

# Nuclear Fuel Performance

NE-533  
Spring 2022

## Last time

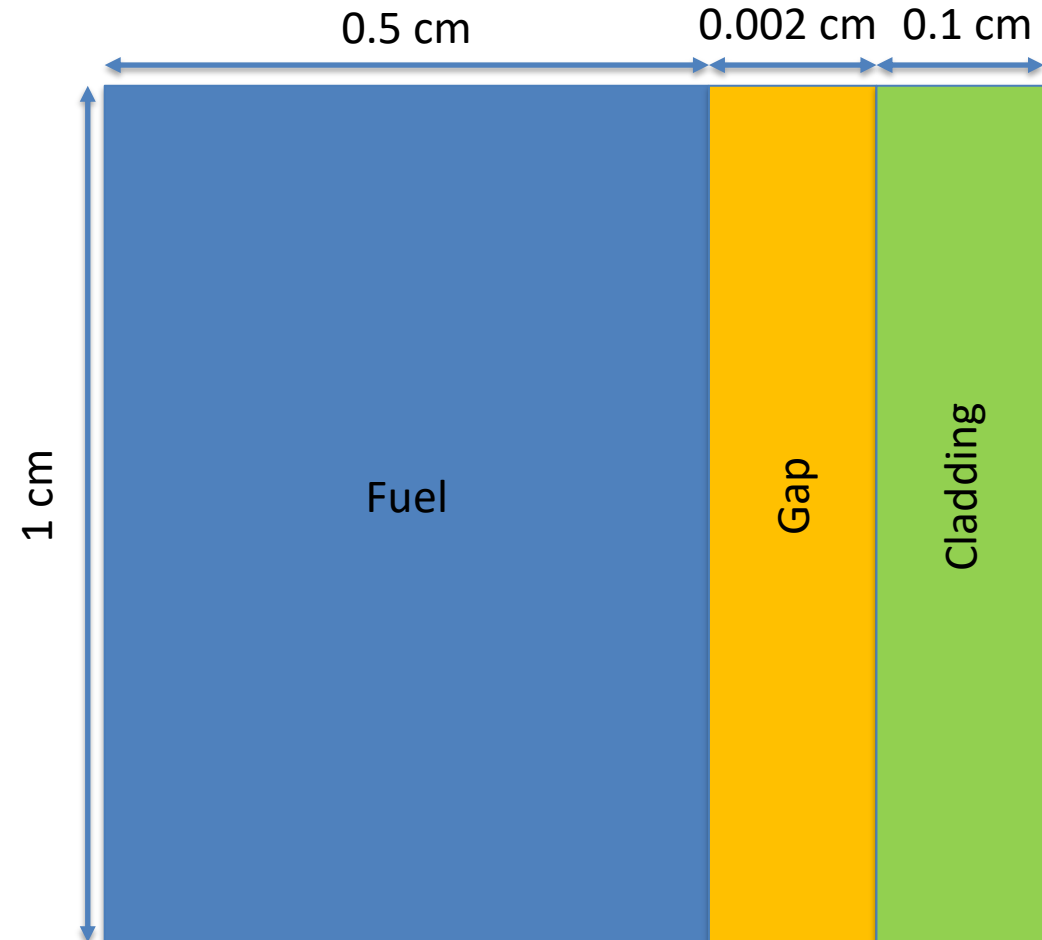
- Fission gas release models are used to understand fission gas experiments and to predict gas release for fuel performance codes
- Spherical grain models predict a fraction of gas release for post-irradiation annealing or for in-pile gas release
- Booth model takes Stage 1 into account; Forsberg-Massih model takes Stage 1 and 2 into account
- Fission gas diffusivity behavior changes with temperature and fission rate

# MOOSE Project

- Three-part project
- Will upload input and output files to Moodle
- Will upload a final written report, 5-10 pages (including figures), times new roman, 12pt, 1.5 space, pdf
- Due April 26 – Tuesday before Finals week
- This is an individual project, but some collaboration is encouraged

