Nuclear Fuel Performance

NE-591-010

Spring 2021

Housekeeping

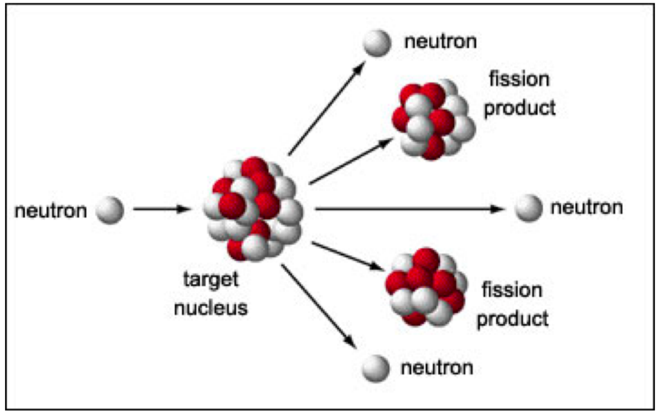
- Will be posting an optional assignment
  - If completed will count as 10% of your total grade, and other assignments will be adjusted uniformly
  - Details will be including within the post, but will be a written report, due April 22

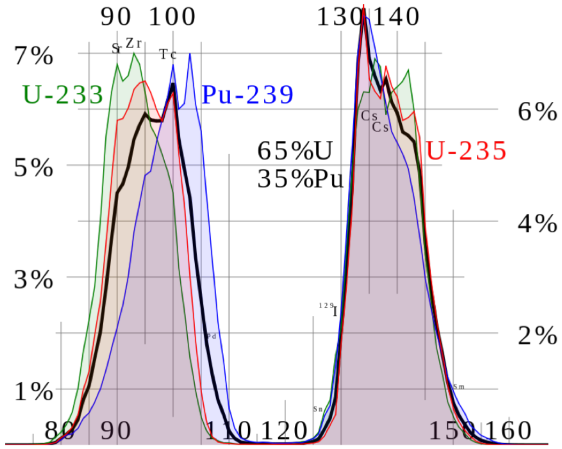
- Graded exams will be emailed out later today

FISSION PRODUCTS

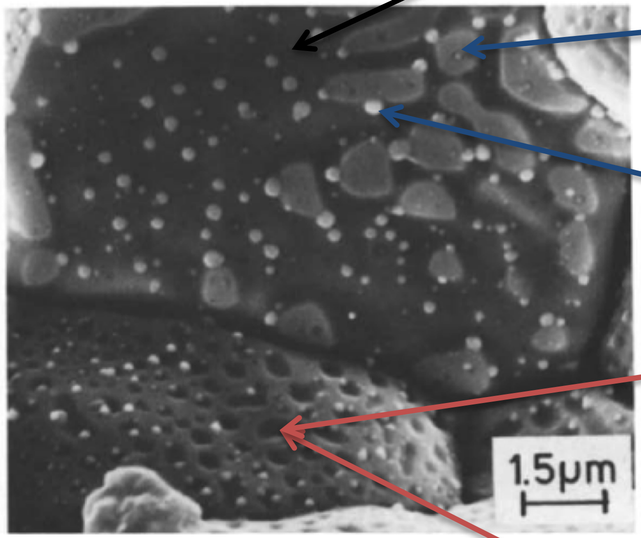
Fission Product Generation

- Fission releases around 200 MeV
  - The fission fragments have 169 MeV of kinetic energy
  - 2 to 3 neutrons with an average energy of 2 MeV
  - 7 MeV of prompt gamma ray photons
  - The remaining energy is released by beta decay
- Every fission product that is produced is now in the crystal lattice of the fuel, changing the microstructure





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



Soluble oxides (Y, La and the rare earths)
- Dissolved in the cation sublattice

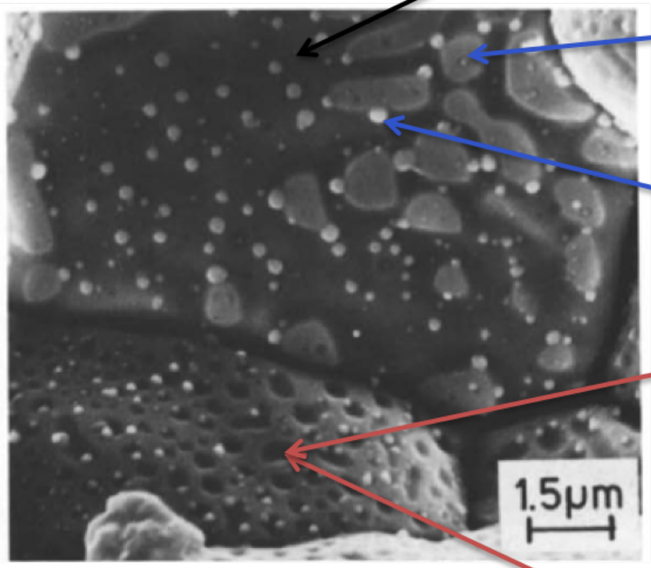
Insoluble oxides (Zr, Ba and Sr)
- Form insoluble oxides in the fluorite lattice

