Nuclear Fuel Performance

NE-533

Spring 2025

Housekeeping

- Gone to TMS next week
- Lectures will be recorded and posted on Panopto
- This means no office hours next week either
- Reach out on slack for questions!
- Can maybe squeeze in a zoom call or two if it is really needed

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

- Released fission gas enters the gap and plenum, causing various problems
- Xe and Kr have very low thermal conductivities, reducing the gap conductance
- The plenum pressure increases
- The volatile fission gases corrode the cladding
- They are also radioactive and hazardous, causing problems when the cladding is breached
- Fission gas release experiments:
  - Post irradiation annealing
    - Fuel is irradiated at low temperature; Fuel is then placed into a furnace and heated; Gas atom release is then measured
  - In-pile release
    - Gas release is measured during reactor operation; It is much more difficult than post-irradiation annealing; Total amount released is measured by puncturing cladding after irradiation; Release with time can be estimated using a pressure transducer inside an instrumented fuel rod

Fission Gas Release

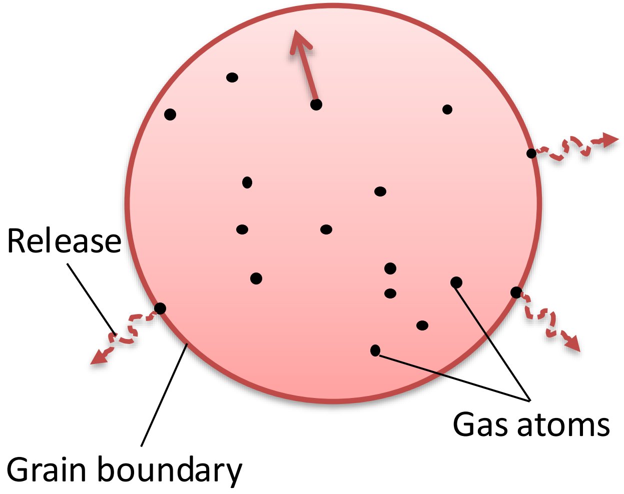
- Fission gas release models attempt to predict the rate at which gas is released from the fuel
- To model fission gas release, ideally, we must model all three stages of gas release
  - Diffusion of gas atoms to grain boundaries
  - Growth and interconnection of grain boundary bubbles
  - Transport of gas atoms through interconnected bubbles to free surfaces
- The earliest models only considered Stage 1
- Most models now consider stage 1 and 2
- There are no models that consider all three stages, but some are under development

Booth Model

$$\dot{c}_g = k_{c_g} + \nabla \cdot D \nabla c_g$$

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

- The Booth model is the earliest model of fission gas release and only considers stage 1
- A grain is considered as a simple sphere
- Gas atoms are released at the grain boundary
- The model solves the diffusion equation in 1D spherical coordinates
- Assumptions
  - $c_g(r, t)$
  - All grains are spheres of radius *a*
  - D is constant throughout the grain
  - Gas is produced uniformly throughout the grain
  - Gas is released once it reaches the grain boundary



<u>ICs and BCs</u>

$c_g(r, 0) = 0$

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

$c_g(a, t) = 0$ (release)

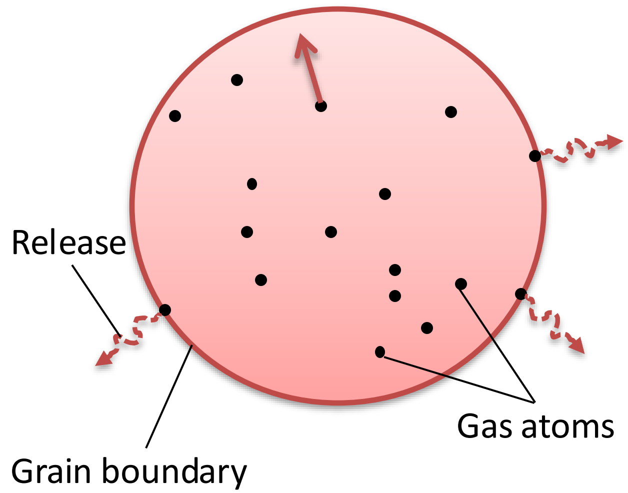
Modeling post-irradiation annealing

- The initial gas concentration is c_g^0
- No gas is produced

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

- Solving this equation tells us the value of c_g at any radius or time
- However, we want to know the fraction of gas atoms that have made it to the grain boundary
- We use the flux at the grain boundary

$$J_a = -D\left(\frac{\partial c_g}{\partial r}\right)_a \qquad f = \frac{4\pi a^2 \int_0^t J_a dt}{4/3\pi a^3 c_g^0} = \frac{3}{a c_g^0}\int_0^t J_a dt$$



ICs and BCs

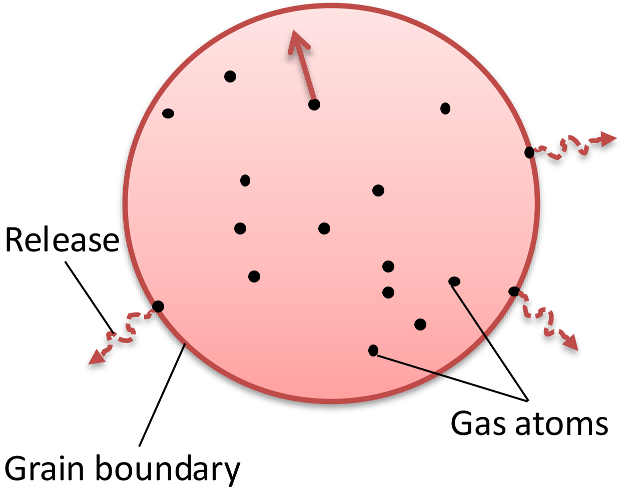
$c_g(r, 0) = c_g^0$

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

$c_g(a, t) = 0$ (release)