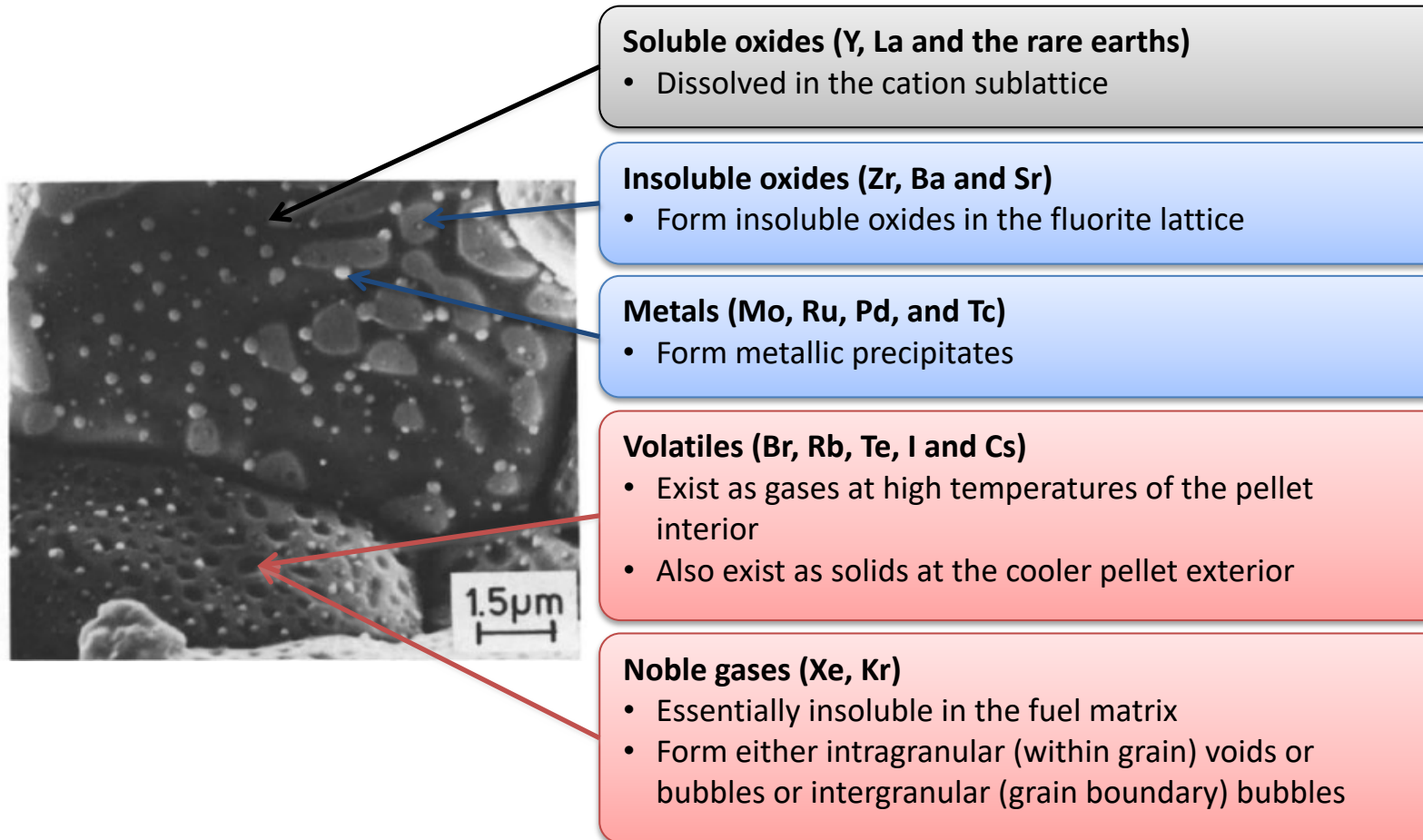
# MOOSE Project Part 1

- Fuel pin dimensions listed
- This is a 1-D problem, but I want your geometry to be set up in 2-D RZ
- Assume reasonable values for thermal conductivities, can assume constant  $k$
- Outer cladding temperature: 500 K
- Mesh: something sufficiently converged
- Solve temperature profile for:
  - Steady-state:  $\text{LHR} = 150 \text{ W/cm}^2$
  - Compare against analytical solution
- Solve for centerline temperature vs time
  - Transient:  $\text{LHR} = 150 \cdot (1 - \exp(-0.05 \cdot \text{time})) + 150$  for up to  $t=100$



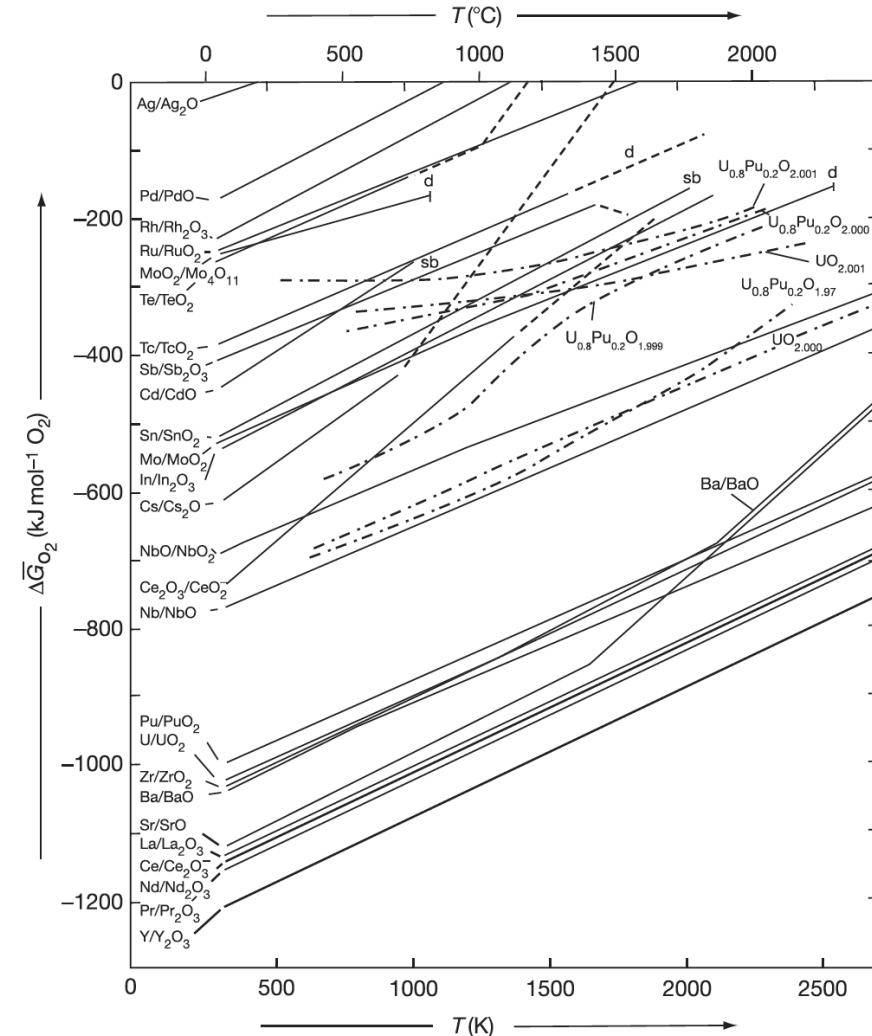
# FISSION PRODUCTS

# There are various types of fission products that form in the fuel



# Oxide Formation

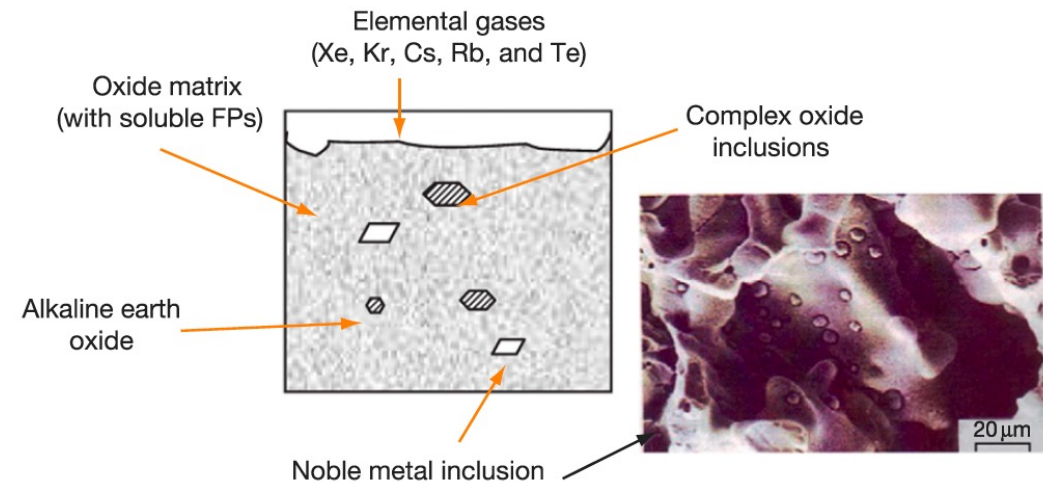
- With burnup, the liberated oxygen will associate with U, Pu, and La but may not be in sufficient supply to combine with Pd and other less oxidizing elements
- If the Gibbs energy of formation of the given fission product lies below the fuel oxygen potential, the element will be capable of forming an oxide
- If the fission product point is above the chemical potential in the fuel, the fission product will exist as an element in the fuel (in a separate metallic phase)



# Fission Products

- When a 4+ valence fission product (e.g.,  $\text{Zr}^{4+}$ ) enters the lattice, there is no change in the electrical neutrality
- If the charge of the fission-product cation is lower than  $\text{U}^{4+}$ , the site occupancy of the lattice is altered to achieve electrical neutrality
- The alkaline earth cations  $\text{Ba}^{2+}$  and  $\text{Sr}^{2+}$  have large ionic radii and form a separate oxide phase
- Fission products that have limited solubility in  $\text{UO}_2$  will segregate to the grain boundaries and voids

Chemical group	Physical state	Probable valence
Zr and Nb	Oxide in fuel matrix; some Zr in alkaline earth oxide phase	4 +
Rare earths	Oxide in fuel matrix	3 +
Ba and Sr	Alkaline earth oxide phase	2 +
Mo	Oxide in fuel matrix or element in metallic inclusion	4 + or 0
Ru, Tc, Rh, and Pd	Elements in metallic inclusion	0 +
Cs and Rb	Elemental vapor or separate oxide phase in cool regions of fuel	1 + or 0
I and Te	Elemental vapor; I may be combined with Cs and CsI	0 or 1 -
Xe and Kr	Elemental gas	0








