^{7}T$ Pa; n = 5; Q = 2.7 × 10^5 J/mol
- Irradiation Creep $\dot{\epsilon}_{ir} = C_0\Phi^{C_1}{\sigma_m}^{C_2}$
  - φ is the fast neutron flux n/(cm^2 s) = 3E11xLHR n/(cm^2-s)
  - Note that SRA stands for stress relief annealed
  - RXA for recrystallization annealed
  - PRXA stands for partially recrystallization annealed

Clad Type	C_0	C_1	C_2
SRA	3.557×10^{-24}	0.85	1.0
RXA	1.654×10^{-24}	0.85	1.0
PRXA	2.714×10^{-24}	0.85	1.0
ZIRLO	2.846×10^{-24}	0.85	1.0

Creep Example

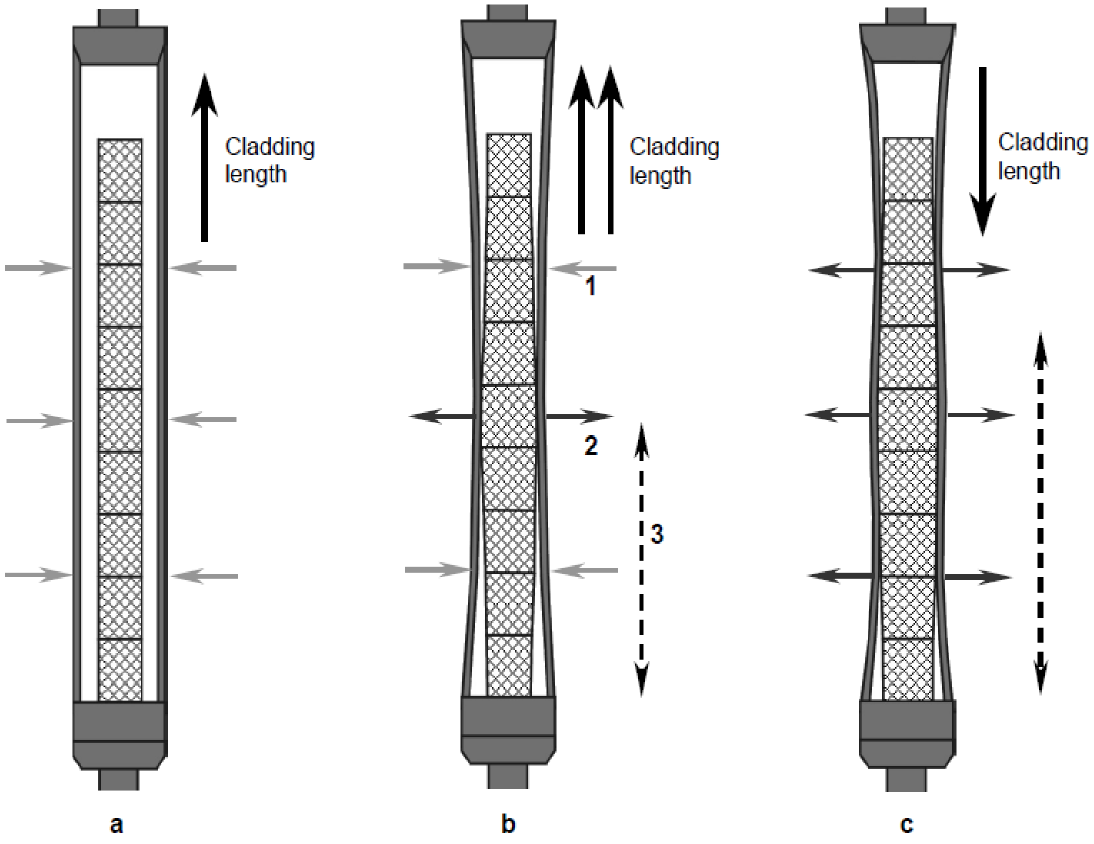
- Consider an SRA cladding tube at T = 600 K and LHR = 250 W/cm, with a stress σ_m = 200 MPa. What is the total creep strain after three years?
- First, we will calculate the thermal creep
  - $A_0 = 3.14 \times 10^{24}$ (1/s)
  - G = $4.2519 \times 10^{10} - 2.2185 \times 10^{7} T$ Pa = 4.2519e10 − 2.2185e7*600 = 2.92e10 Pa
  - Q = 2.7×10^5 J/mol, n = 5, R = 8.3144598 J/(K mol)
  - 3.14e24*(200/2.92e4)^5*exp(-2.7e5/(8.3144598*600)) = 1.48e-10 1/s
- Now we will calculate the irradiation creep
  - C_0 = 3.557e-24, C_1 = 0.85, C_2 = 1.0
  - $\Phi \approx$ 3e11 LHR = 3e11*250 = 7.5e13 n/(cm^2 s)
  - 3.557e-24*(7.5e13)^0.85*200^1 = 4.43e-10 1/s
- The total creep strain rate is 1.48e-10 + 4.43e-10 = 5.91e-10 1/s
- The total creep strain after three years is (assuming constant conditions) 5.91e-10*(3600*24*365*3) = 0.056 = 5.6% strain