Solving the Booth Model

- This equation is solved using a Laplace transform after nondimensionalization
- Will not go through the derivation (shown in Olander)
- | = D x t / a^2



$$f = 6\sqrt{\frac{Dt}{\pi a^2}} - 3\frac{Dt}{a^2} \qquad \tau < \pi^{-2}$$

$$f = 1 - \frac{6}{\pi^2} e^{-\pi^2 \frac{Dt}{a^2}} \qquad \tau \geq \pi^{-2}$$

Booth Example

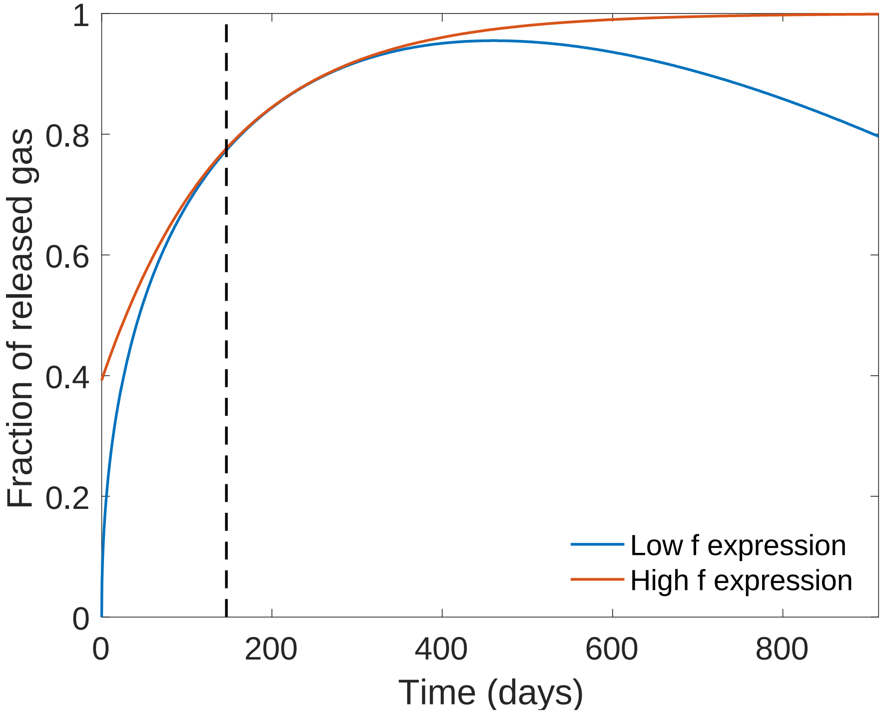
- For a diffusion coefficient for Xe of D = 8e-15 cm^2/s, what fraction of the fission gas trapped in a post-irradiation annealed fuel pellet has escaped after one hour? It has an average grain size of 10 microns
  - D = 8e-15 cm^2/s
  - a = 10e-4 cm
  - t = 3600 s
- Which f? | = D x t/a^2 = 2.88E-4 < π^{-2} = 0.101

$$f = 6\sqrt{\frac{Dt}{\pi a^2}} - 3\frac{Dt}{a^2}$$

  - f = 6*sqrt(8e-15*3600/(pi*(10e-4)^2)) – 3*8e-15*3600/(10e-4)^2 = 0.0181

Different expressions for fission gas release

- Given the data from the previous example, can plot both
  - $\tau < \pi^{-2}$ $$f = 6\sqrt{\frac{Dt}{\pi a^2}} - 3\frac{Dt}{a^2}$$
  - $\tau > \pi^{-2}$ $$f = 1 - \frac{6}{\pi^2} e^{-\pi^2 \frac{Dt}{a^2}}$$



Modeling in-pile release

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

- 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

$$\dot{F} = qN_U\sigma_{f235}\phi_{th}$$

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

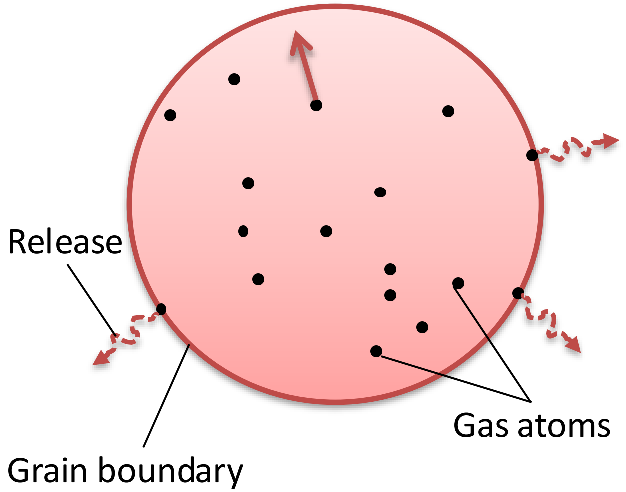
$$f = \frac{3}{ay\dot{F}t}\int_0^t J_a\,dt$$