# Volatile Fission Products


- Xe release occurs at the central region of the pellet where the highest temperatures were achieved during irradiation
- Volatile FPs are released from the fuel matrix similar to that of the noble gases
- Volatile fission products in the gap can react among themselves, resulting in a changing chemical speciation
- The kinetics of formation/decomposition of CsI and Zr iodides are possible factors in the mechanism of SCC

		← A ← T → B →															
I A	II A											III B	IV B	V B	VI B	VII B	
Li	Be											B	C	N	O	F	Ne
Na	Mg	III A	IV A	V A	VI A	VII A		VIII		I B	II B	Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac															
		Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu		
		Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr		

 Volatile fission product

 Metallic precipitates (alloys)

 Ceramic precipitates (oxides)

 Oxides dissolved in the fuel

# Fission Product Segregation

- Fission products can be segregated because of their migration to specific locations such as intragranular segregations, grain boundaries, or pellet surfaces
- Metallic inclusions are commonly observed in ceramographs from irradiated samples
- They are formed by isotopes of Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, and Te
- These inclusions are found at the surface of the grain boundaries and are associated, in general, with grain boundary and intragranular bubbles
- Some of the components of these precipitates can be oxidized or reduced forming other compounds of high volatility that can be released from the fuel matrix
- Cs, Ru, Te, and Ba have been consistently found at the cracks in the pellet and on the clad inner surface
- The white inclusions are metallic precipitates, which are composed of Mo, Tc, Rh, Ru, and Pd, form a quinary alloy in an hcp structure
- A “gray oxide phase” perovskite structure containing different combinations of Ba, Cs, Zr, Mo, and U can also form

## FP Interaction with Zr Cladding

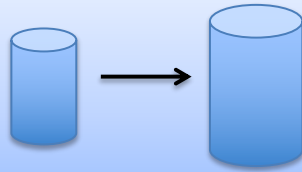
- Fission fragments will deposit on the inner surface of the cladding
- Some of these species can diffuse into the clad, while others can attack the clad thereby initiating cracks that can later progress with the formation of through-wall cracks
- Oxygen will diffuse into the cladding and contribute to its oxidation state
- Measurements have shown deposits of Sr, Cs, Pu, and Am
- The corrosion behavior of zirconium by Te has been reported
- The chemical reaction between some corrosive fission products and the cladding can lead to PCI
- Fission products such as Cs, Cd, or I can attack the cladding inducing crack initiation, which then progress through the cladding by intragranular and transgranular cracking modes

# FUEL SWELLING/DIMENSIONAL CHANGE

# Fuel changes size and shape under reactor operation

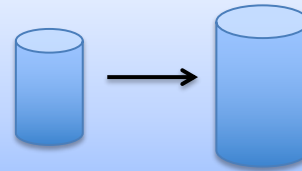
## Thermal Expansion

- Increase in volume
- Decrease in density
- Caused by increasing temperature



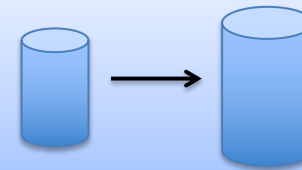
## Densification

- Decrease in volume
- Increase in density
- Caused by shrinking of porosity left after sintering



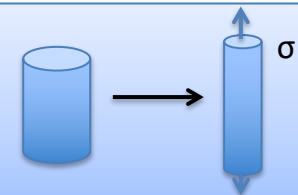
## Swelling

- Increase in volume
- Decrease in density
- Caused by fission products



## Irradiation Creep

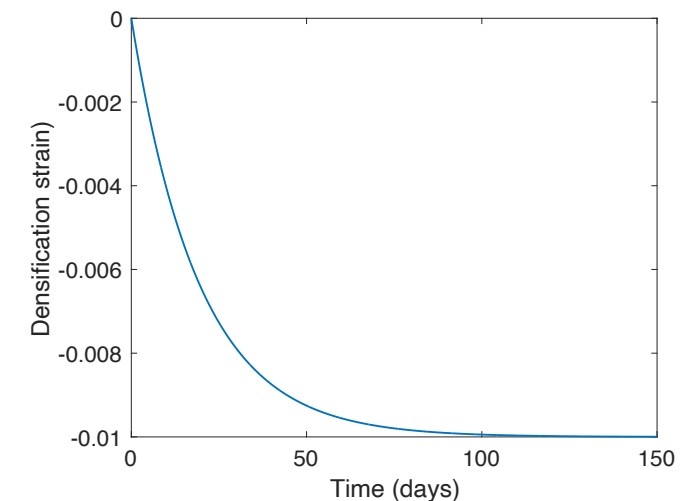
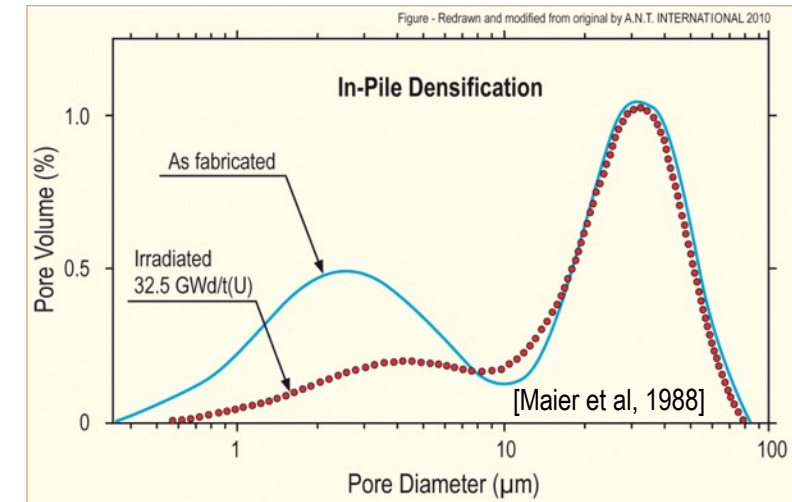
- Change in shape
- Constant density
- Occurs under stress with  $\sigma < \sigma_y$