$$\dot{\epsilon}_{ss} = A_0 \left(\frac{\sigma_m}{G}\right)^n e^{\left(\frac{-Q}{RT}\right)}$$

$$\dot{\epsilon}_{ir} = C_0 \Phi^{C_1} {\sigma_m}^{C_2}$$

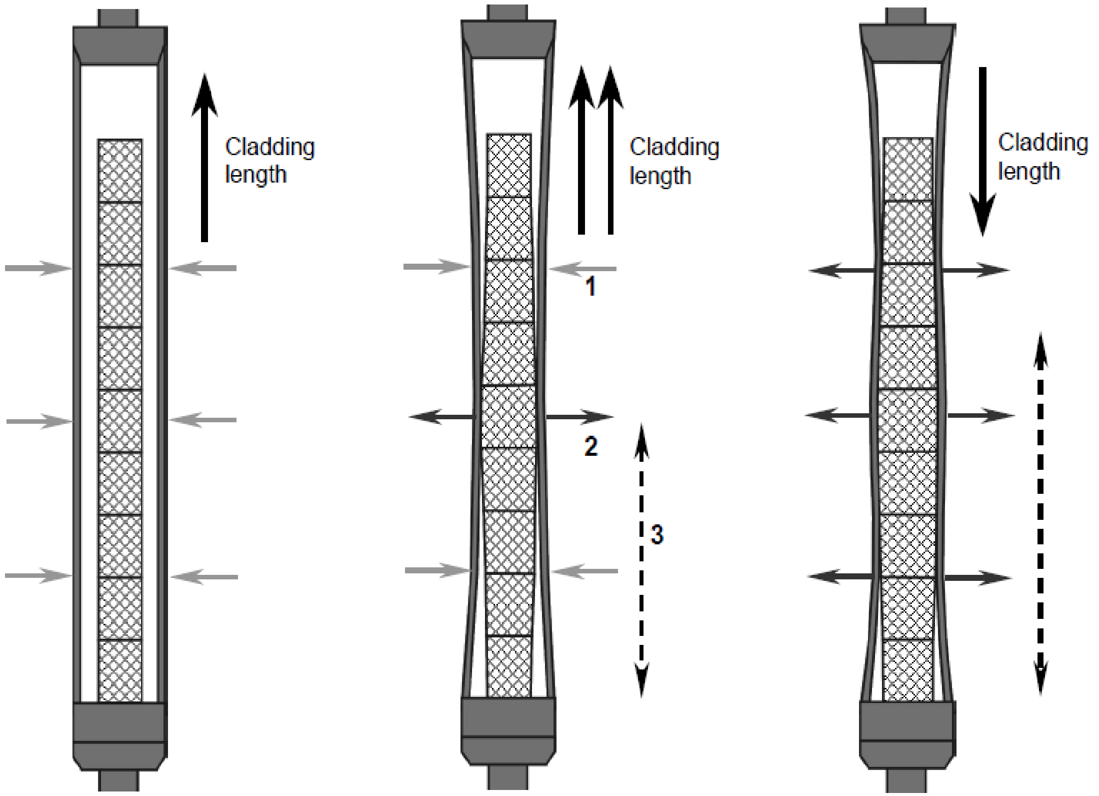
Creep

- Creep impacts fuel performance by shrinking the gap and then conforming to the pellets
- a) before “fuel-cladding” interaction: The stress due to coolant pressure exceeds the internal stress from the gap; The diameter decreases due to thermal creep and irradiation creep, where the thermal creep decreases rapidly as irradiation damage builds up; The length increases due to anisotropic creep and irradiation growth