- After solving with with a Laplace transform

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

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

- The total gas production is $y\dot{F}t$ gas atoms/cm^3



<u>ICs and BCs</u>

$c_g(r, 0) = 0$

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

$c_g(a, t) = 0$ (release)

Example

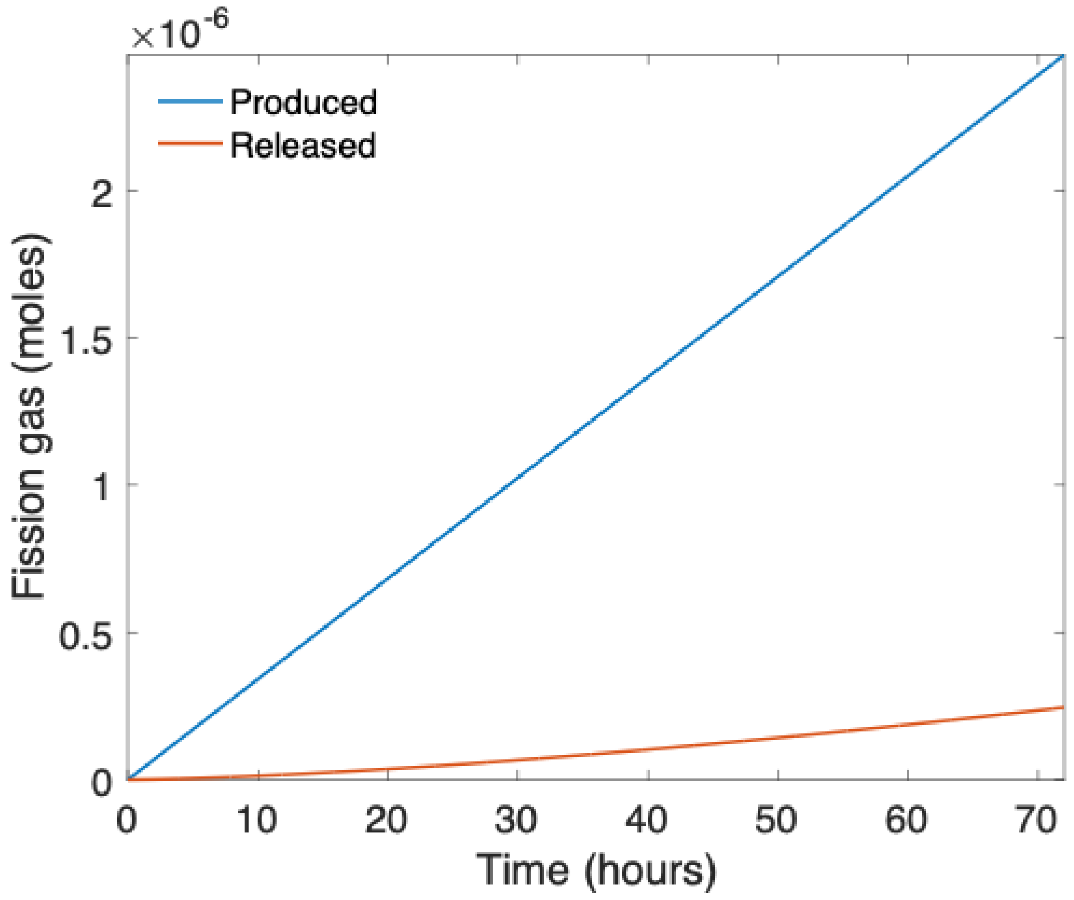
- For a diffusion coefficient for Xe of D = 8e-15 cm^2/s, what fraction of the fission gas trapped in an in-pile fuel pellet has escaped after one hour? It has an average grain size of 10 microns.
  - D = 8e-15 cm^2/s
  - a = 10e-4 cm
  - We have a short time (t=3600 s, $\tau < \pi^{-2}$), so we can use:

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

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

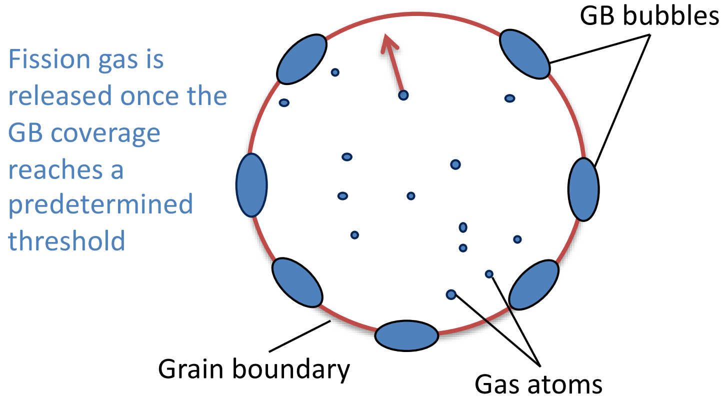
  - f = 4*sqrt(8e-15*3600/(pi*(10e-4)^2)) – 3/2*8e-15*3600/(10e-4)^2 = 0.0121

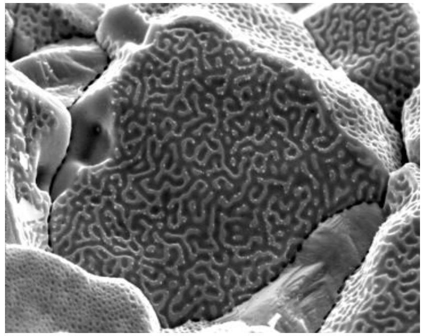
As time progresses, both the fraction released and the produced gas increase