# Densification

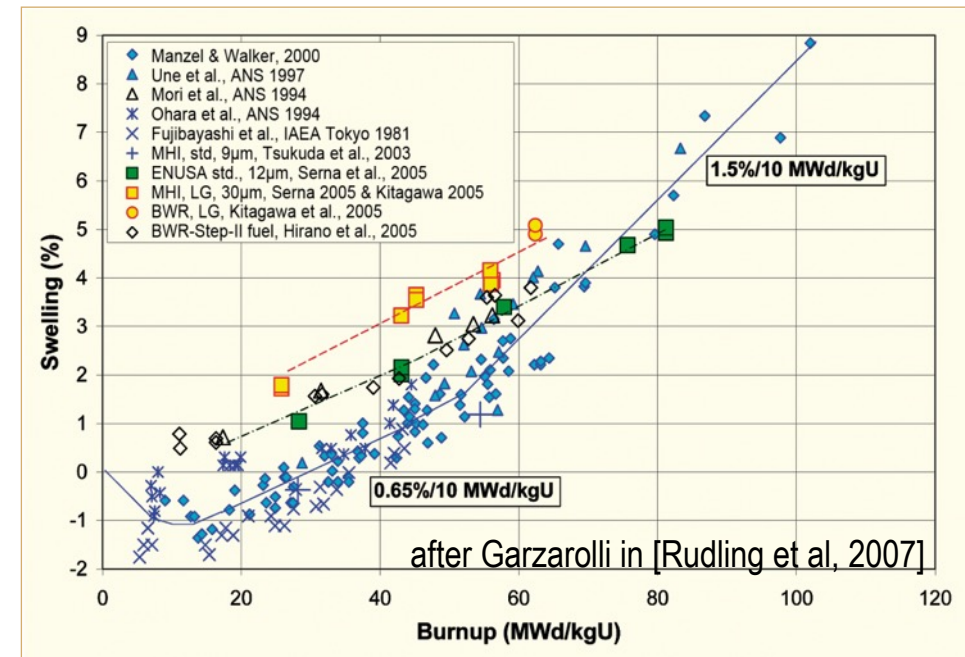
- Densification takes place during initial 5 - 10 MWd/kgU
  - Small, as-built pores close due to effects of fission spikes and vacancy diffusion
  - Large pores stable (in absence of large hydrostatic stress)
- Empirical correlation for densification is a function of
  - $\beta$  - Burnup (in FIMA)
  - $\Delta\rho_0$  - Total densification that can occur (a common value is 0.01)
  - $\beta_D$  - Burnup at which densification stops ( a common value is 5 MWD/kgU)
  - $C_D = 7.235 - 0.0086 (T(^{\circ}\text{C}) - 25)$  for  $T < 750^{\circ}\text{C}$  and  $C_D = 1$  for  $T \geq 750^{\circ}\text{C}$

$$\epsilon_D = \Delta\rho_0 \left( e^{\frac{\beta \ln 0.01}{C_D \beta_D}} - 1 \right)$$



# Fission product induced swelling

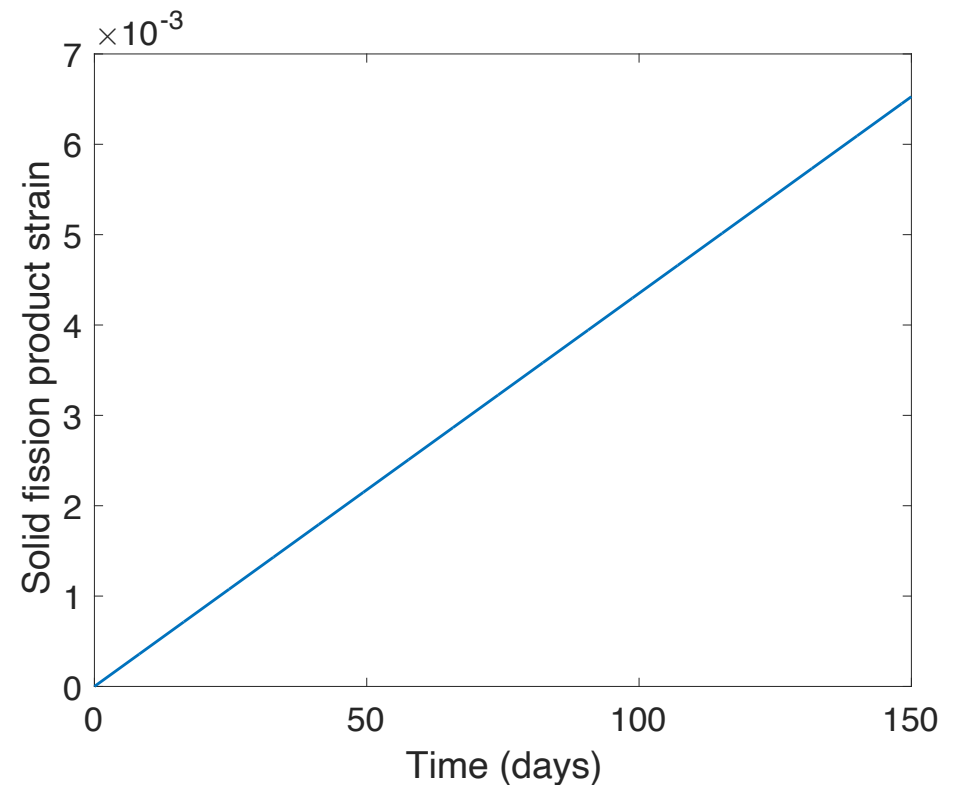
- Fission product swelling results from three changes in the fuel microstructure
  - Solid swelling: Accumulation of soluble and insoluble fission products in fuel matrix
  - Gaseous swelling: Accumulation of gaseous and volatile fission products in intragranular and intergranular pores
  - High burnup swelling: Restructuring of pellet rim with the accumulation of fission gas in a large number of small pores



## Solid fission product swelling

- The solid fission product swelling model is a function of:
  - B – Burnup (in FIMA)
  - $\rho$  – Initial  $\text{UO}_2$  density ( $\text{g/cm}^3$ )

$$\epsilon_{sfp} = 5.577 \times 10^{-2} \rho \beta$$

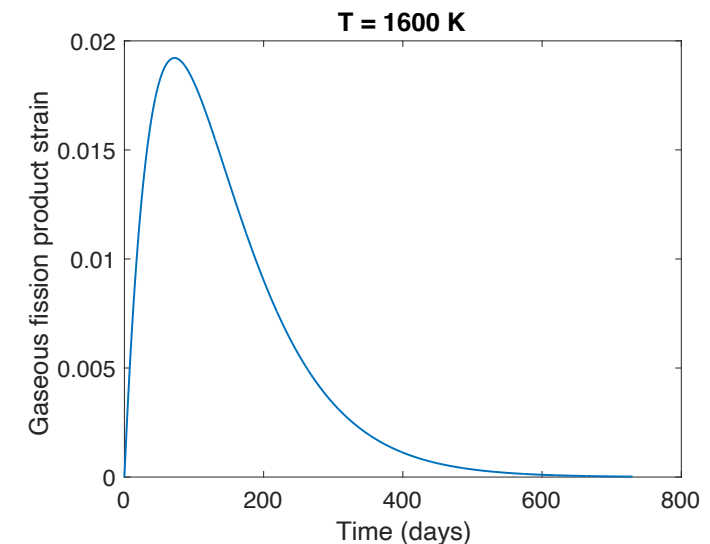
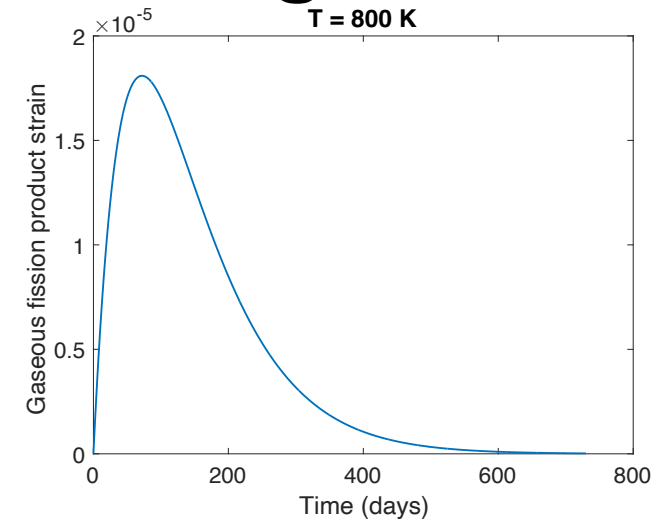




