

Nuclear Fuel Performance

NE-533
Spring 2024

Upcoming

- Paper presentations next time
- Upload presentations by end of the day March 20
- Will do random number draw to assign presentation order

MOOSE Project

- MOOSE part 1 notes/grade have been emailed back everyone
- I realized that the MOOSE project grading delineation was not clear in the syllabus
 - Part 1: 20%, Part 2: 30%, Part 3: 50%
 - Since this was not included previously, I need agreement
 - Bear in mind later parts factor in the accuracy of earlier parts
- Some notes
 - Do mesh convergence testing! A finer mesh is not always the answer
 - These are reports we are writing, make them such, subsections and so on
 - Do all of the assigned parts
 - Ask questions and come talk to me!

Last time

- Five families of fission products, which change the fuel behavior
 - soluble oxides, insoluble oxides, noble metals, volatiles, noble gases
- Fission gas is released in three stages
 1. Fission gas production and diffusion to grain boundaries
 2. Grain boundary bubble nucleation, growth, and interconnection
 3. Gas transport through interconnected bubbles to free surfaces
- 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

Modeling in-pile release

- The initial gas concentration is 0
- Gas is produced due to fission, where y is the chain yield ($y = 0.3017$ for Xe and Kr) and the fission rate

$$\dot{F} = qN_U\sigma_f{}_{235}\phi_{th}$$

- Gas can also decay, where λ is the decay constant
 - If we only consider stable stable products, $\lambda = 0$
- For in pile release, the fraction is equal to

$$f = \frac{3}{ay\dot{F}t} \int_0^t J_a dt$$

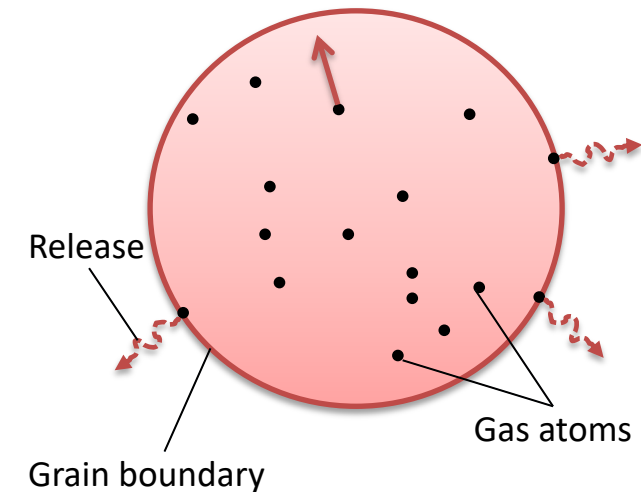
- After solving with with a Laplace transform

$$f = 4\sqrt{\frac{Dt}{\pi a^2}} - \frac{3}{2} \frac{Dt}{a^2} \quad \tau < \pi^2$$

$$f = 1 - \frac{0.0662}{\frac{Dt}{a^2}} \left(1 - 0.93e^{-\pi^2 \frac{Dt}{a^2}}\right) \quad \tau \geq \pi^2$$

- The total gas production is $y\dot{F}t$ gas atoms/cm³

$$\dot{c}_g = y\dot{F} + D \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial c_g}{\partial r} \right) - \lambda c_g$$



ICs and BCs

$$c_g(r, 0) = 0$$

$$c_{g,r}(0, t) = 0$$

$$c_g(a, t) = 0 \text{ (release)}$$

Example

- For a diffusion coefficient for Xe of $D = 8\text{e-}15 \text{ cm}^2/\text{s}$, what fraction of the fission gas trapped in an in-pile fuel pellet has escaped after one hour? It has an average grain size of 10 microns.

- $D = 8\text{e-}15 \text{ cm}^2/\text{s}$

- $a = 10\text{e-}4 \text{ cm}$

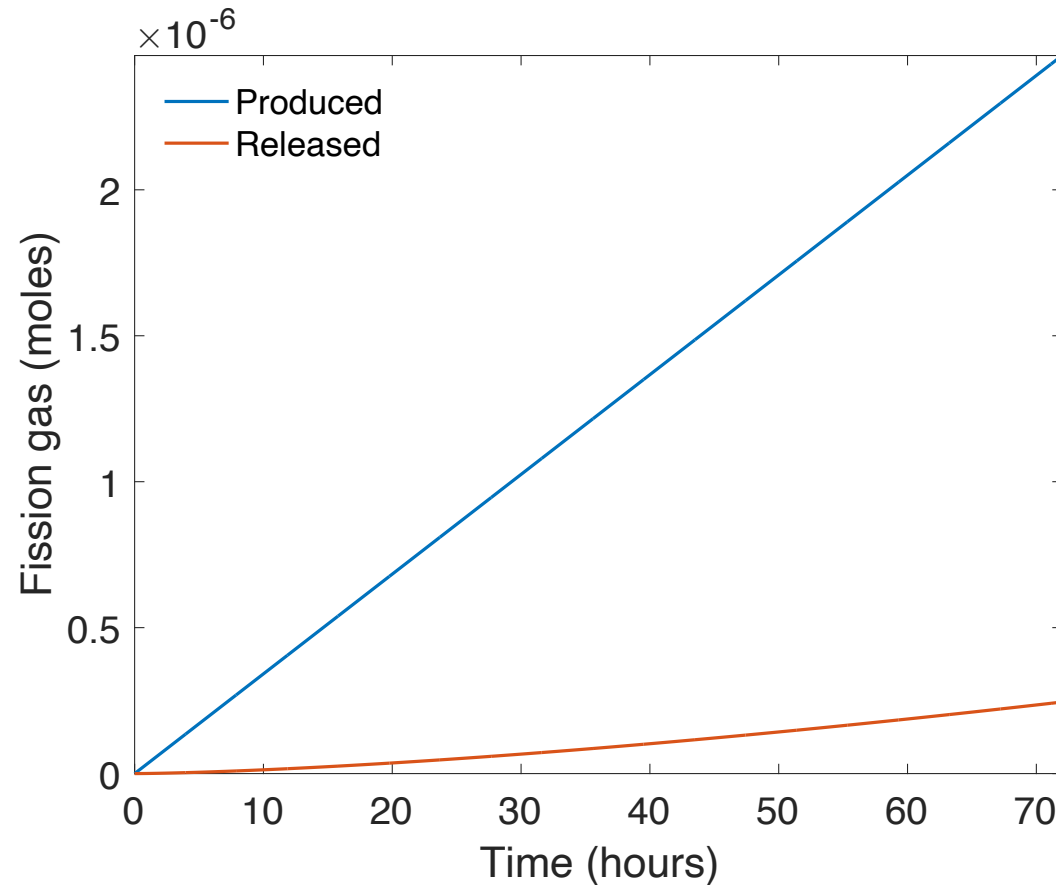
$$\tau = \frac{Dt}{a^2}$$

- We have a short time ($t=3600 \text{ s}$, $\tau < \pi^{-2}$), so we can use:

$$f = 4\sqrt{\frac{Dt}{\pi a^2}} - \frac{3}{2} \frac{Dt}{a^2}$$

- $f = 4*\text{sqrt}(8\text{e-}15*3600/(\text{pi}*(10\text{e-}4)^2)) - 3/2*8\text{e-}15*3600/(10\text{e-}4)^2 = 0.0121$