Metals (Mo, Ru, Pd, and Tc)
- Form metallic precipitates

Volatiles (Br, Rb, Te, I and Cs)
- Exist as gases at high temperatures of the pellet interior
- Also exist as solids at the cooler pellet exterior

Noble gases (Xe, Kr)
- Essentially insoluble in the fuel matrix
- Form either intragranular (within grain) voids or bubbles or intergranular (grain boundary) bubbles

All of the fission products impact the behavior of the fuel



Soluble oxides (Y, La and the rare earths)
- Cause swelling, decrease thermal conductivity

Insoluble oxides (Zr, Ba and Sr)
- Can cause swelling

Metals (Mo, Ru, Pd, and Tc)
- Slightly raise thermal conductivity,

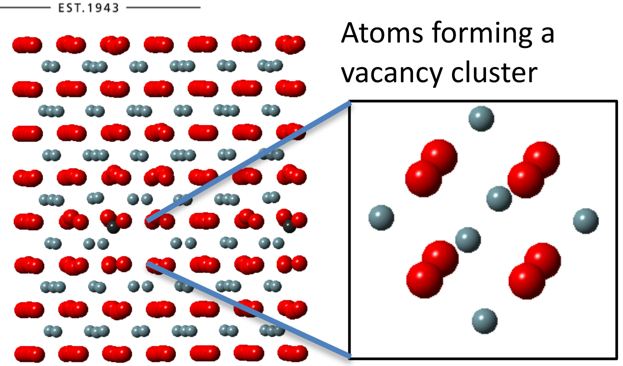
Volatiles (Br, Rb, Te, I and Cs)
- Cause swelling, decrease thermal conductivity
- Escape from fuel, corrode the cladding

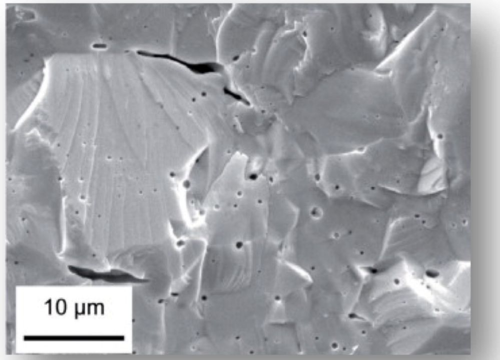
Noble gases (Xe, Kr)
- Cause swelling
- Decrease thermal conductivity
- After release, raise gap pressure and lower thermal conductivity

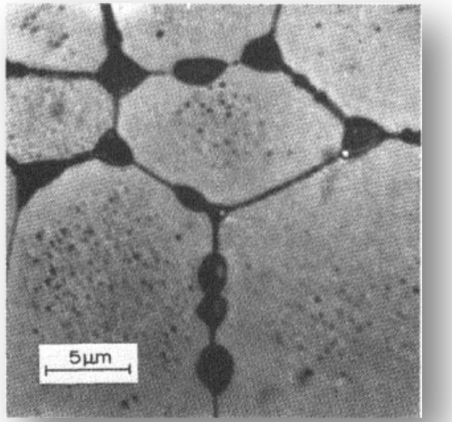
Fission Gas Release

- Fission gases (Xe, Kr) are released in a process composed of three stages in UO2
- Stage 1: Gas atoms are produced throughout the fuel due to fission and diffuse towards grain boundaries
- Small intragranular bubbles form within the grains, but never get larger than a few nm radius due to resolution from energized particles
- Gas atoms that don't get trapped within the intragranular bubbles migrate to grain boundaries