# Gaseous fission product swelling

- Gaseous swelling varies strongly with temperature, fission rate and stress
- $T < 1000\text{K}$ 
  - Fission gas atoms remain in fuel matrix or collect in small, isolated, intragranular pores (<1 nm)
  - Intragranular pore size limited by fission spikes that drive gas back into fuel matrix
  - Gaseous swelling constrained by fission gas release
- $T = 1000$  to  $1700\text{ K}$ 
  - Swelling takes place at hot interior of pellet
  - Gas atoms in fuel matrix diffuse to grain boundaries and collect in pores
  - Gas pressure causes bubbles to increase in size and to coalesce into larger pores
  - Gaseous swelling opposed by applied stress
  - Gaseous swelling also constrained by fission gas release

$$\epsilon_{gfp} = 1.96 \times 10^{-28} \rho \beta (2800 - T)^{11.73} e^{-0.0162(2800 - T)} e^{-17.8 \rho \beta}$$



## Total change in volume

- The total change in volume is found by adding all components of dimensional change
  - $\epsilon_{\text{tot}} = \epsilon_{\text{th}} + \epsilon_{\text{D}} + \epsilon_{\text{sfp}} + \epsilon_{\text{gfp}}$
- Example:
  - fission rate =  $2.5\text{e}13 \text{ f}/(\text{cm}^3 \text{ s})$
  - $T(\text{fuel}) = 1400 \text{ K}$
  - $T_{\text{ref}} = 300 \text{ K}$
  - For densification:  $\Delta\rho_0 = 0.01$  and  $\beta_{\text{D}} = 5 \text{ MWD/kgU}$
  - Total time: 2 weeks

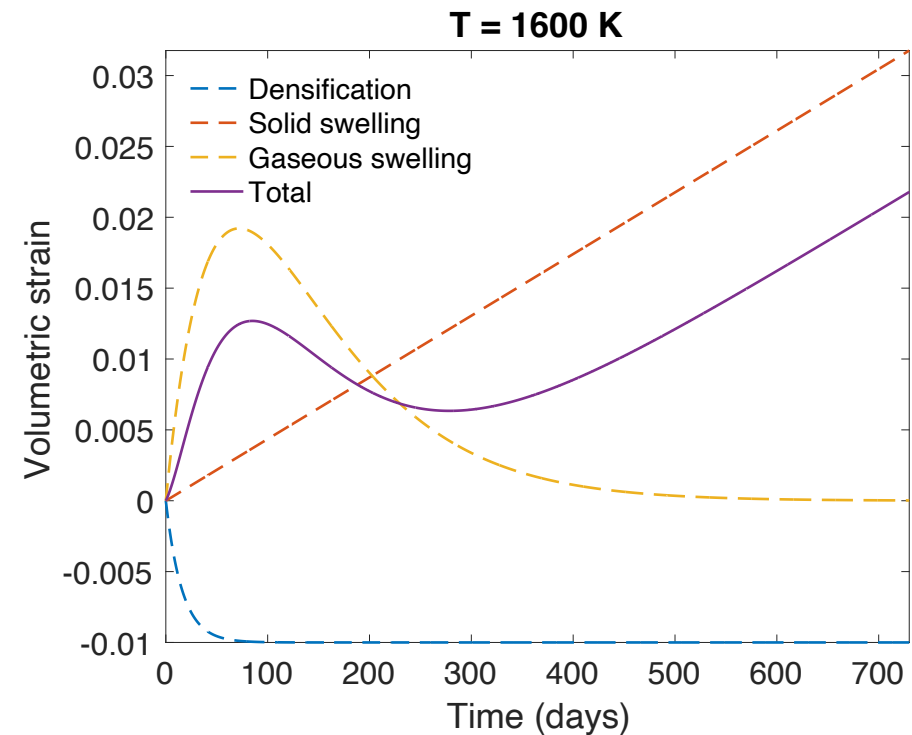
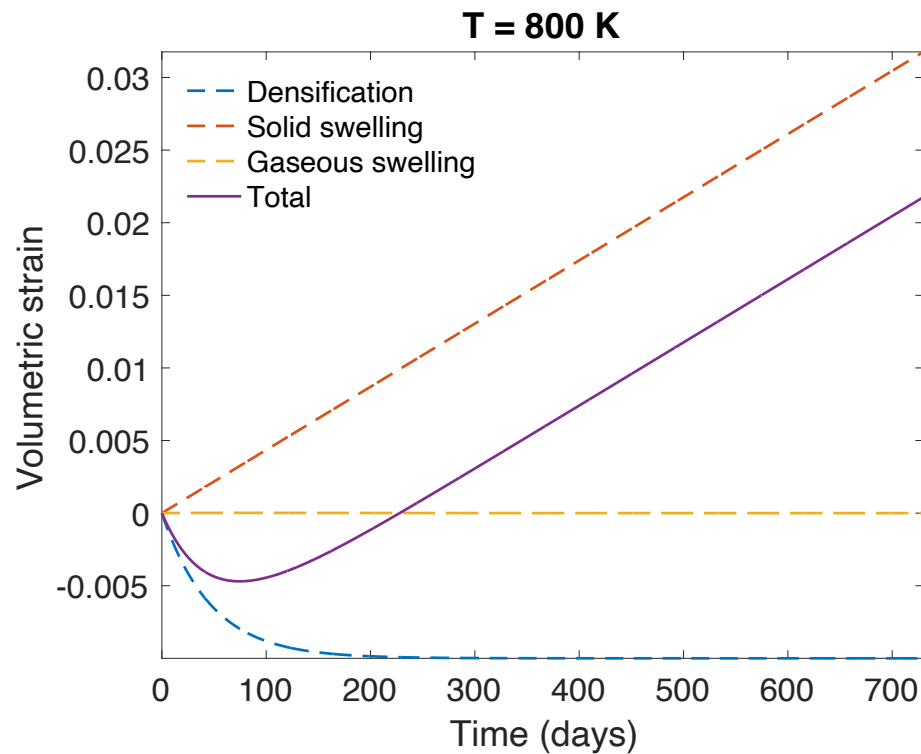
## Change in Volume Example

- First, we need to calculate the burnup, enrich 5%,  $\beta = \dot{F}t/N_U$ 
  - $M_{UO_2} = 16 \cdot 2 + 238 \cdot 0.95 + 235 \cdot 0.05 = 269.9 \text{ g/mol}$
  - $N_U = N_a \rho_U / M_U = 6.022e23 \cdot 10.97 / 269.9 = 2.45e22 \text{ atoms of U/cm}^3$
  - $\beta = 2.5e13 \cdot 3600 \cdot 24 \cdot 7 \cdot 2 / 2.45e22 = 0.0012 \text{ FIMA}$
- Next, we need to determine the strain from thermal expansion
  - $\epsilon_{th} = \alpha \Delta T = 11e-6 \cdot (1400 - 300) = 0.0121$
- Then, we consider densification,  $\epsilon_D = \Delta \rho_0 \left( e^{\frac{\beta \ln 0.01}{C_D \beta_D}} - 1 \right)$ 
  - We need to calculate  $C_D$ , but because we are higher than 750C,  $C_D = 1$
  - We need to convert the burnup to FIMA,  $\beta_D = 5 \text{ MWd/kgU} / 950 = 0.0053 \text{ FIMA}$
  - $\epsilon_D = 0.01 \cdot (\exp(0.0012 \cdot \log(0.01) / (1 \cdot 0.0053)) - 1) = -0.0065$