Creep

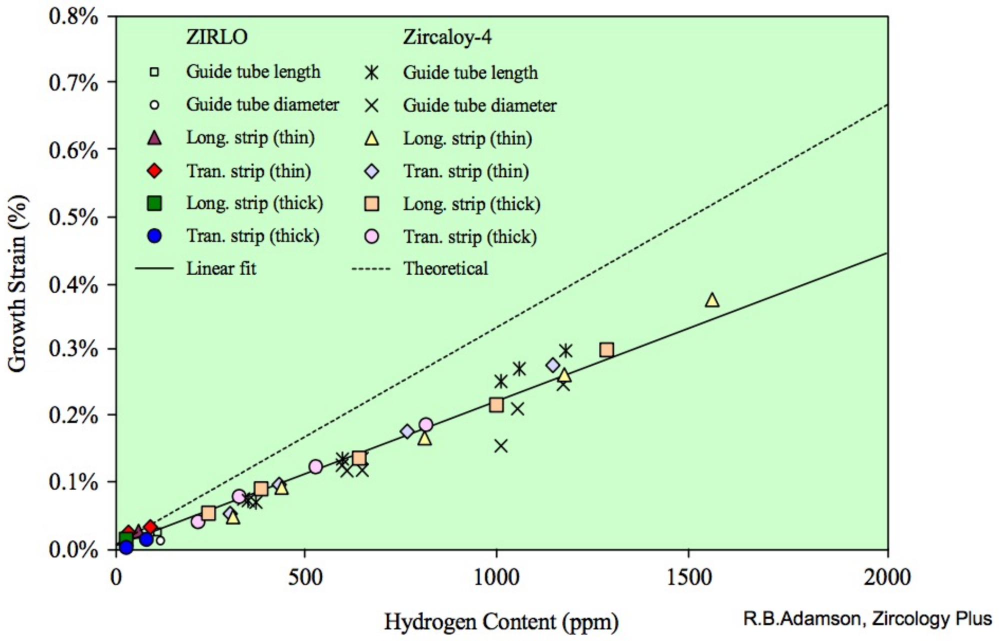
- b) start of "fuel-cladding" interaction: At the contact points the diameter increases, causing some contraction in rod length, and the expanding fuel imparts a local axial tensile stress on the rod, causing an increase in length; Irradiation growth causes an increase in rod length; The net change in length is the sum of the various inputs, but the net is very likely an increase in rod length
- c) "fuel-cladding interaction" over most of fuel column: The fuel pellets stress the cladding outward, increasing the diameter of the rod; Anisotropic creep decreases the rod length; Axial pellet-cladding stresses and irradiation growth increase the rod length; The net change in rod length could be positive or negative



a b c

Zirconium Hydrides

- Hydrides cause size change because the hydride lattice is larger than the zirconium lattice
- There are a number of different Zr-H phases and morphologies that can present
- 0.1% H can cause 0.2% growth



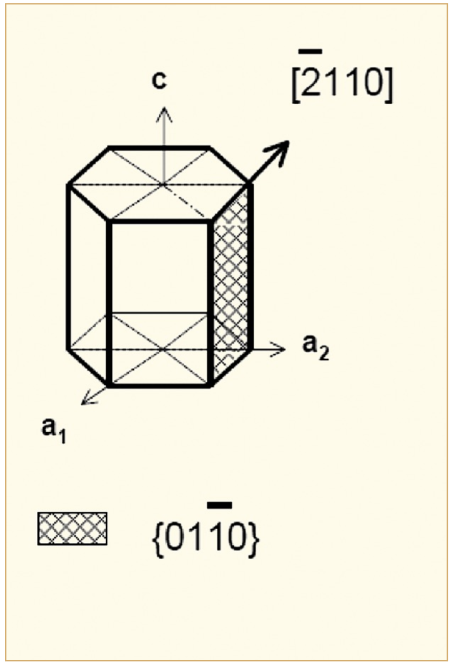
1000 ppm hydrogen can cause 0.2% dimension change