Los Alamos NATIONAL LABORATORY EST. 1943

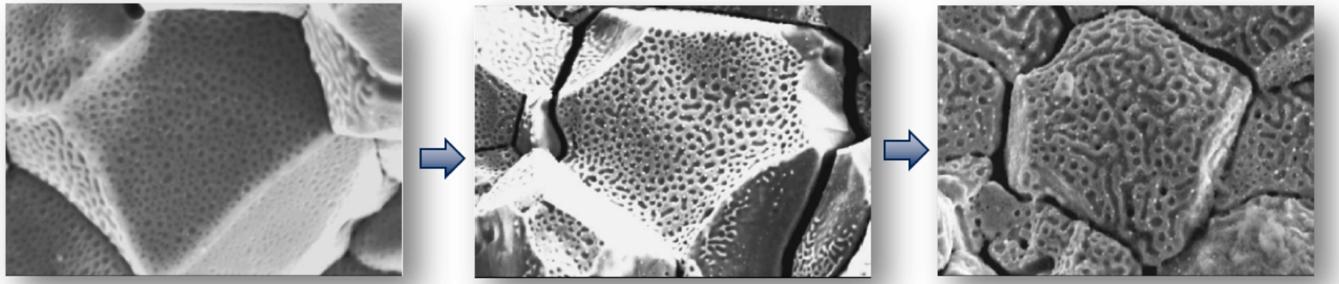






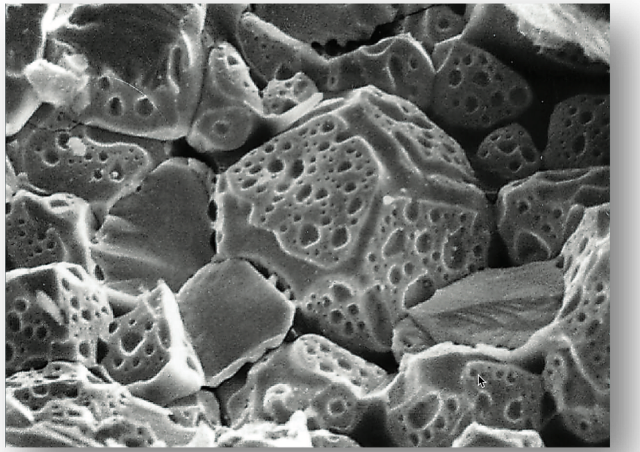
Fission Gas Release

- Stage 2: Gas bubbles nucleate on grain boundaries, growing and interconnecting



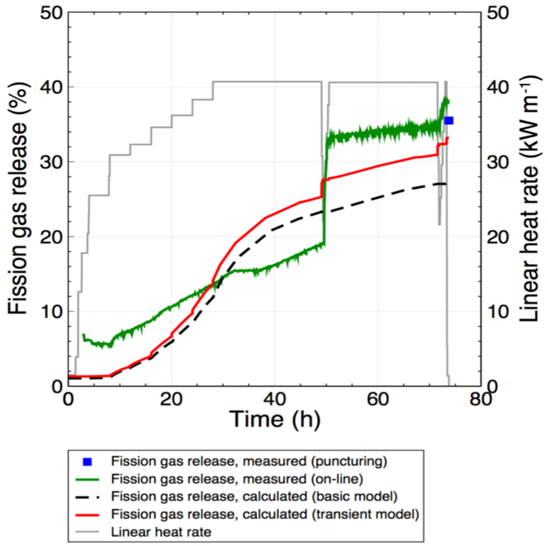
Nucleation Growth Percolation

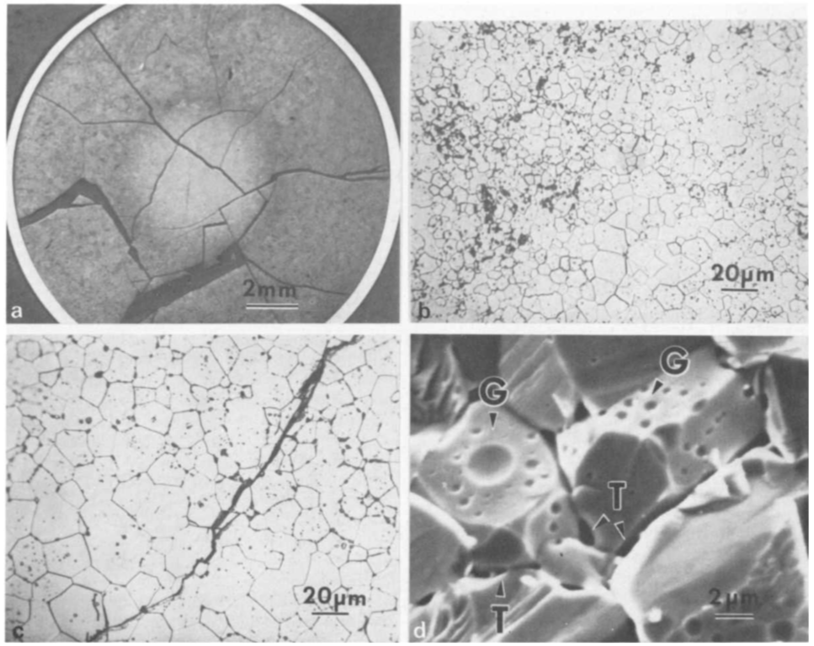
- Stage 3: Gas travels through interconnected bubbles to a free surface