## Change in Volume Example

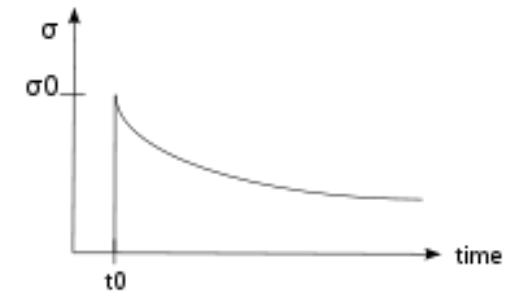
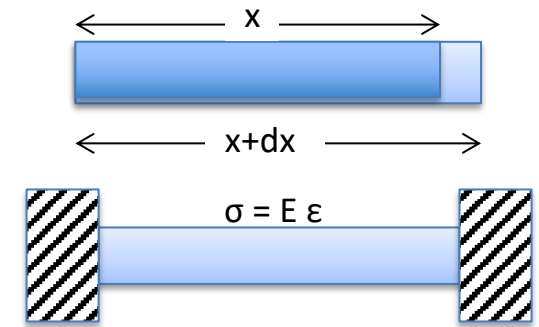
- For solid fission product swelling,  $\epsilon_{sfp} = 5.577 \times 10^{-2} \rho \beta$ 
  - For the density of  $\text{UO}_2$ ,  $\rho = 10.97 \text{ g/cm}^3$
  - $\epsilon_{sfp} = 5.577 \times 10^{-2} \times 10.97 \times 0.0012 = 7.34 \times 10^{-4}$
- Finally, gaseous fission product swelling
 
$$\epsilon_{gfp} = 1.96 \times 10^{-28} \rho \beta (2800 - T)^{11.73} e^{-0.0162(2800 - T)} e^{-17.8 \rho \beta}$$
  - $\epsilon_{gfp} = 1.96 \times 10^{-28} \times 10.97 \times 0.0012 \times (2800 - 1400)^{11.73} \times \exp(-0.0162 \times (2800 - 1400)) \times \exp(-17.8 \times 10.97 \times 0.0012) = 0.0023$
- Total:
  - $\epsilon_{\text{tot}} = \epsilon_{\text{th}} + \epsilon_{\text{D}} + \epsilon_{\text{sfp}} + \epsilon_{\text{gfp}} = 0.0121 - 0.0065 + 7.34 \times 10^{-4} + 0.0023 = 0.0086$

# The overall swelling behavior depends on temperature



# Creep

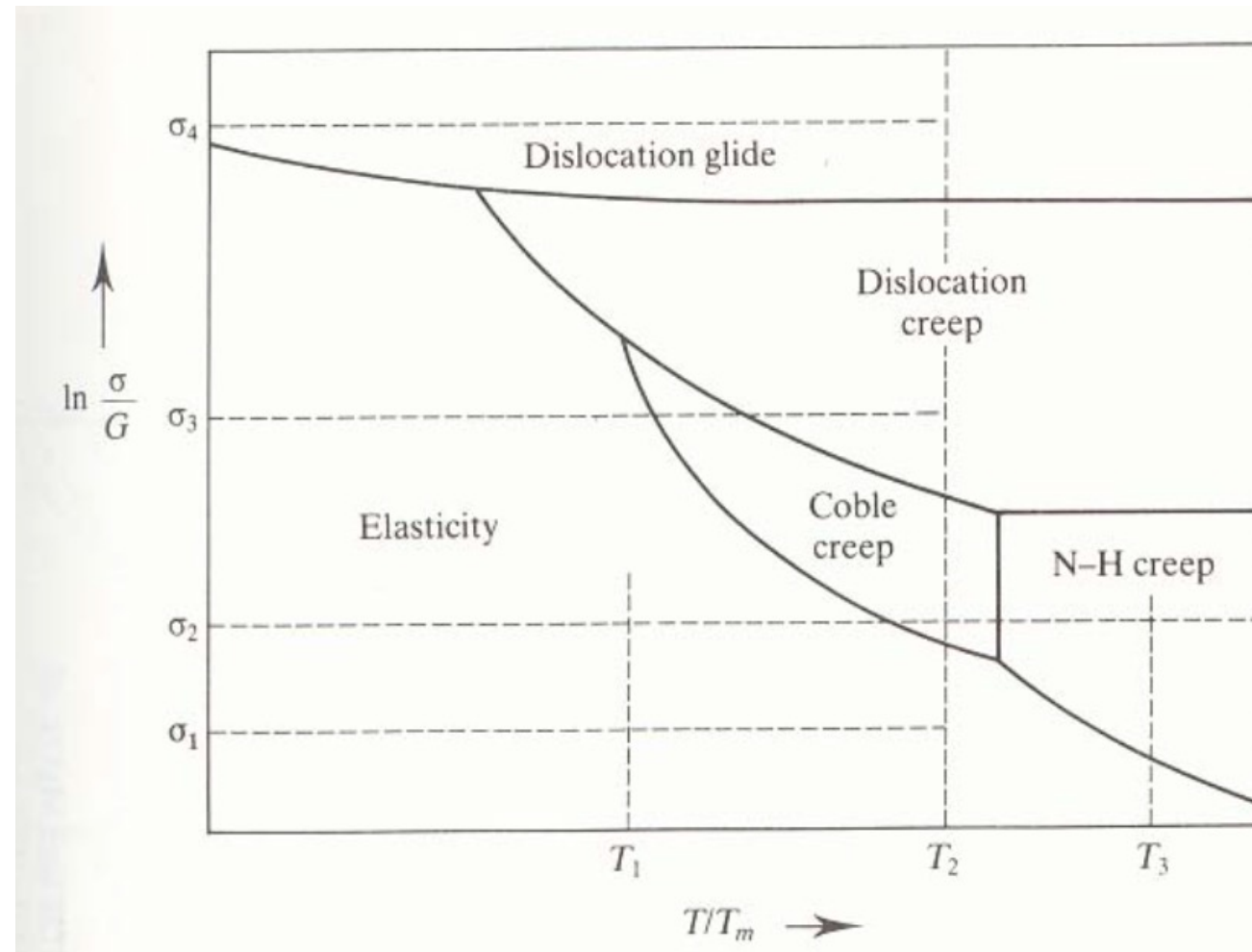
- Creep is a general mechanism for plastic deformation that occurs over time when  $\sigma < \sigma_y$
- Consider a heated metal beam so it expands some distance  $dx$
- We then fix it between two walls and let it cool down
- Because  $\sigma < \sigma_y$ , that stress remains constant
- In creep, defect diffusion is induced by the stress to cause permanent deformation and reduce the stress
- Therefore, creep
  - Occurs over time
  - Increases with increasing number of diffusing defects
    - High temperature (**thermal creep**)
    - Irradiation (**irradiation creep**)