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)





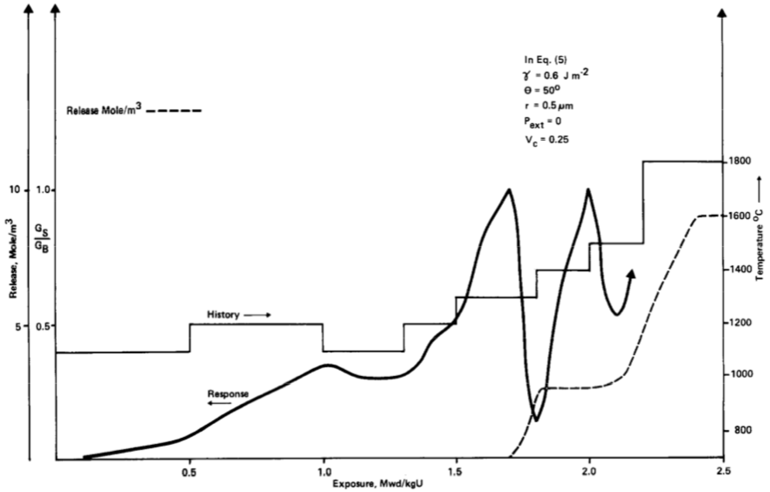
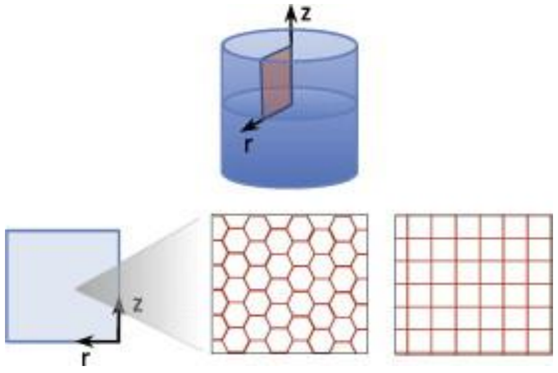
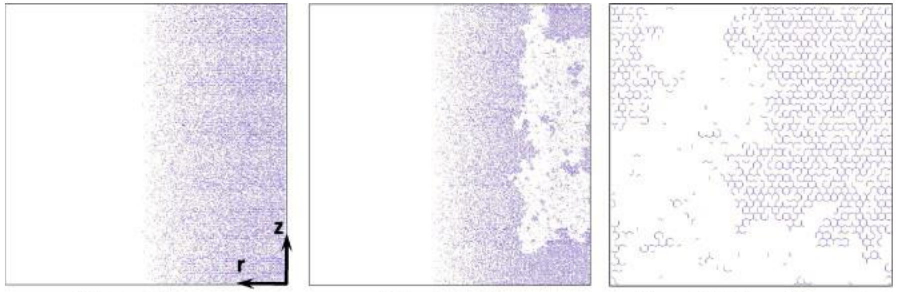


Fig. 1. Fraction of gas atoms on grain boundary, G_s/G_B, as a function of exposure for downward fuel cascading temperature history. γ is the bubble surface tension, 2θ is the angle where two free surfaces meet at a grain boundary, r is average bubble radius, V_c is the fractional coverage of the grain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

Forsberg-Massih model

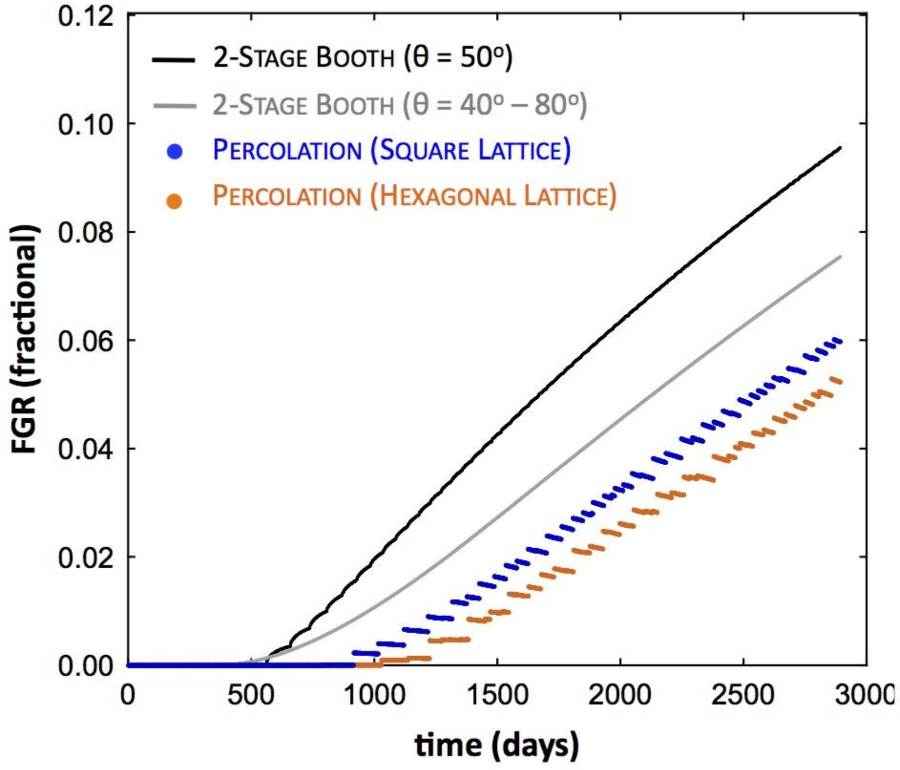
- 2-stage F-M model over-predicts gas release because it neglects grain boundary bubble percolation (Stage 3)