As time progresses, both the fraction released and the produced gas increase



Forsberg-Massih model

- The Booth model ONLY considers stage one of fission gas release
- Two stage Forsberg-Massih mechanistic model
 - Considers intragranular diffusion diffusion to grain boundaries (stage 1)
 - Also, grain boundary gas accumulation, resolution back into grain, saturation (stage 2)
 - Assumes that once the bubbles on the grain face are interconnected, it is released (no stage 3)

Fission gas is released once the GB coverage reaches a predetermined threshold

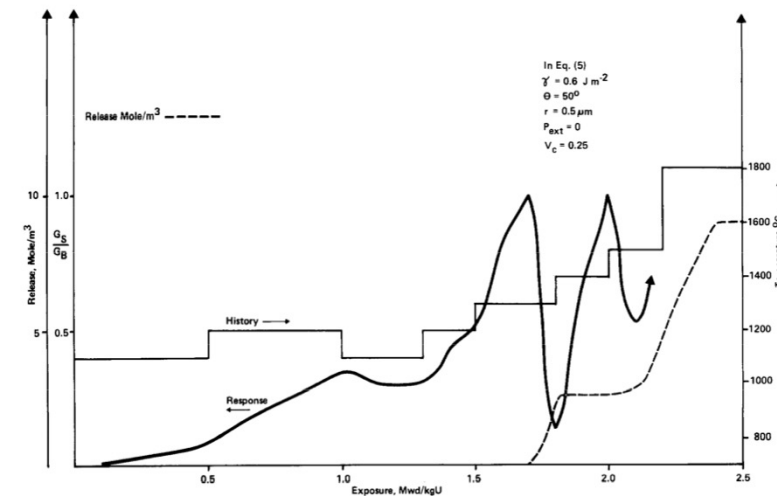
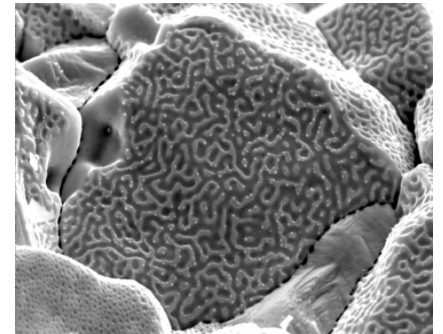
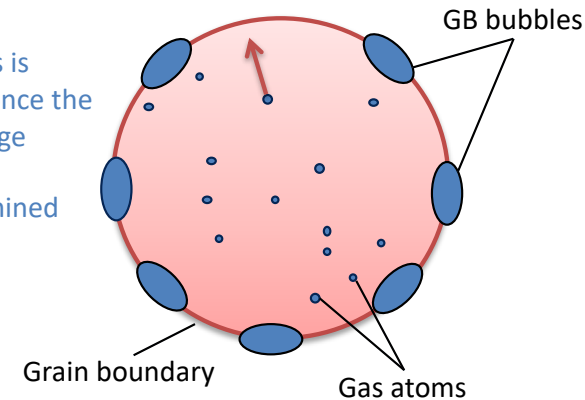
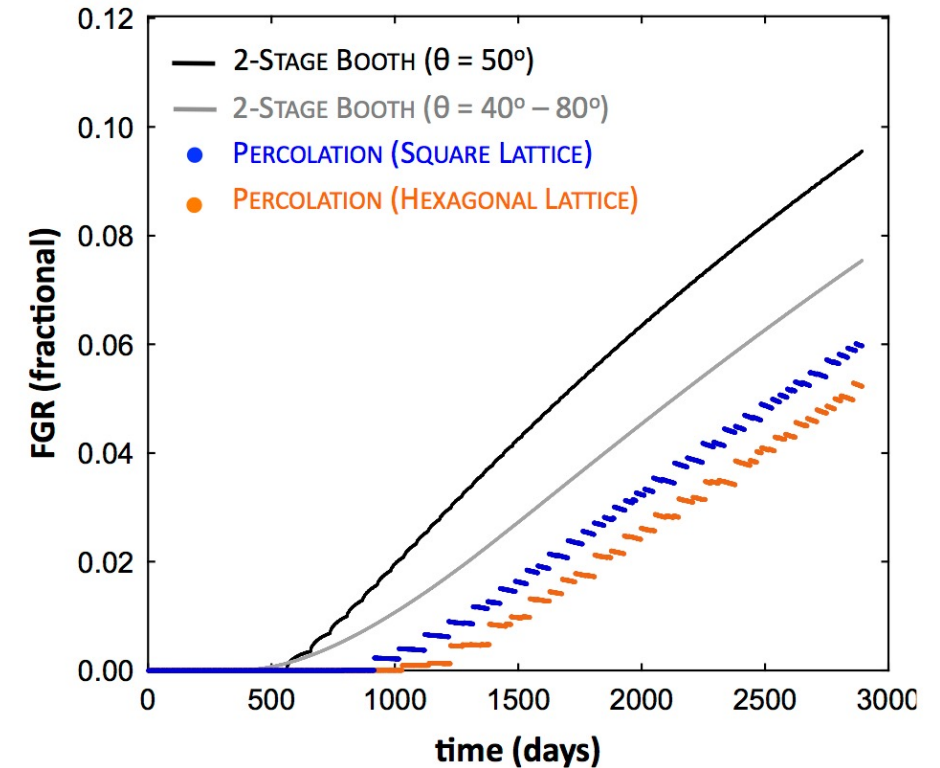
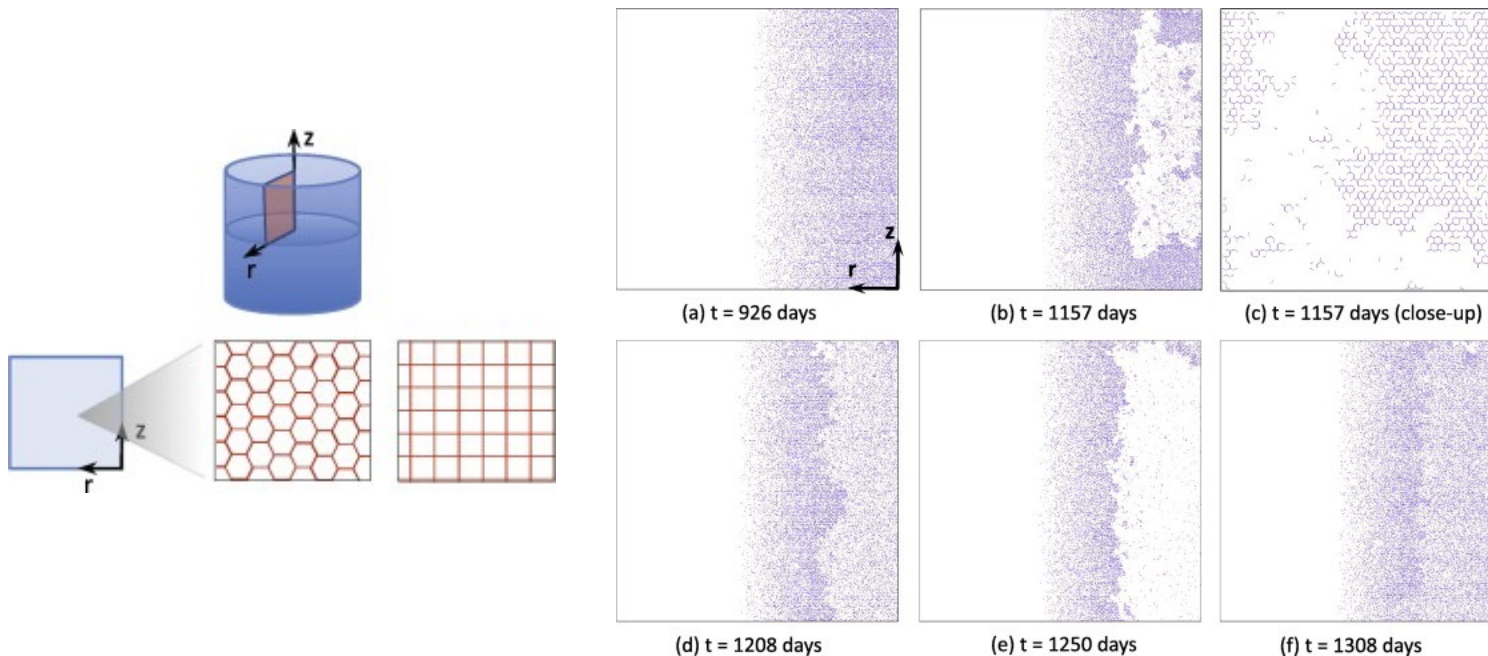