# Creep

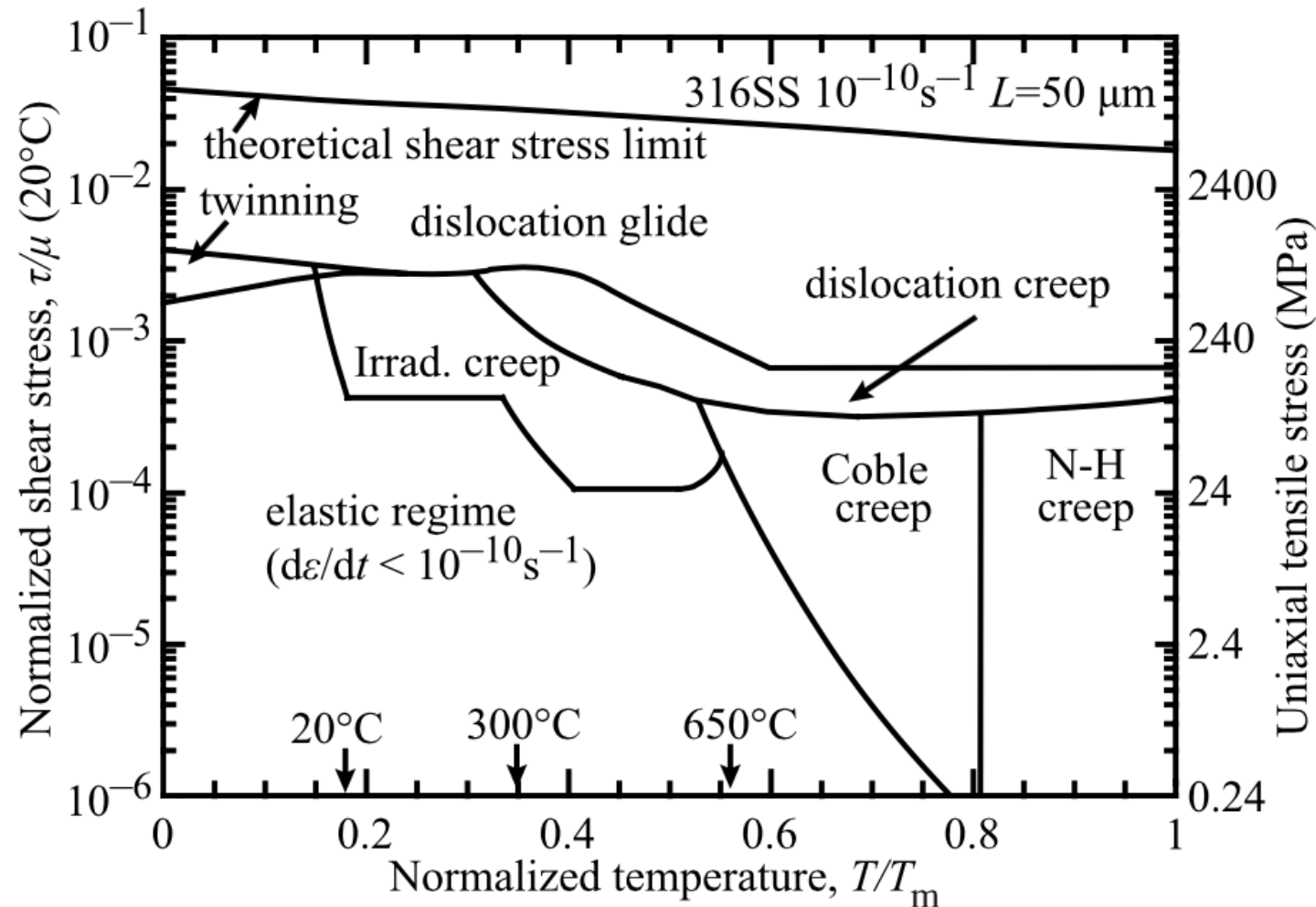
- General creep equation: 
$$\dot{\epsilon} = \frac{C\sigma^m}{D_{gr}^b} e^{\frac{-Q}{k_b T}}$$
- Creep can be caused by various microstructural mechanisms
- Bulk Diffusion (Nabarro-Herring creep)
  - Atoms diffuse (high T), causing grains to elongate along the stress axis
  - $Q = Q(\text{self diffusion})$ ,  $m = 1$ , and  $b = 2$
- Grain boundary diffusion (Coble creep)
  - Atoms diffuse along grain boundaries to elongate the grains along the stress axis
  - $Q = Q(\text{grain boundary diffusion})$ ,  $m = 1$ , and  $b = 3$
- Dislocation creep
  - Dislocations glide under a high stress
  - Dislocations climb due to defects to avoid obstacles
  - $Q = Q(\text{self diffusion})$ ,  $m = 4-6$ , and  $b = 0$

# Different creep mechanisms are active for different combinations of stress and temperature





# The behavior of creep changes in irradiated materials

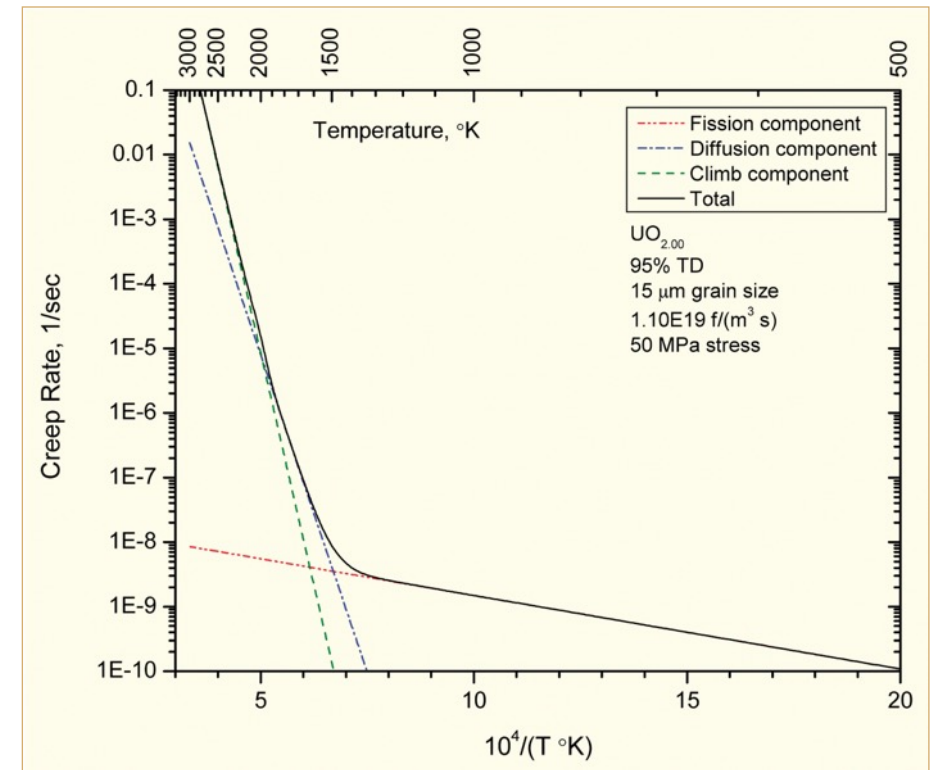


# Irradiation and Creep

- Irradiation accelerates creep, causing it to be significant at lower temperatures
- Irradiation has little effect on diffusional creep, but it accelerates dislocation creep in cubic materials
- The dislocation creep rate can be written as  $\dot{\epsilon} = \rho_d^m b v_d$ 
  - $\rho_d^m$  is the density of mobile dislocations
  - $b$  is the burgers vector
  - $v_d$  is the dislocation velocity
- Gliding dislocations quickly get pinned by obstacles
- As the dislocations absorb defects created by irradiation, they climb to different slip planes to avoid the obstacles
- More interstitials are absorbed than vacancies due to the higher sink strength for interstitials