Fission Gas Release

- Fission gas release also occurs due to mechanisms that don’t depend on diffusion
- Release can occur to particle recoil and knockout at low temperature
- It can occur due to fracture during rapid transients





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

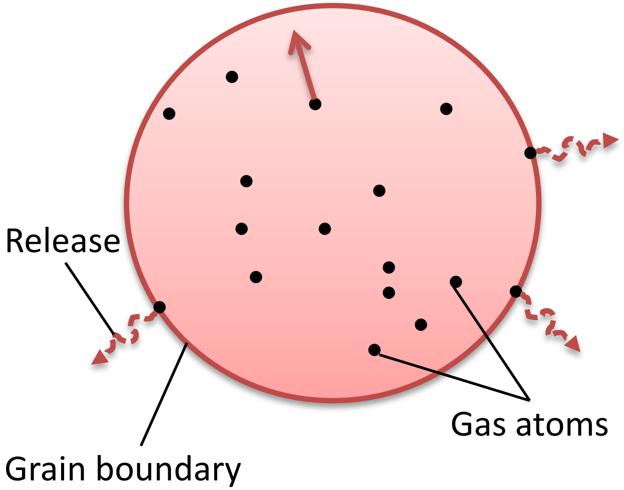
- Fission gas release models attempt to predict the rate at which gas is released from the fuel
- To model fission gas release, ideally we must model all three stages of gas release
  - Diffusion of gas atoms to grain boundaries
  - Growth and interconnection of grain boundary bubbles
  - Transport of 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)