(a) t = 926 days (b) t = 1157 days (c) t = 1157 days (close-up)

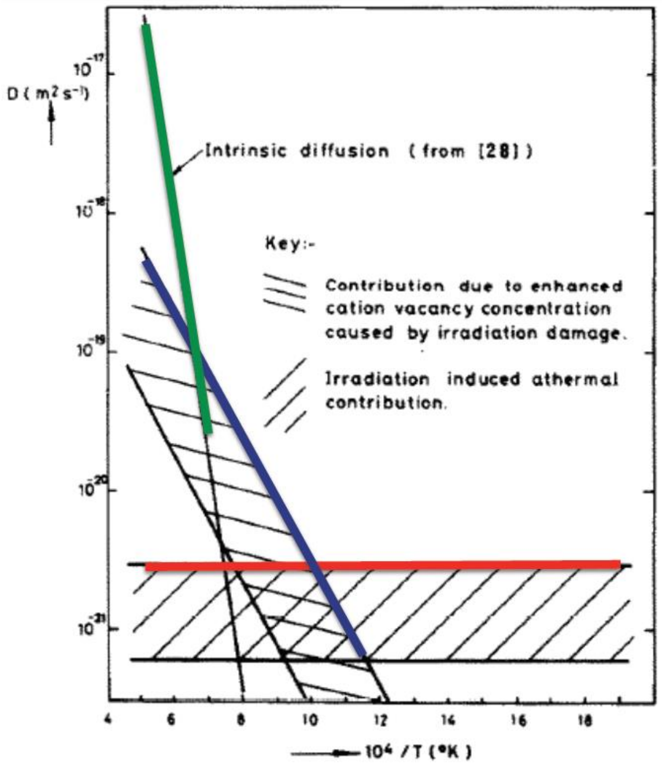
(d) t = 1208 days (e) t = 1250 days (f) t = 1308 days



Millett, Tonks, and Biner. JNM, 424.1 (2012): 176-182.