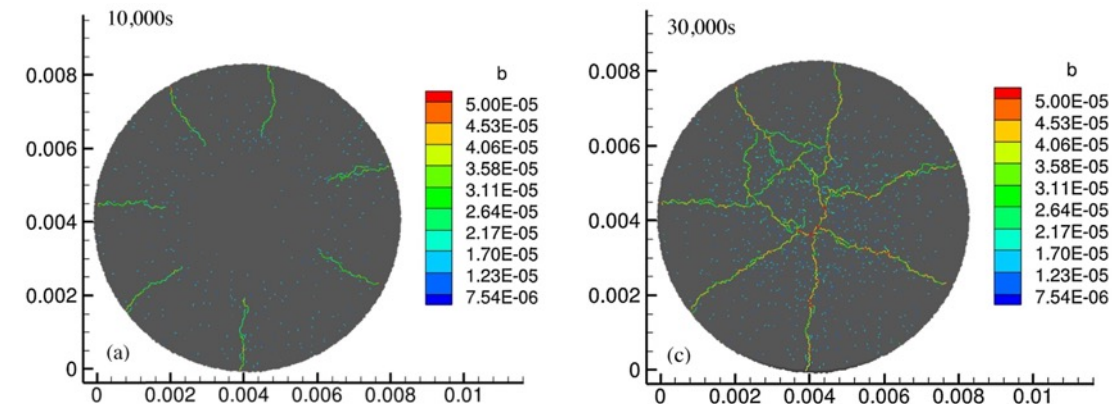
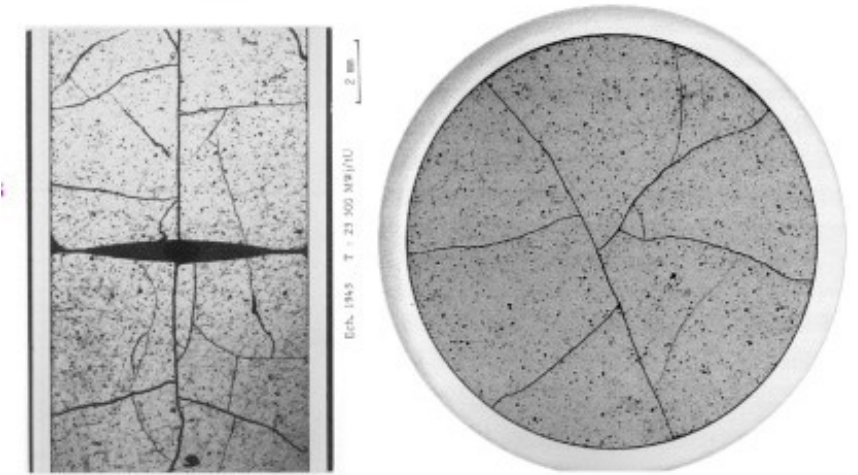
# Fuel Creep

- Like other materials, the fuel also undergoes creep
- The fuel creep (In UO<sub>2</sub>) is a combination of diffusion creep and irradiation creep
- It is expected that fuel creep plays a major role in dimensional change in metallic fuels, largely via N-H and Coble creep, but still unproven experimentally and no good creep models exist for metallic fuels



# Fracture

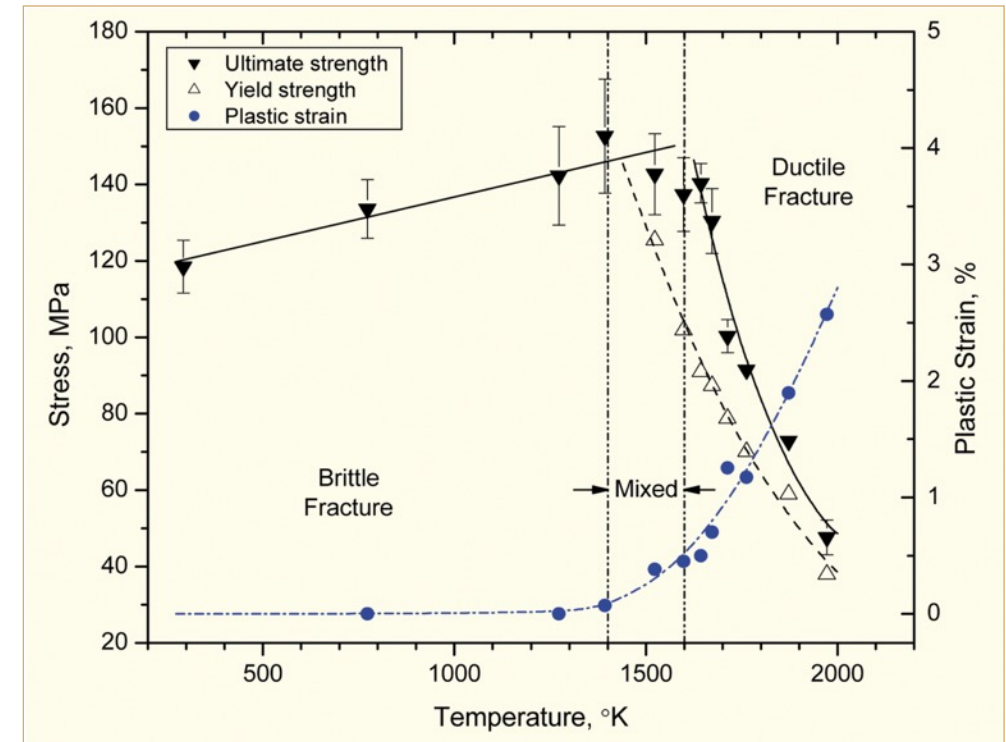
- $\text{UO}_2$  pellets fracture during changes in temperature due to large thermal stresses
- Fracture results in:
  - Increased gap reduction
  - Reduced thermal conductivity
  - Increased avenues for fission gas release
- Fracture has been typically modeled in two ways:
  - Empirical relocation model 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)
  - $\sigma_{\text{frac}} = G^{-m} \sigma_{\text{frac, ref}}$ ,  $m = 0.04 - 0.05$  (vs.  $m \sim 0.5$  for metal)
  - Increasing grain size from  $10 \mu\text{m}$  to  $100 \mu\text{m}$  reduces  $\sigma_{\text{frac}}$  by  $\sim 10\%$
- Ductility transition temperature is lower in-reactor than in thermal tests
- Fracture strength is  $\sim 10 \times$  higher in compression than in tension
- Load-deformation behavior strongly affected by creep under in-reactor conditions



# Summary

- 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 also decreases the gap, as fractures pieces shift outward