Fig. 1. Fraction of gas atoms on grain boundary, G_s/G_B , as a function of exposure for downward fuel cascading temperature history. γ is the bubble surface tension, 2θ is the angle where two free surfaces meet at a grain boundary, r is average bubble radius, V_c is the fractional coverage of the grain boundaries at saturation and the grain radius is taken to be $5 \mu\text{m}$.

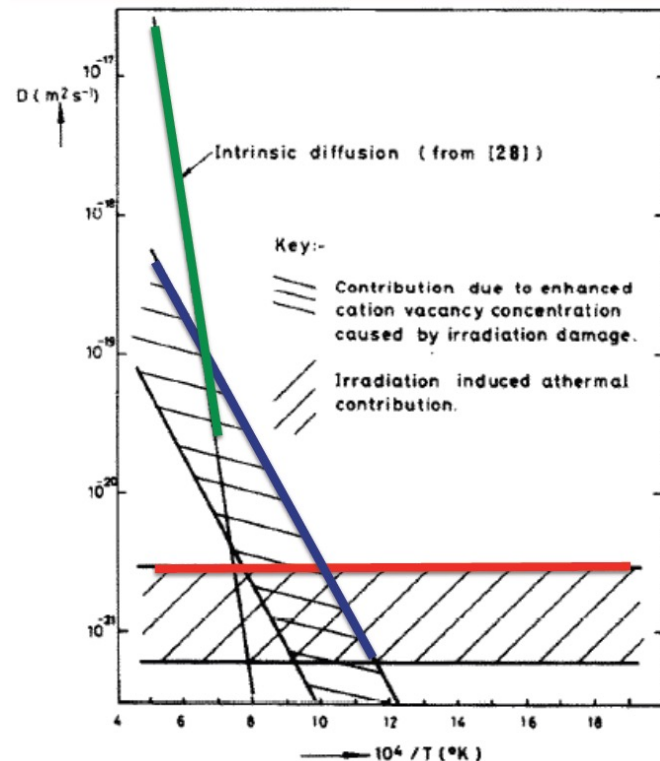
Forsberg-Massih model

- 2-stage F-M model over-predicts gas release because it neglects grain boundary bubble percolation (Stage 3)



Gas diffusion

- The diffusivity of the fission gas depends on temperature and on irradiation
- Experimental data shows three different regimes for the diffusivity