Gas diffusion

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

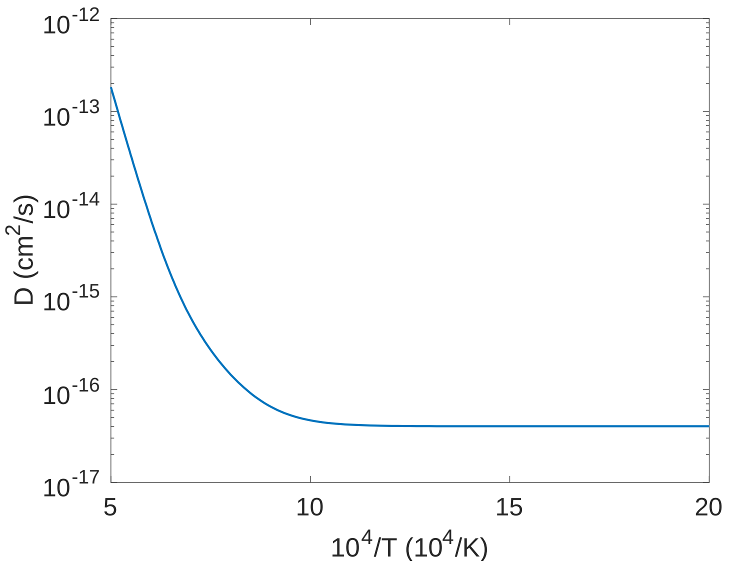


$$D = D_1 + D_2 + D_3 \text{ cm}^2/\text{s}$$

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

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

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



Gas diffusion

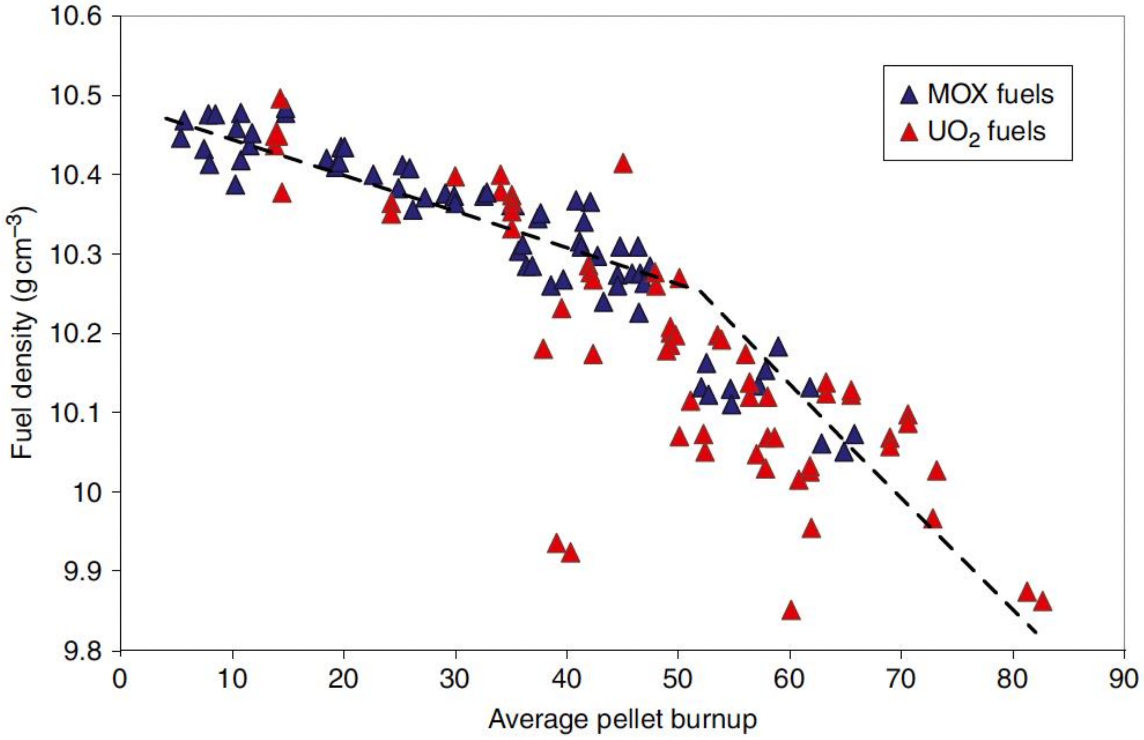
- The effective fission gas diffusivity is slower due to trapping by intragranular bubbles
- As the gas atoms diffuse towards the grain boundary, some are trapped by the small intragranular bubbles
- Some are later knocked out by energized particles (called resolution)
- The effective diffusion constant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

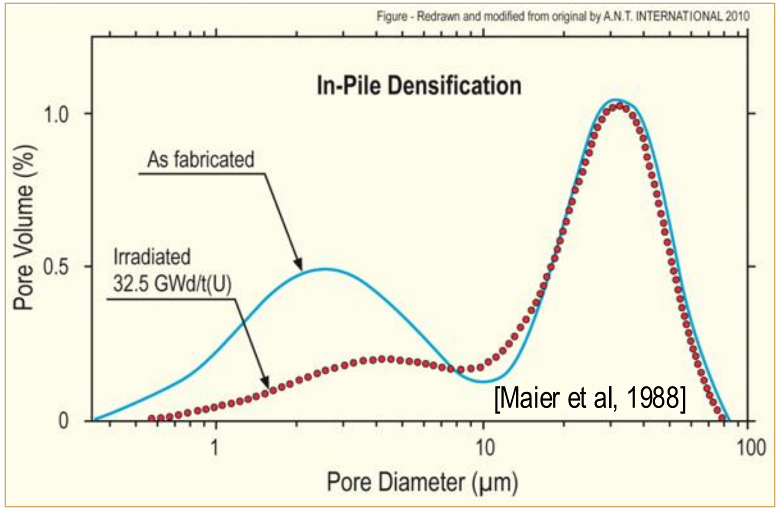
- Thermal expansion:
  - increase in volume, decrease in density, caused by increasing temperature
- Densification:
  - Decrease in volume, increase in density, caused by shrinking of porosity left after sintering
- Swelling:
  - Increase in volume, decrease in density, caused by fission products
- Irradiation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

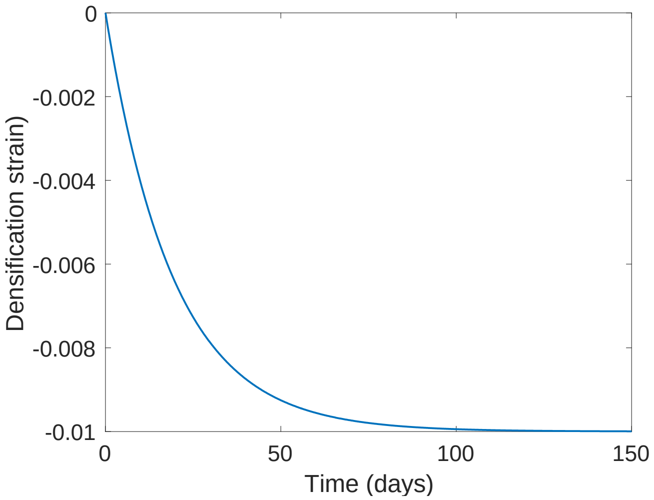


Densification

- 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(° C) − 25) for T < 750° C and CD = 1 for T ≥ 750 ° C