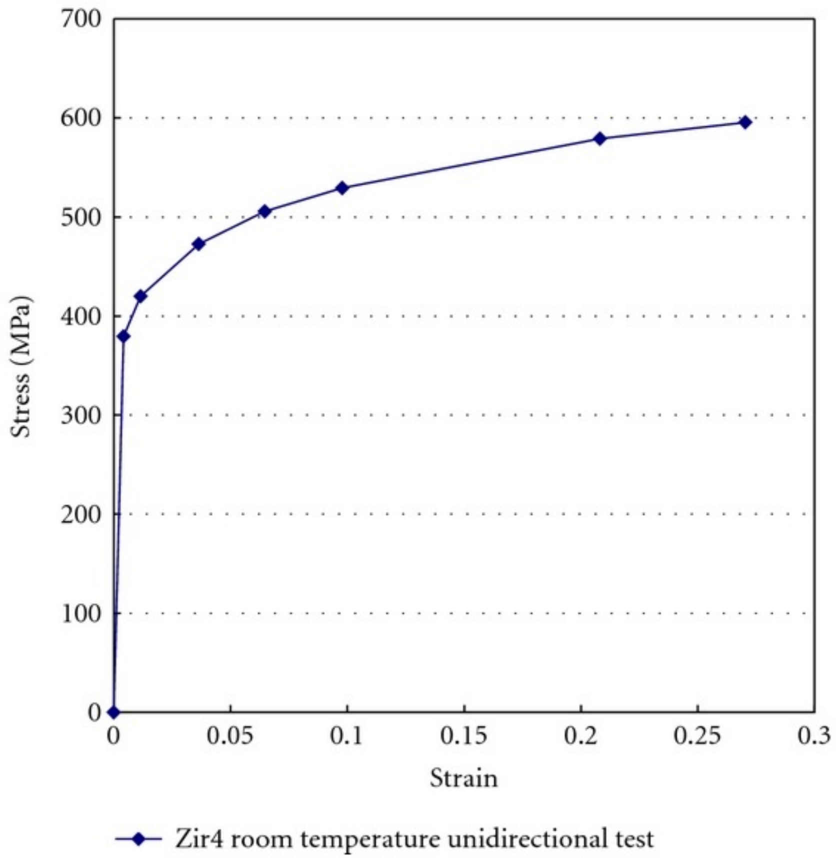
Summary of Creep and Irradiation Growth

- Growth and creep are the major mechanisms for dimensional instability in zirconium alloy cladding
- Growth results from the clustering of interstitials on prismatic planes, and eventually from the clustering of vacancies on basal planes such that the material shrinks in the axial direction
- Growth depends on the fluence, coldwork, texture, temperature, and composition

Mechanical Behavior of Zr Cladding

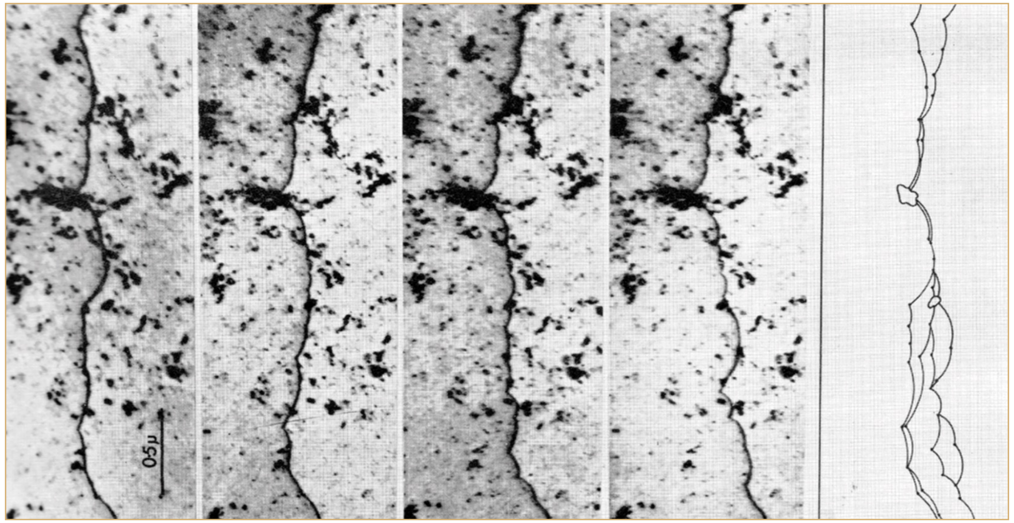
- Zirconium alloys plastically deform primarily due to dislocation motion on prismatic planes