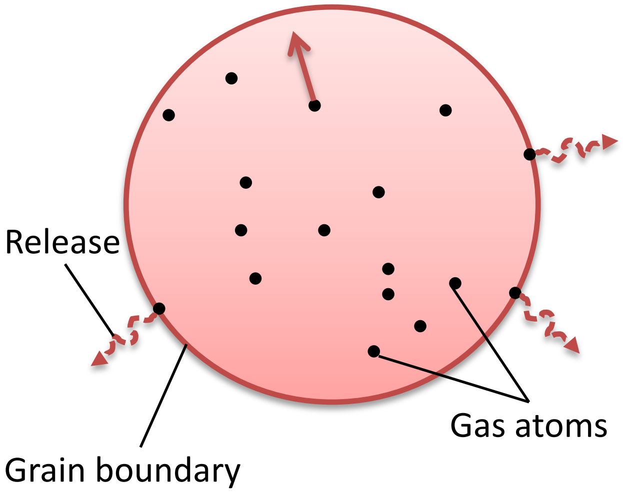
Modeling post-irradiation annealing

- 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

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



<u>ICs and BCs</u>

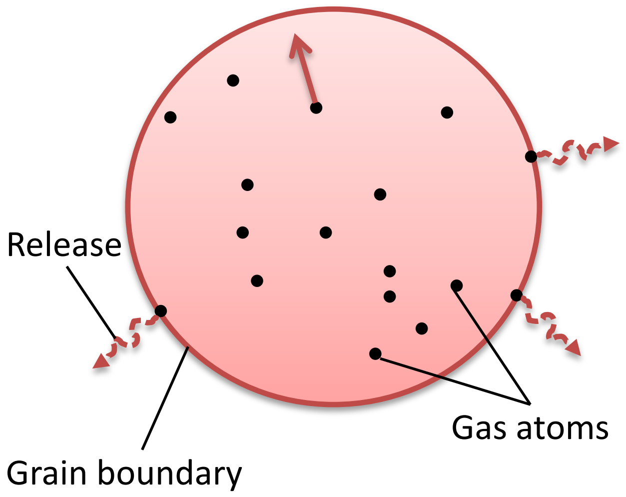
$c_g(r, 0) = c_g^0$

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

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

Solving the Booth Model

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



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

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

Booth Example

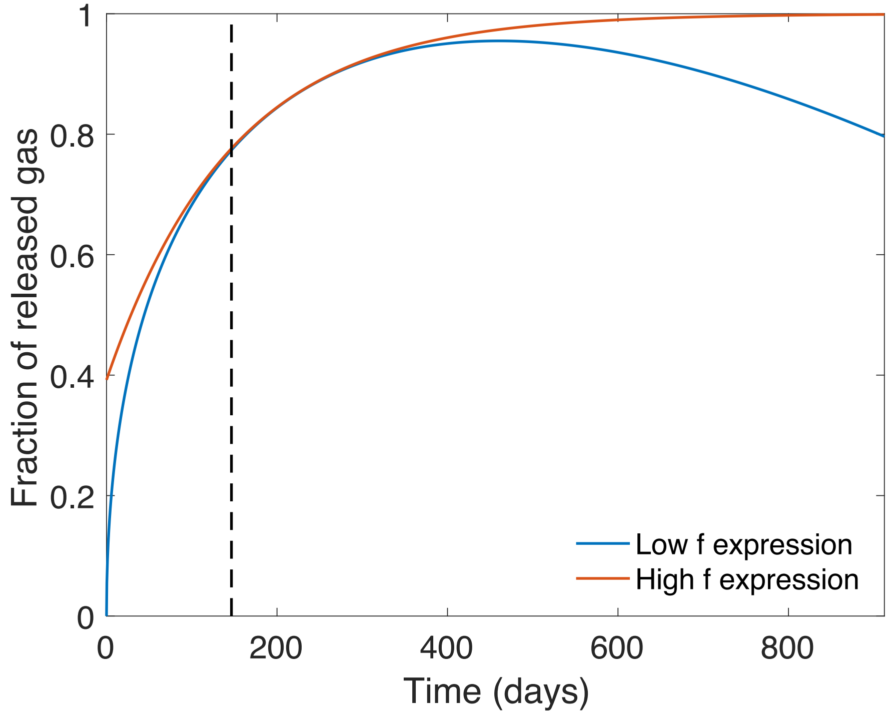
- For a diffusion coefficient for Xe of D = 8e-15 cm^2/s, what fraction of the fission gas trapped in an post-irradiation annealed fuel pellet has escaped after one hour? It has an average grain size of 10 microns
  - D = 8e-15 cm^2/s
  - a = 10e-4 cm
  - t = 3600 s
- Which f? $\tau = D \times t/a^2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

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

  - 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

- 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

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

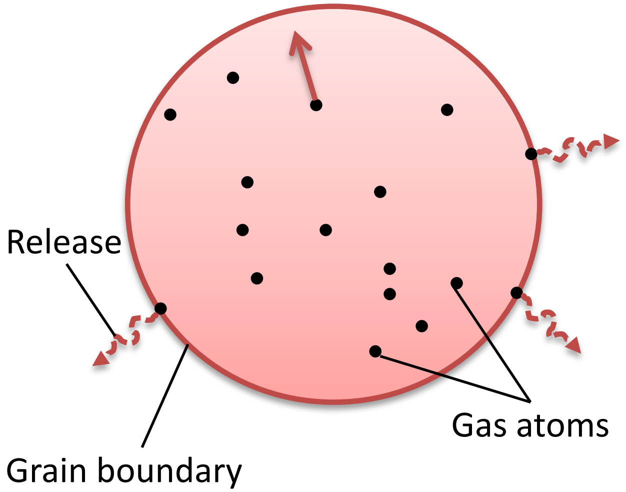
- After solving with with a Laplace transform

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

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

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

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



<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), so we can use:

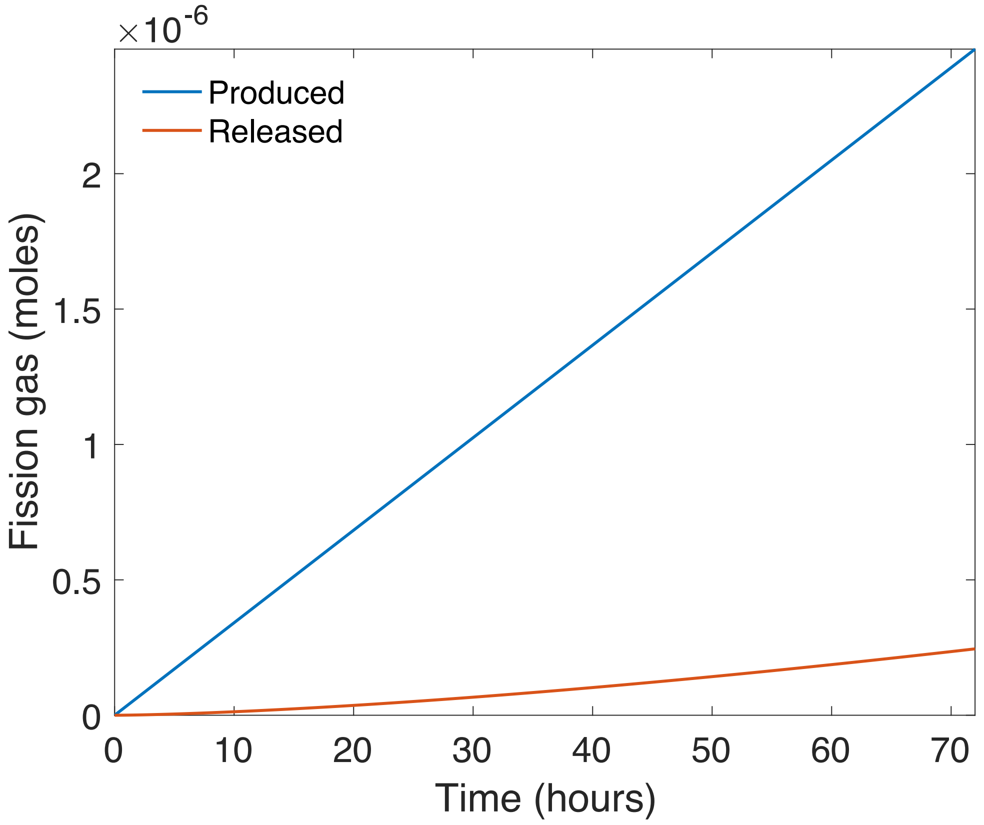
$$f = 4\sqrt{\frac{Dt}{\pi a^2}} - \frac{3}{2}\frac{Dt}{a^2}$$