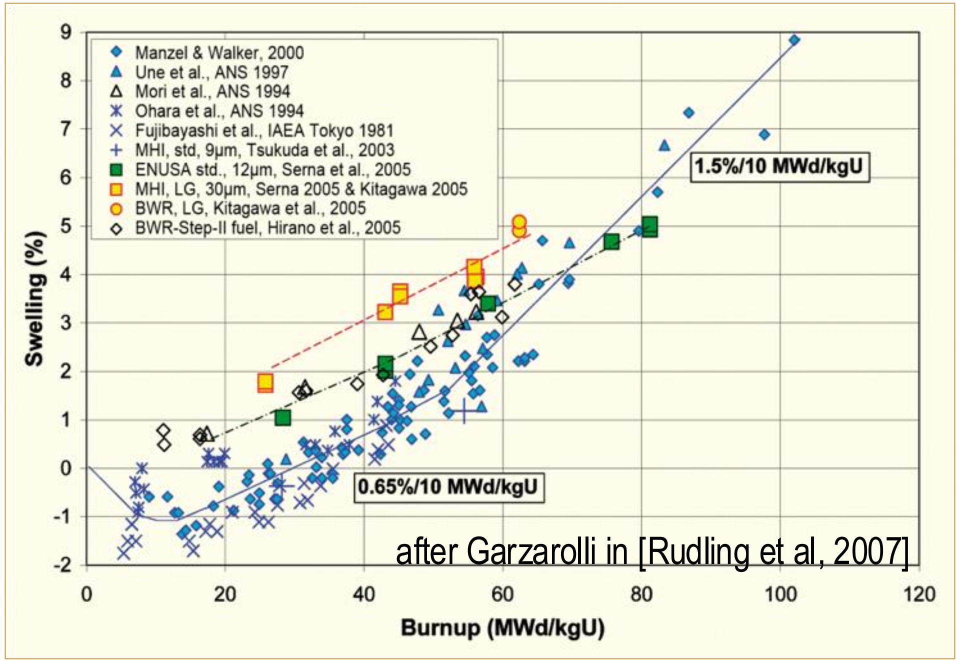
$$\epsilon_D = \Delta\rho_0 \left(e^{\frac{\beta \ln 0.01}{C_D \beta_D}} - 1 \right)$$





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

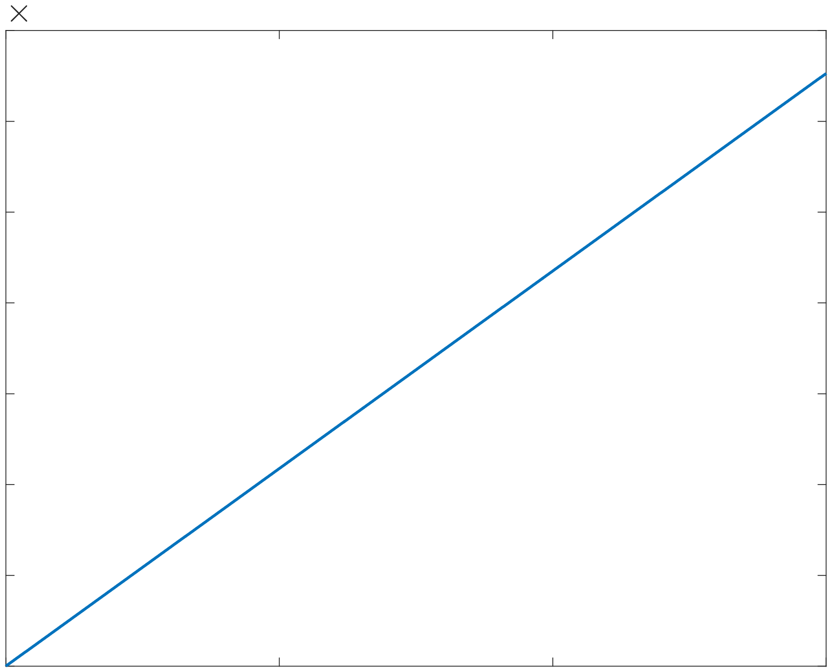


after Garzarolli in [Rudling et al, 2007]

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

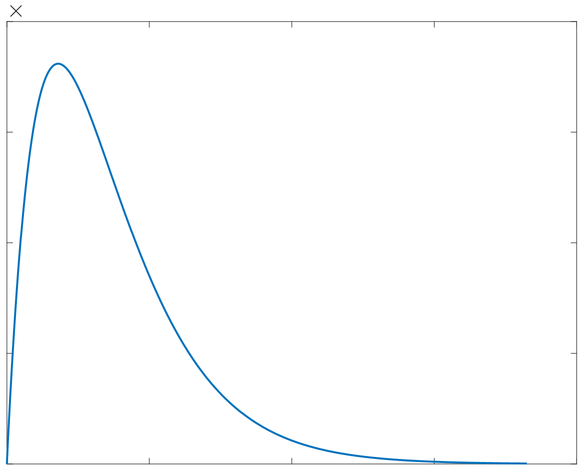
$$\epsilon_{sfp} = 5.577 \times 10^{-2} \rho\beta$$