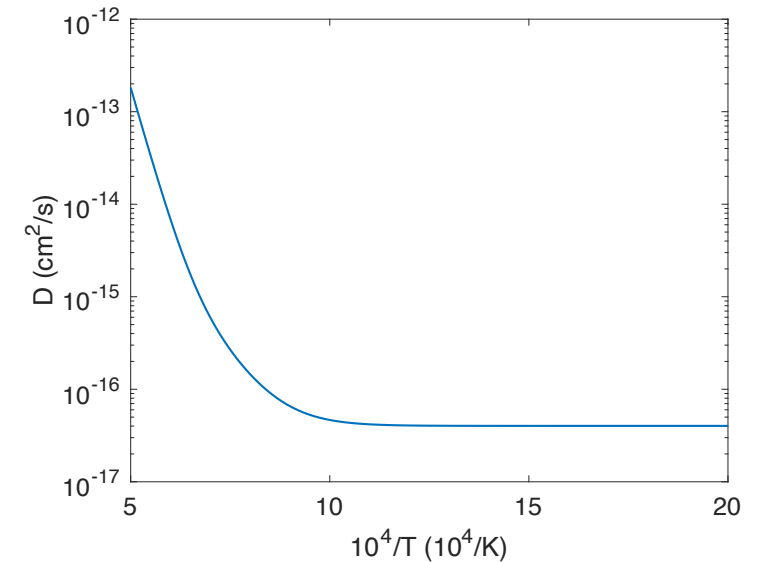


$$D = D_1 + D_2 + D_3 \text{ cm}^2/\text{s}$$

$$D_1 = 7.6 \times 10^{-6} e^{-\frac{3.03 \text{ eV}}{k_b T}}$$

$$D_2 = 1.41 \times 10^{-18} e^{-\frac{1.19 \text{ eV}}{k_b T}} \sqrt{\dot{F}}$$

$$D_3 = 2.0 \times 10^{-30} \dot{F}$$



Gas diffusion

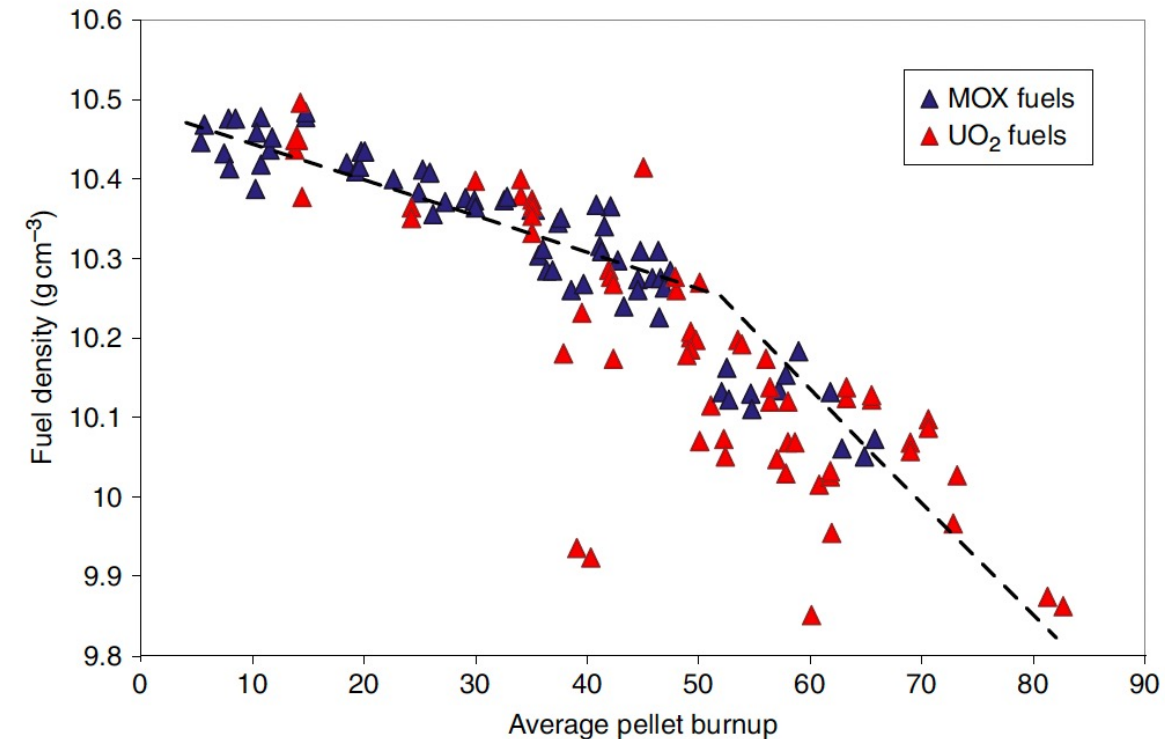
- The effective fission gas diffusivity is slower due to trapping by intragranular bubbles
- As the gas atoms diffuse towards the grain boundary, some are trapped by the small intragranular bubbles
- Some are later knocked out by energized particles (called resolution)
- The effective diffusion constant depends on the trapping rate r_t and the resolution rate r_r

$$D_{eff} = \left(\frac{r_r}{r_r + r_t} \right) D$$

FUEL SWELLING/DIMENSIONAL CHANGE

Fuel changes size and shape under reactor operation

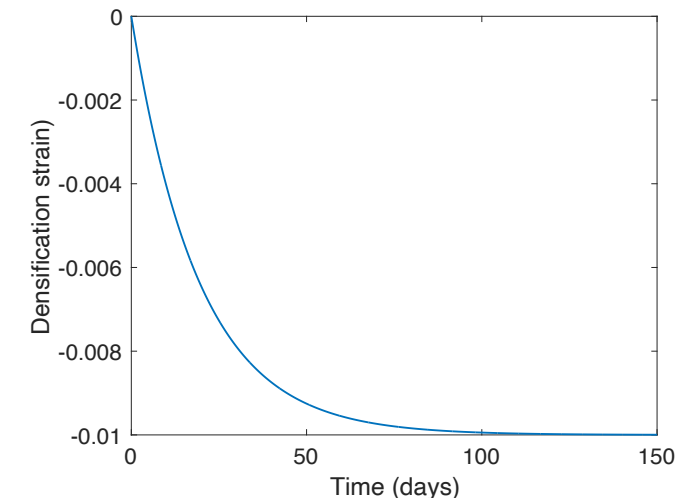
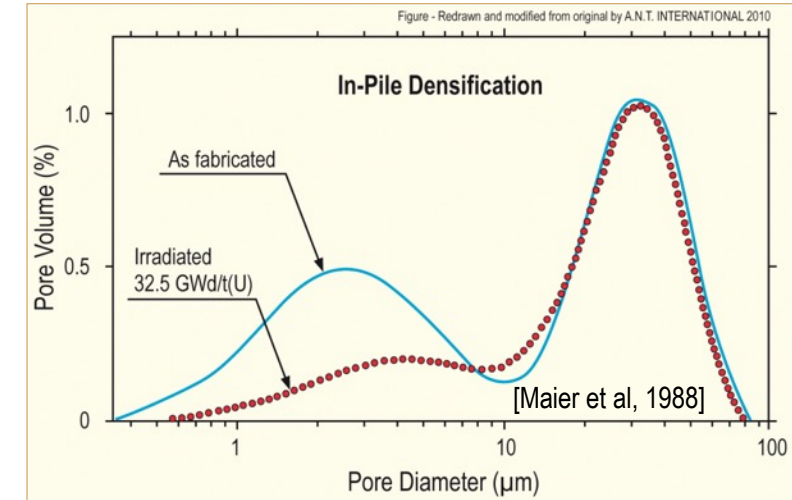
- Thermal expansion:
 - increase in volume, decrease in density, caused by increasing temperature
- Densification:
 - Decrease in volume, increase in density, caused by shrinking of porosity left after sintering
- Swelling:
 - Increase in volume, decrease in density, caused by fission products
- Irradiation Creep:
 - Change in shape, constant density, occurs with applied stress less than σ_y



Densification

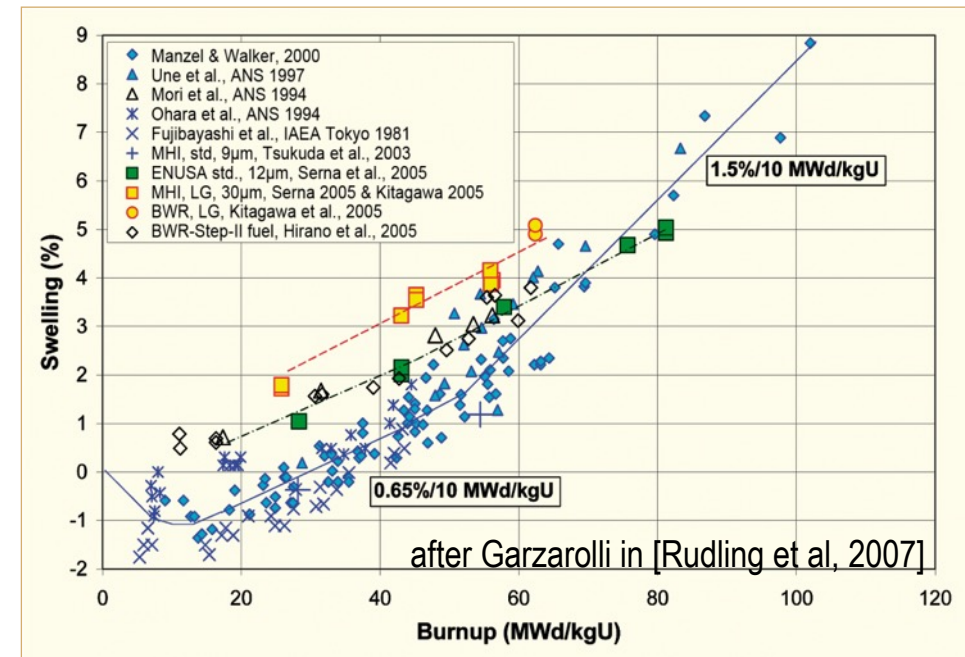
- Densification takes place during initial 5 - 10 MWd/kgU
 - Small, as-built pores close due to effects of fission spikes and vacancy diffusion
 - Large pores stable (in absence of large hydrostatic stress)
- Empirical correlation for densification is a function of
 - β - Burnup (in FIMA)
 - $\Delta\rho_0$ – Total densification that can occur (a common value is 0.01)
 - β_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