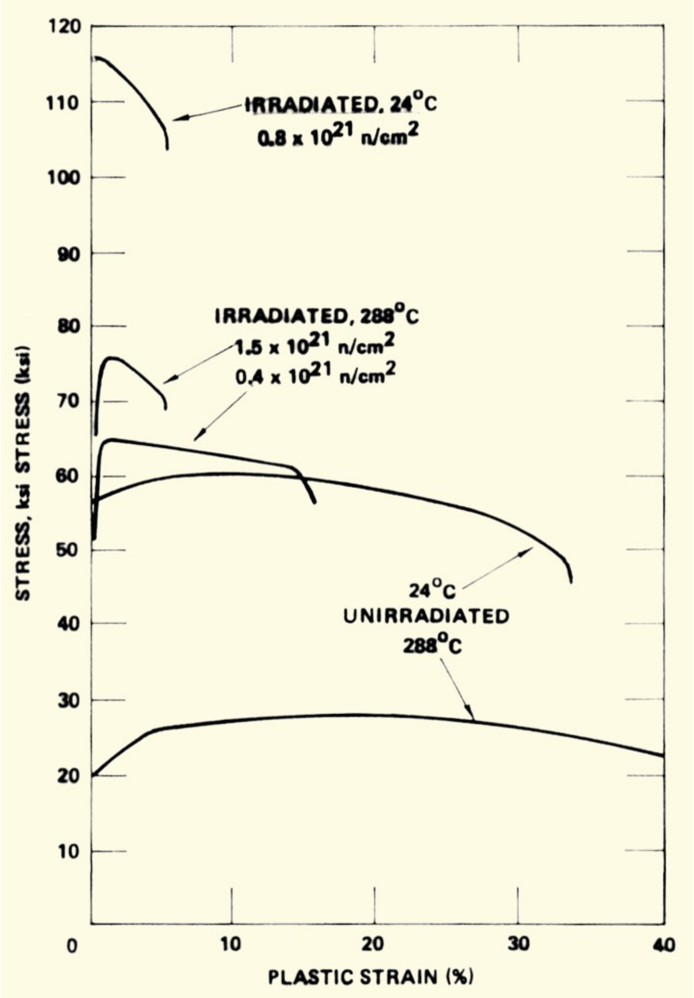


Irradiation Hardening

- Under irradiation, the interstitial loops that buildup on the prismatic planes cause hardening
- The UTS goes up with fluence, while the strain before fracture goes down