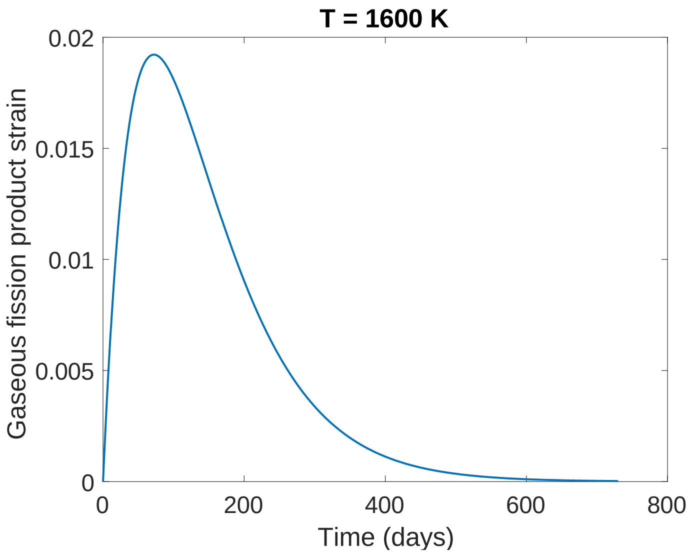


Gaseous fission product swelling

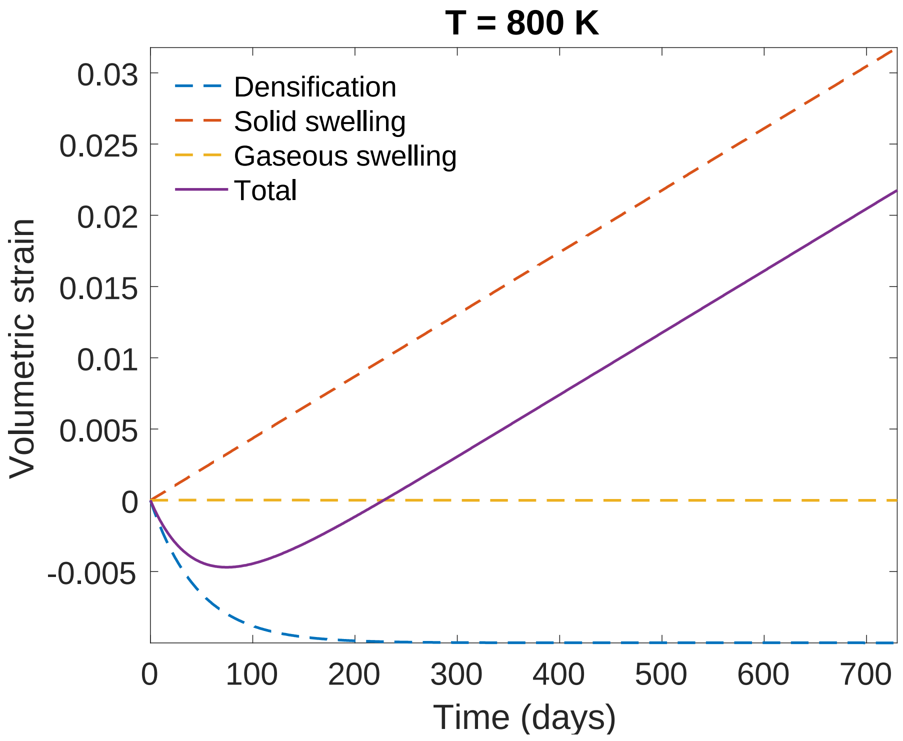
- Gaseous swelling varies strongly with temperature, fission rate, and stress
- Empirical relationships dependent upon burnup and temperature
- T < 1000K
  - Fission gas atoms remain in fuel matrix or collect in small, isolated, intragranular pores (<1 nm)
  - Intragranular pore size limited by fission 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

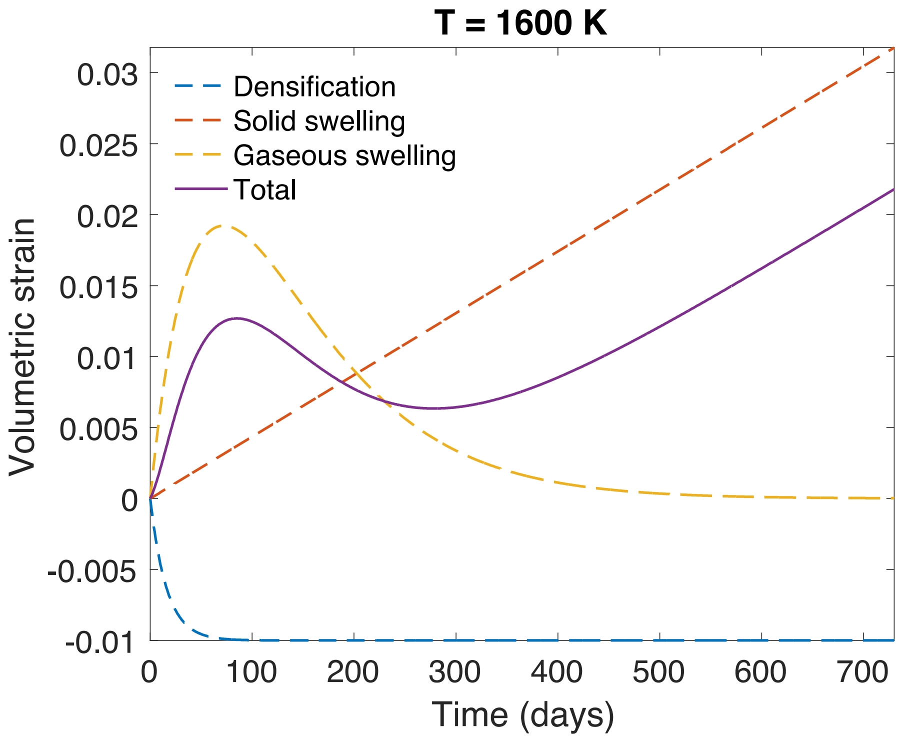
$$\epsilon_{gfp} = 1.96 \times 10^{-28} \rho\beta (2800 - T)^{11.73} e^{-0.0162(2800-T)} \, e^{-17.8\rho\beta}$$





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
  - $\varepsilon_{tot} = \varepsilon_{th} + \varepsilon_{D} + \varepsilon_{sfp} + \varepsilon_{gfp}$
- Example:
  - fission rate = 2.5e13 f/(cm^3 s)
  - T(fuel) = 1400 K
  - T_{ref} = 300 K
  - For densification: $\Delta\rho_0$ = 0.01 and β_D = 5 MWD/kgU
  - Total time: 2 weeks

Example