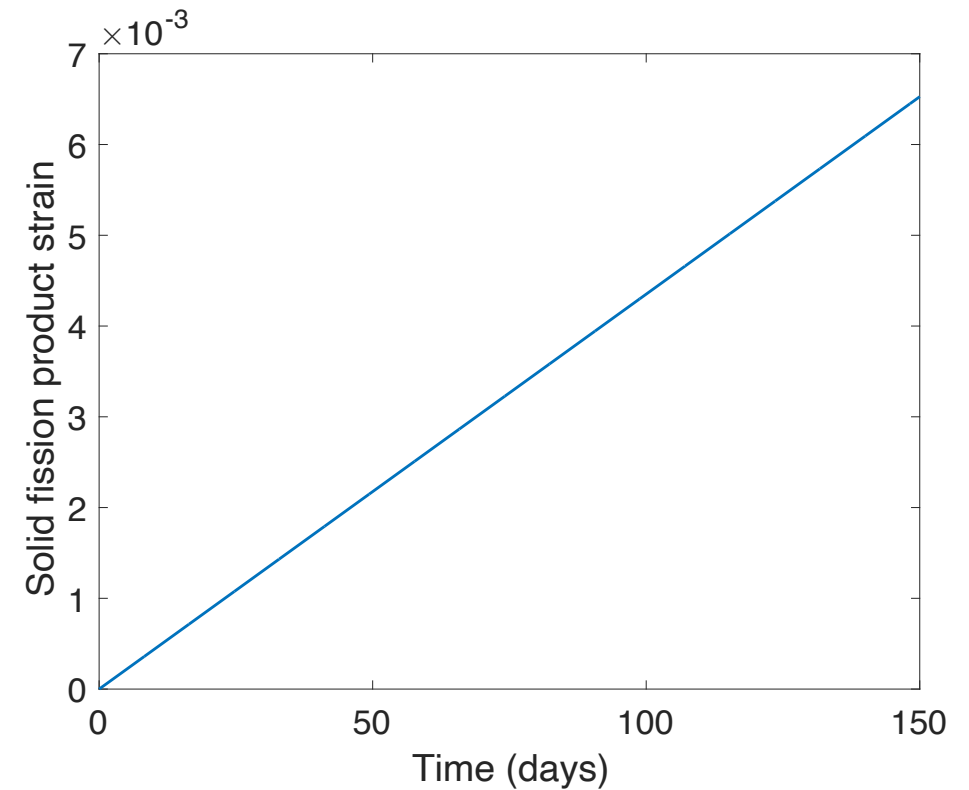
- Fission product swelling results from three changes in the fuel microstructure
 - Solid swelling: Accumulation of soluble and insoluble fission products in fuel matrix
 - Gaseous swelling: Accumulation of gaseous and volatile fission products in intragranular and intergranular pores
 - High burnup 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:
 - β – Burnup (in FIMA)
 - ρ – Initial UO_2 density (g/cm^3)
- Includes contributions from soluble oxides, insoluble oxides, and metallic precipitates

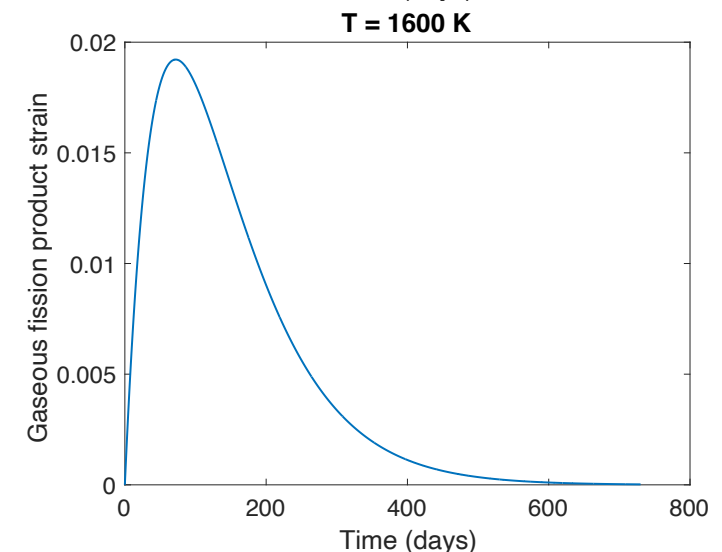
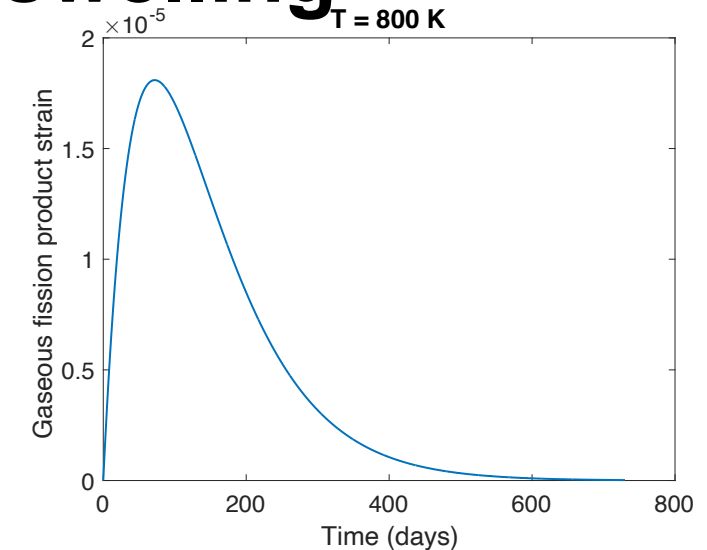
$$\epsilon_{sfp} = 5.577 \times 10^{-2} \rho \beta$$