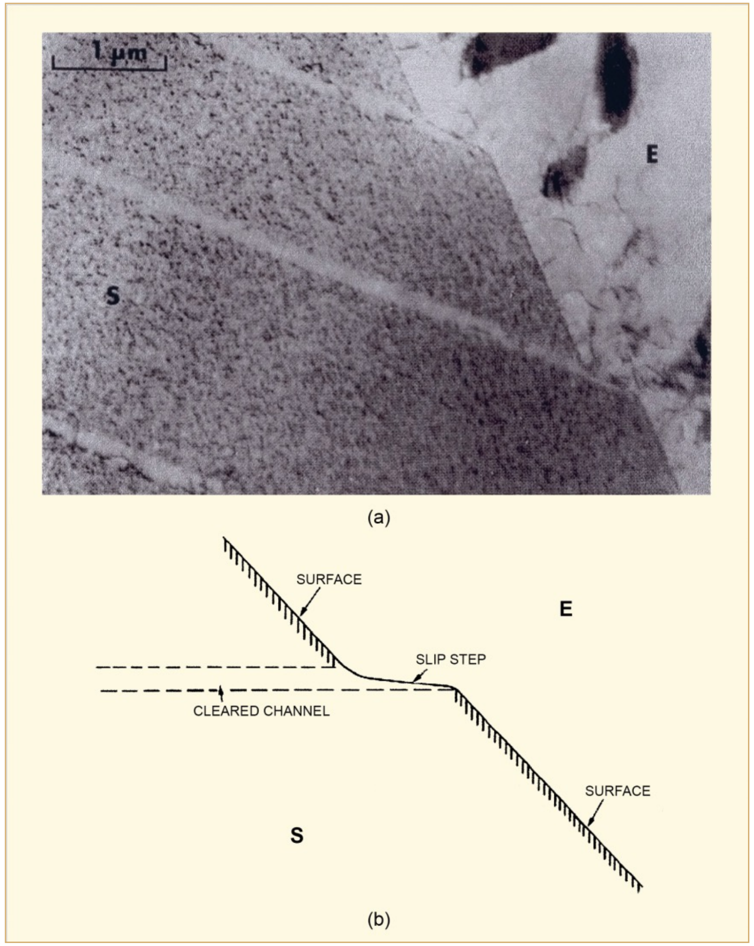


dislocation is moving →



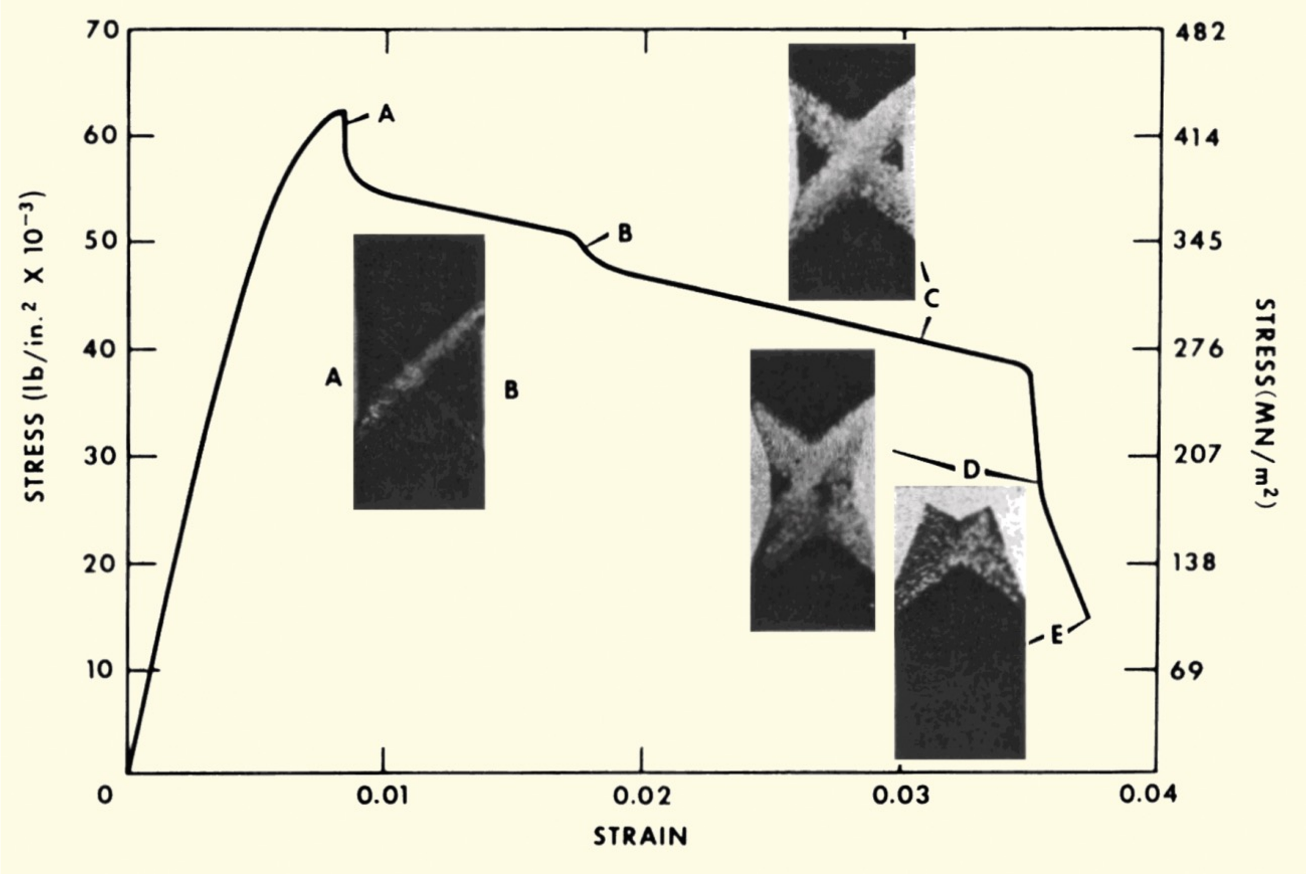
Dislocation Channels

- When stress is high enough, loops are "swept away", forming a clean channel
- Once a channel is cleaned out, dislocations continue to move along it
- This results in lots of deformation in that one area, creating slip steps
- In HCP zircaloy, channels form on basal planes or prism planes depending on
  - Load direction
  - Temperature
  - Oxygen content in the alloy
  - Fluence