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

Gas release

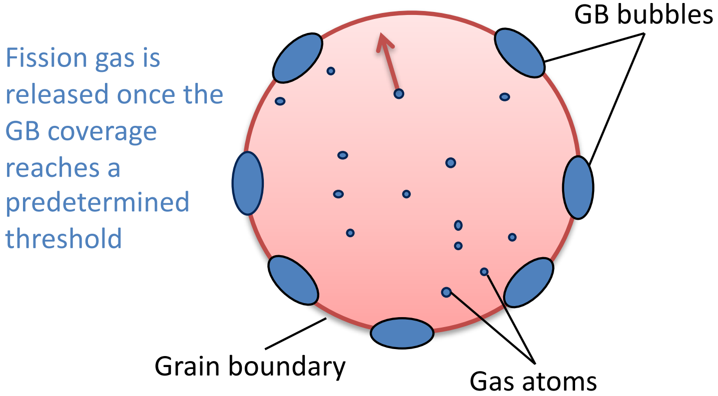
- We have calculated the fraction, but how much gas was actually released?
- Consider 3% enriched UO_2 with a thermal fission cross section of U-235, σ_{f235} = 570 barns or 570 $\times$ 10^{-24} cm^2 and a density of UO2, ρ_U = 10.97 g/cm^3.
- What would be the number of fission gas atoms released after one hour with a neutron flux of 3×10^{13} n/(cm^2 s) from a pellet of radius 0.5 cm and height 1.2 cm?
  - Molar mass of 3% enriched UO2: 235*0.03 + 238*0.97 + 2*16 = 269.9 g/mol
  - Atom density of U-235 = 10.97 x 1/269.9 x 6.022E23 x 1/1 x 0.03 = 7.34E20 atoms/cm^3
  - First, we have to calculate the fission rate: Fdot = 7.34E20*570E-24*3e13 = 1.26E13 fissions/(cm^3 s)
  - Next we calculate the total amount of fission gas produced
    - N_{FG} = y Fdot t = 0.3017*1.26E13*3600 = 1.36E16 fission gas atoms/cm^3
    - Vol = π r^2 h = pi*0.5^2*1.2 = 0.94 cm^3
    - NFG*Vol = 1.36E16*0.94 = 1.28e16 fission gas atoms
  - Finally we calculate the total amount of gas released using the fraction
    - f = 0.0121
    - Gas released = 0.0121*1.28e16 atoms produced = 1.55e14 atoms released

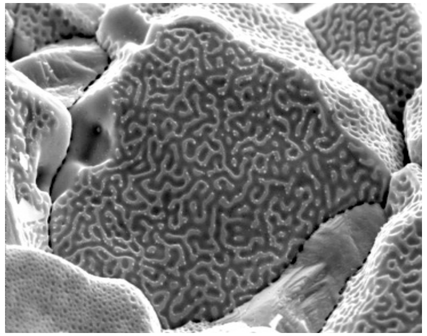
As time progresses, both the fraction released and the produced gas increase



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)