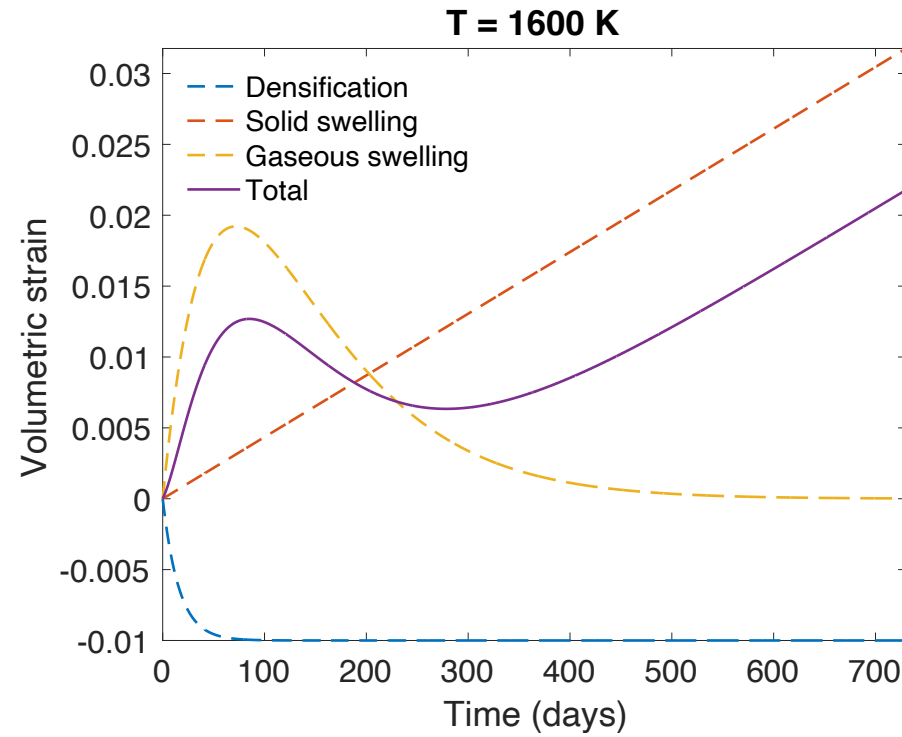
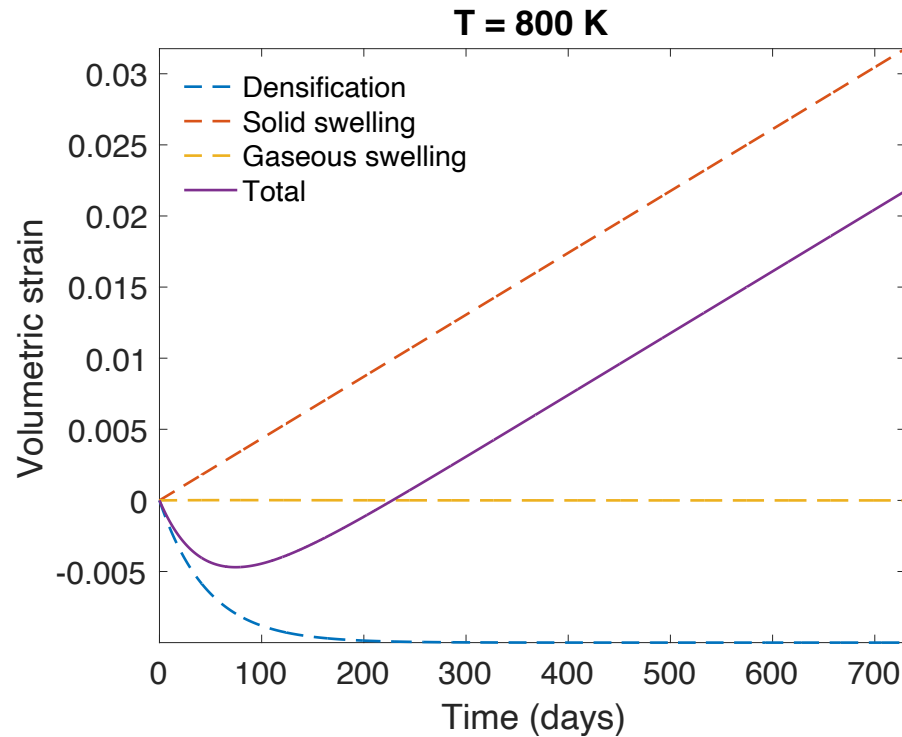
Gaseous fission product swelling

- 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 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

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



The overall swelling behavior depends on temperature



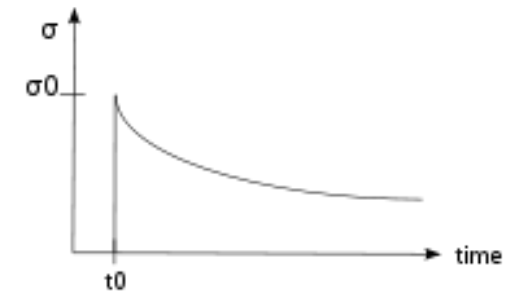
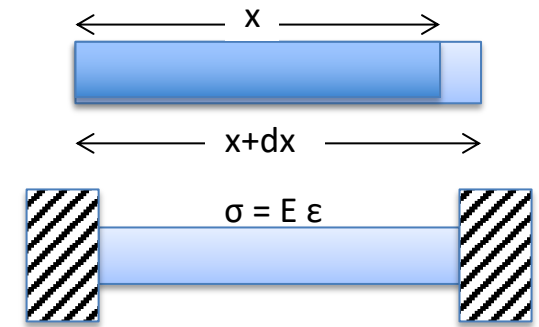
Total change in volume