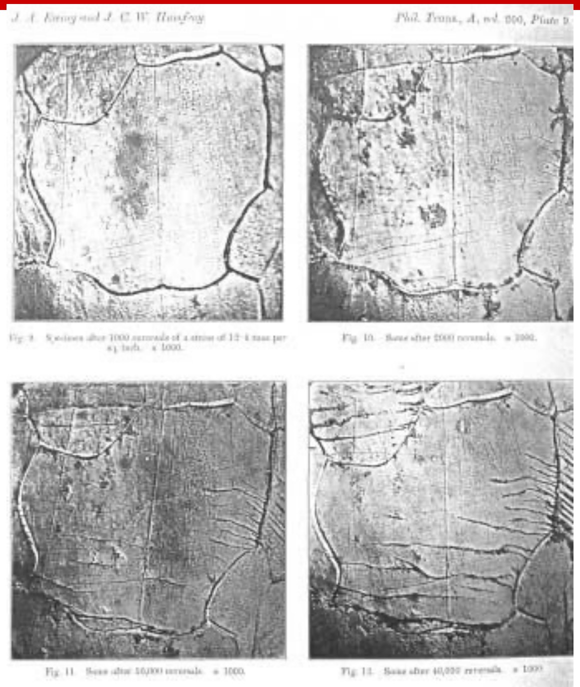
Dislocation Channels

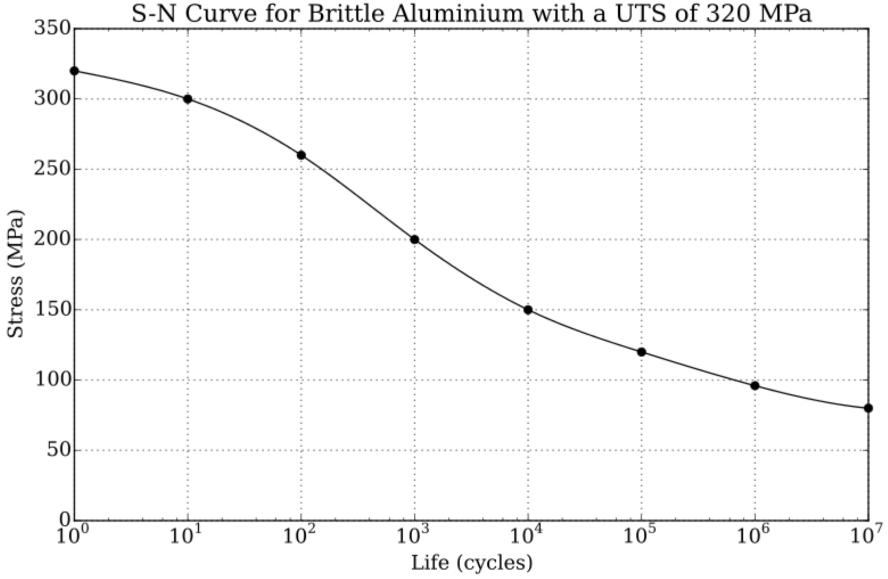
- Channel formation is an important behavior in irradiated zircaloy
- Dislocation channel deformation occurs at high load or high fluence



Fatigue

- Material fatigue is the weakening of a material caused by repeated applied loads
- Materials under cyclic loading can experience brittle like fracture at stresses significantly below their UTS
- It is caused by the slow propagation of microstructure damage
- The engineering tool for investigating fatigue is the S-N curve
- An S-N curve is a graph of the magnitude of the cyclic stress (S) against the logarithmic scale of cycles to failure (N).





Zr Fatigue

- Zircaloy experiences fatigue like other metals
- Zircaloy has a fatigue limit at about 10^5 cycles with 180 Mpa (The UTS is >500 Mpa)
- Irradiation slightly lowers the fatigue life in the low cycle range, less than about 10^4 cycles
- Irradiation has no effect on the fatigue life on the high cycle range, greater than about 10^5 cycles
- Design must demonstrate a factor of safety of 2 on stress and 20 on cycles