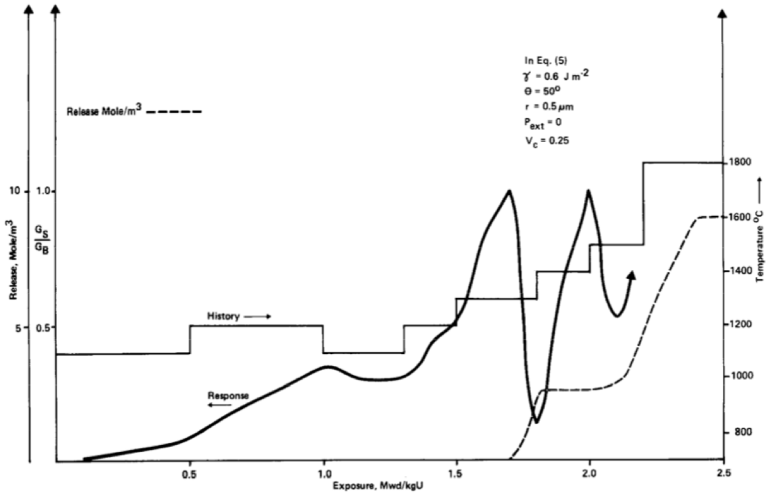
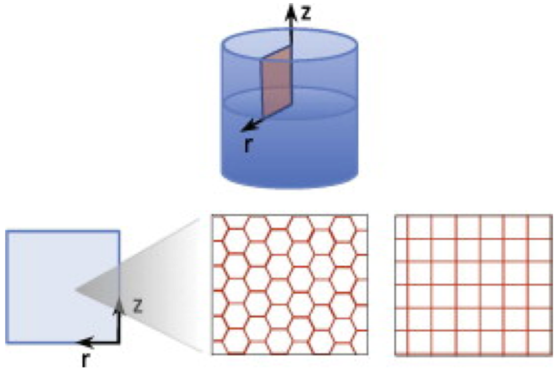
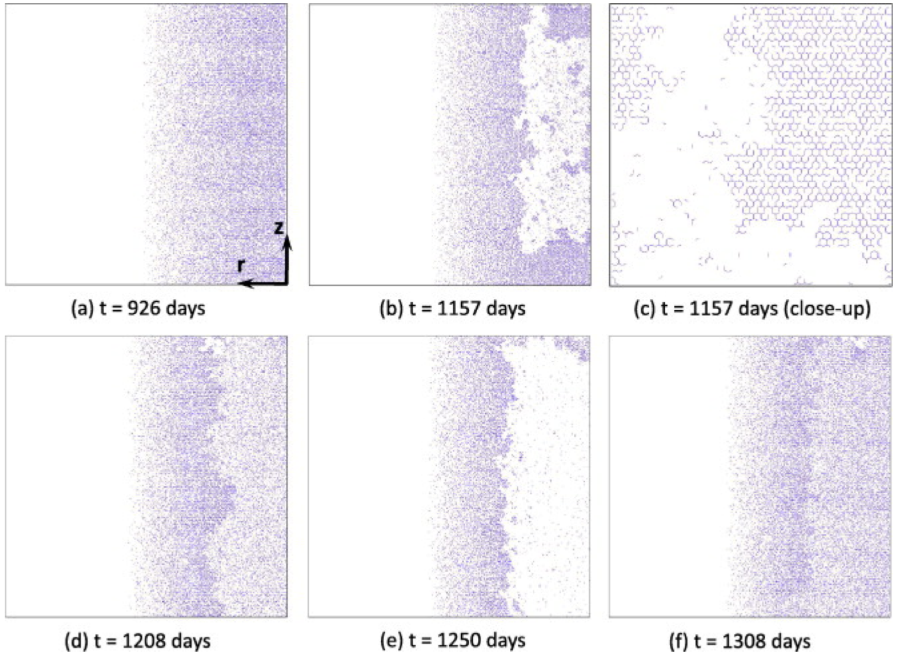


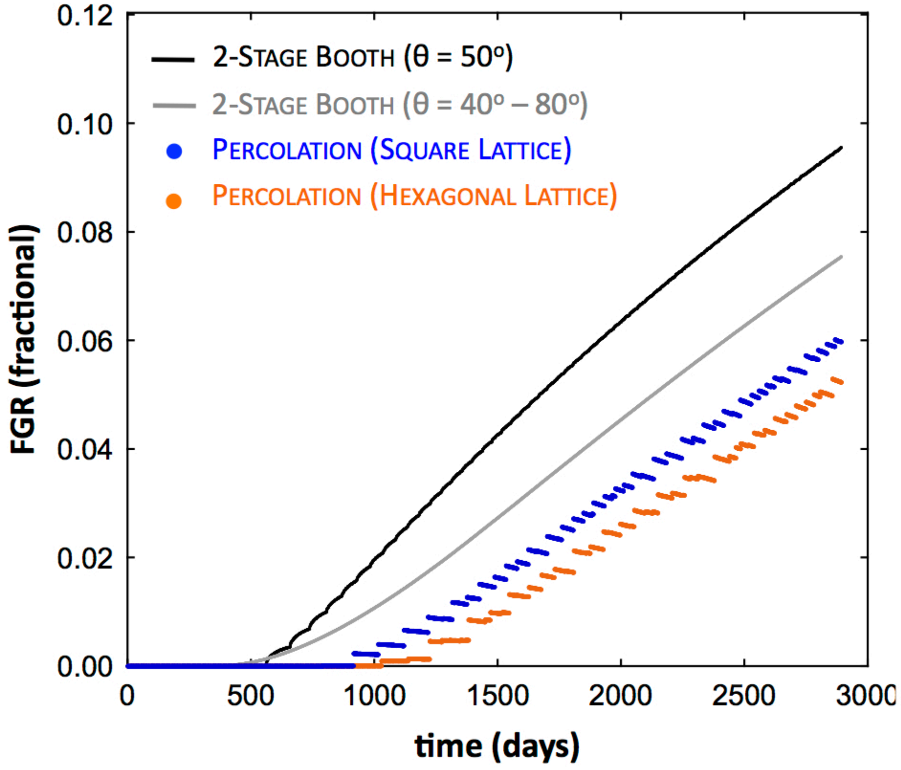
Fig. 1. Fraction of gas atoms on grain boundary, G_s/G_B, as a function of exposure for downward fuel cascading temperature history. γ is the bubble surface tension, 2θ is the angle where two free surfaces meet at a grain boundary, r is average bubble radius, V_c is the fractional coverage of the grain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