- 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

Example

Creep

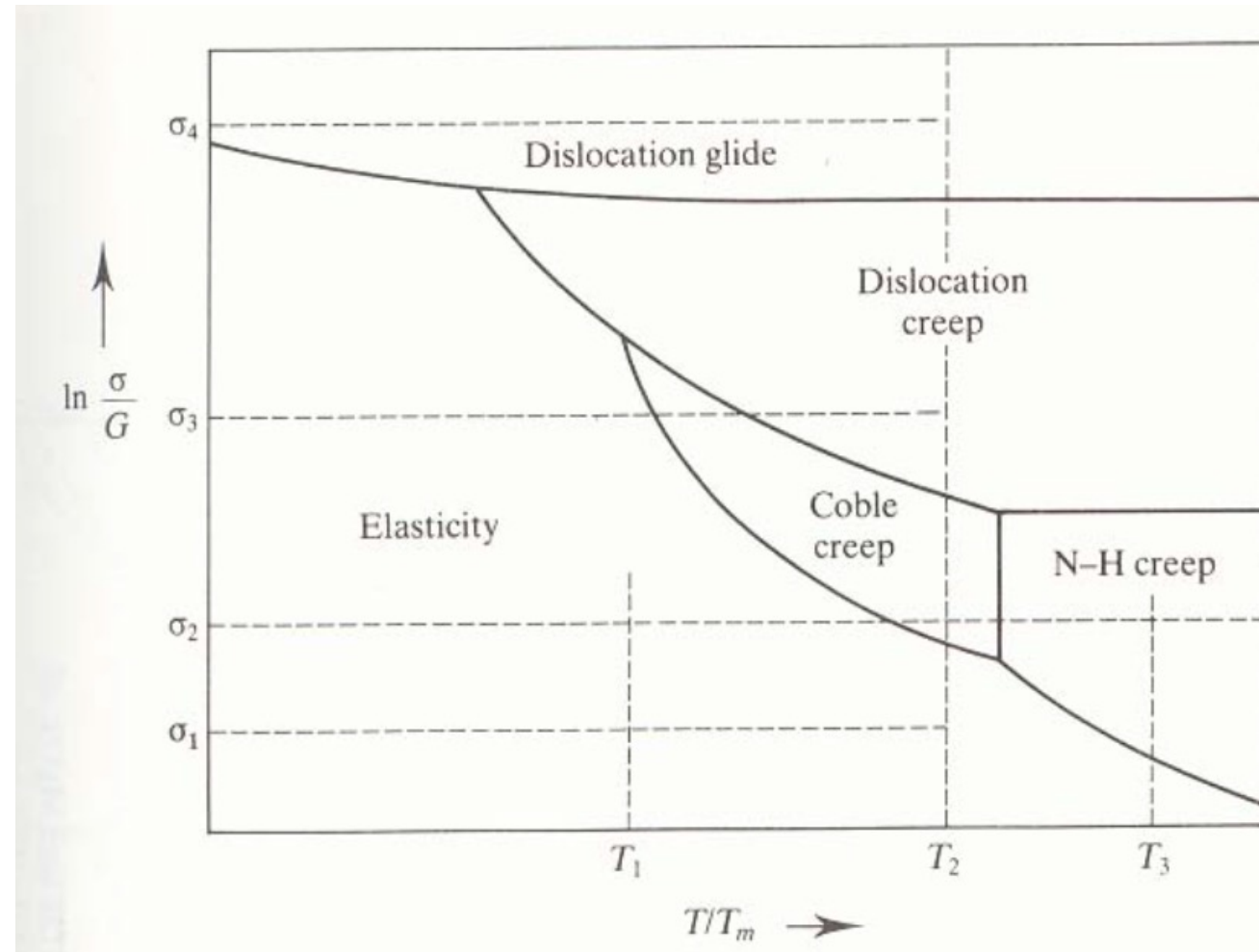
- Creep is a general mechanism for plastic deformation that occurs over time when $\sigma < \sigma_y$
- Consider a heated metal beam so it expands some distance dx
- We then fix it between two walls and let it cool down
- Because $\sigma < \sigma_y$, that stress remains constant
- In creep, defect diffusion 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**)