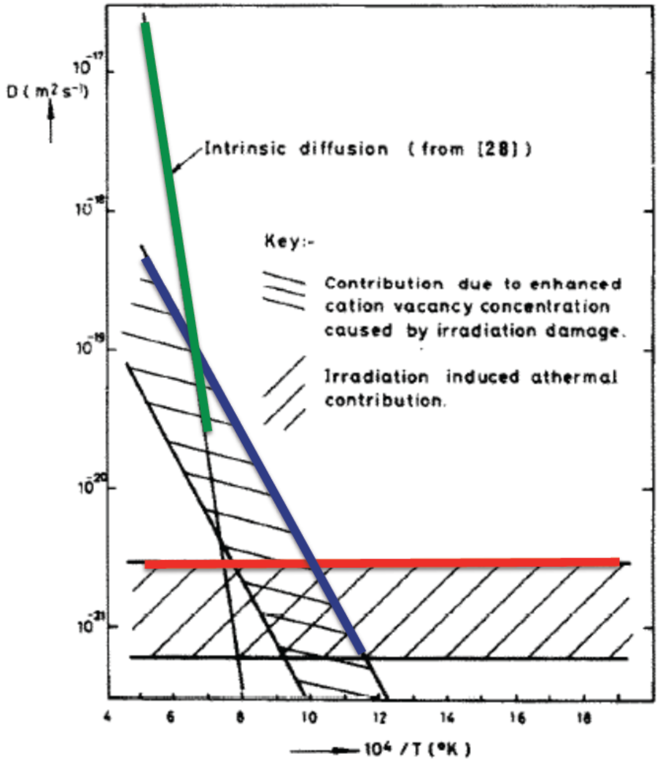




Millett, Tonks, and Biner. JNM, 424.1 (2012): 176-182.

Gas diffusion

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

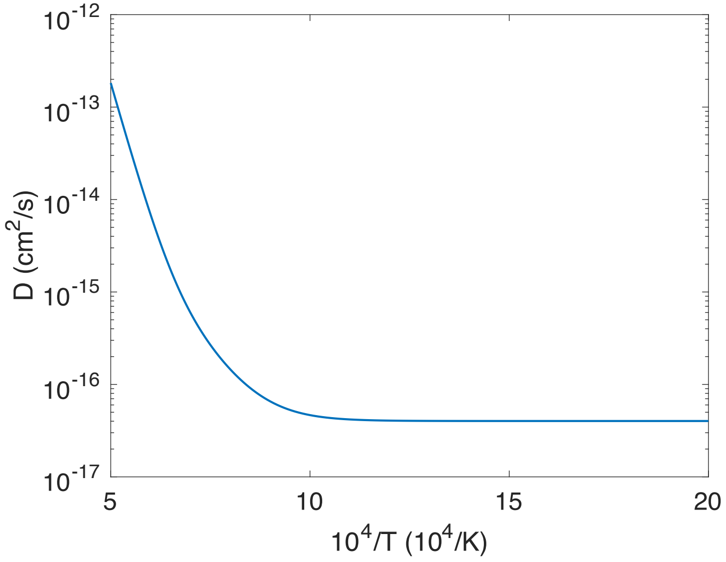


$$D = D_1 + D_2 + D_3 \text{ cm}^2/\text{s}$$

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

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

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



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

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

Summary

- Fission products change the fuel behavior
- 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 is measured using post-irradiation annealing and/or in pile experiments
- 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
- Fission gas diffusivity behavior changes with temperature and fission rate