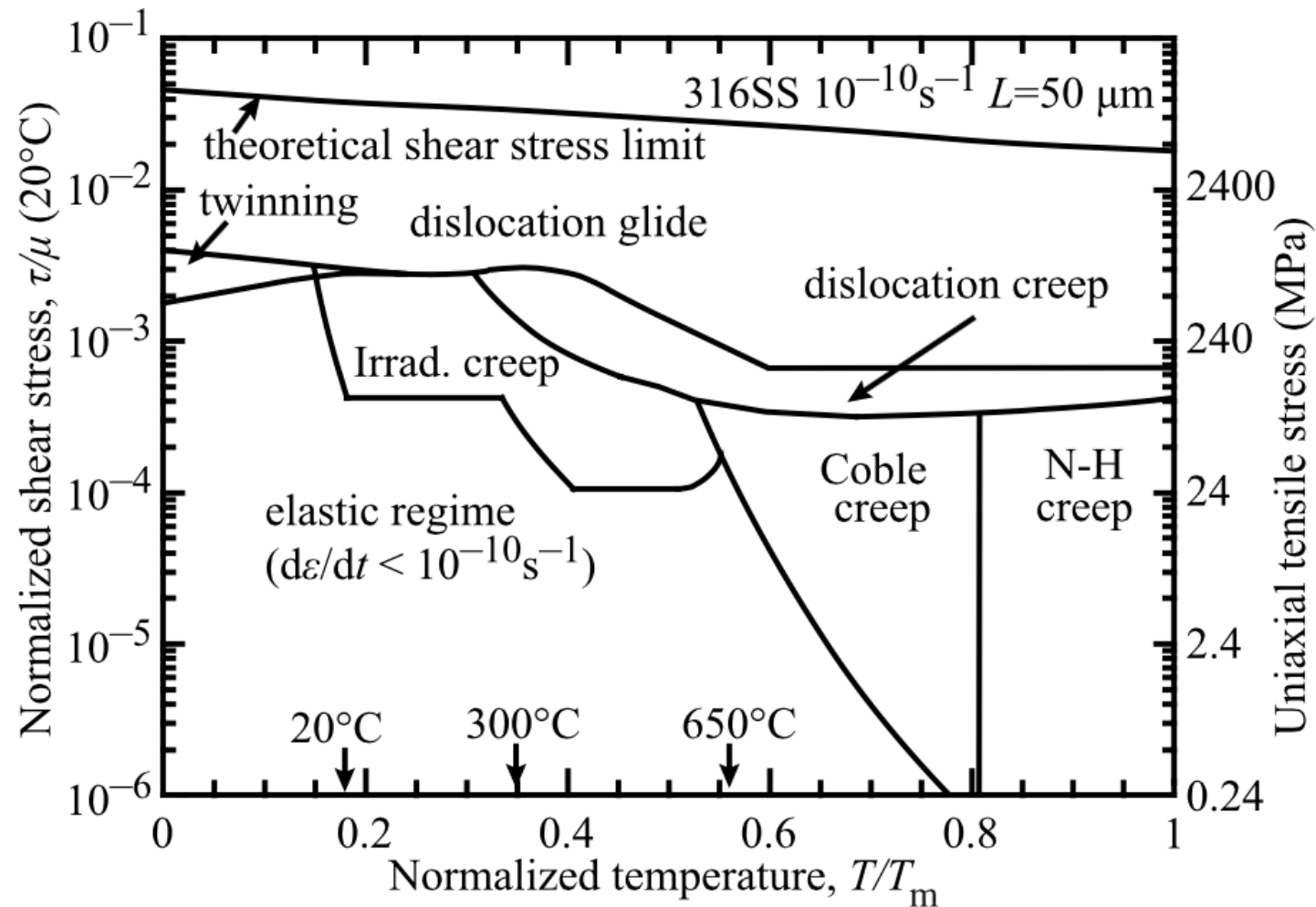
Creep

- 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

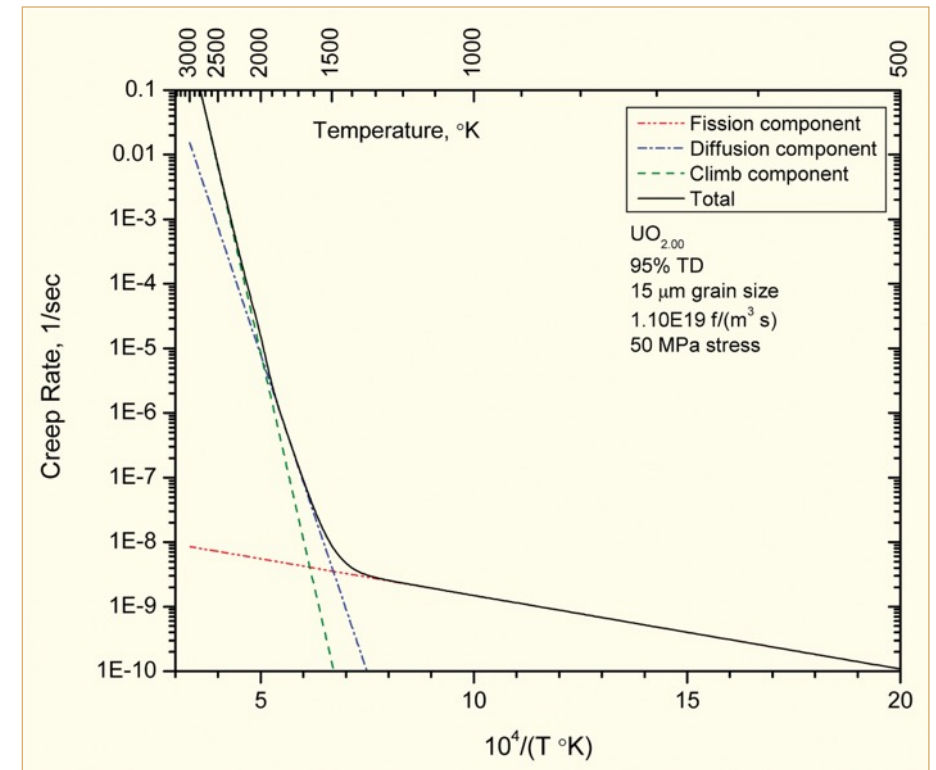


Irradiation and Creep

- 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 $\dot{\epsilon} = \rho_d^m b v_d$
 - ρ_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

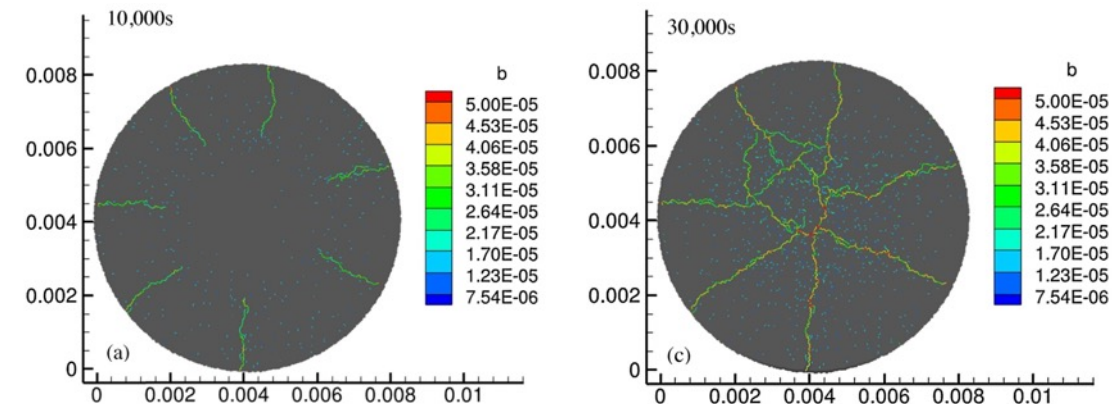
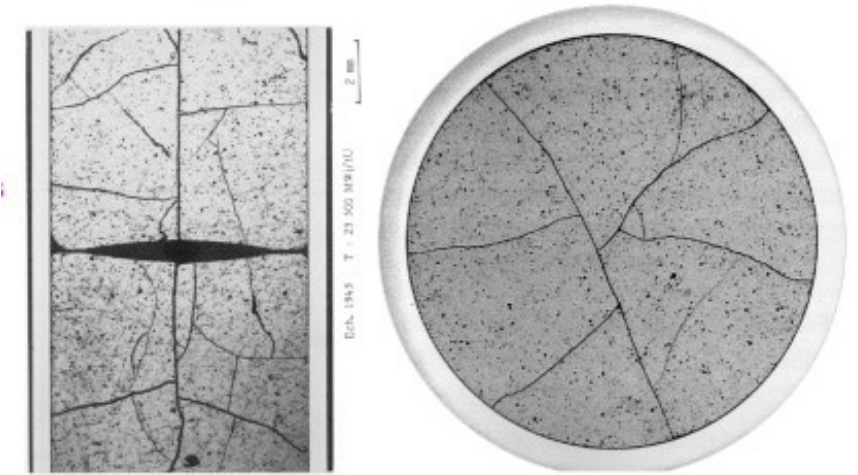
Fuel Creep

- Like other materials, the fuel also undergoes creep
- The fuel creep (In UO₂) 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

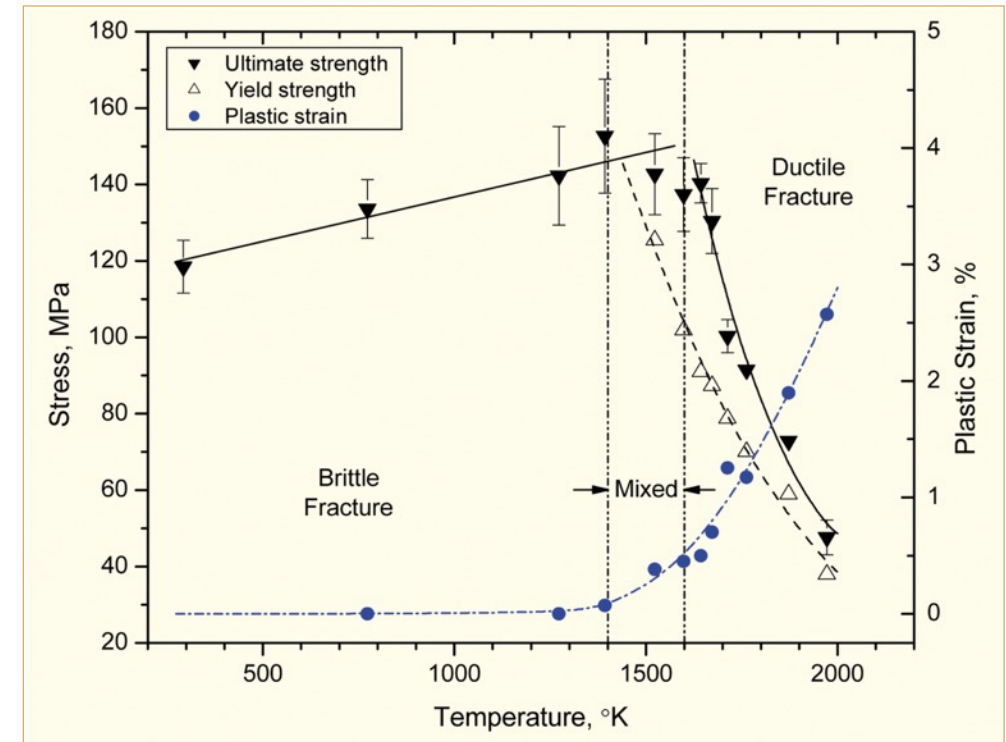
- UO_2 pellets fracture during changes in temperature due to large thermal stresses
- Fracture results in:
 - Increased gap reduction
 - Reduced thermal conductivity
 - Increased avenues for fission gas release
- Fracture has been typically modeled in two ways:
 - Empirical relocation model that is a 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 σ_{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