NucE 497: Reactor Fuel Performance

Lecture 25: Fission gas release

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Content taken from Ch 15 of Olander's book

Today we will discuss modeling of fission gas release in UO_2

- Module 1: Fuel basics
- Module 2: Heat transport
- Module 3: Mechanical behavior
- Module 4: Materials issues in the fuel
  - Property evolution and Intro to materials science
  - Chemistry
  - Grain growth
  - **Fission products and fission gas**
  - Densification, swelling, and creep
  - HBS
  - Fracture
  - Thermal conductivity
- Module 5: Materials issues in the cladding
- Module 6: Accidents, used fuel, and fuel cycle

Here is some review from last time

- Which is not a fission product that forms within the fuel?
  a) Metal precipitates
  b) Ceramic precipitates
  c) Hydride precipitates
  d) Fission gas bubbles
- What occurs during the third stage of fission gas release?
  a) Fission gas bubbles diffuse to grain boundaries
  b) Fission gas travels through interconnected grain edges to free surfaces
  c) Fission gas bubbles burst open during transients
  d) Fission gas bubbles form and interconnect on grain faces

Two kinds of experiments are used to investigate fission gas release

- 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 models attempt to predict the rate at which gas is released from the fuel

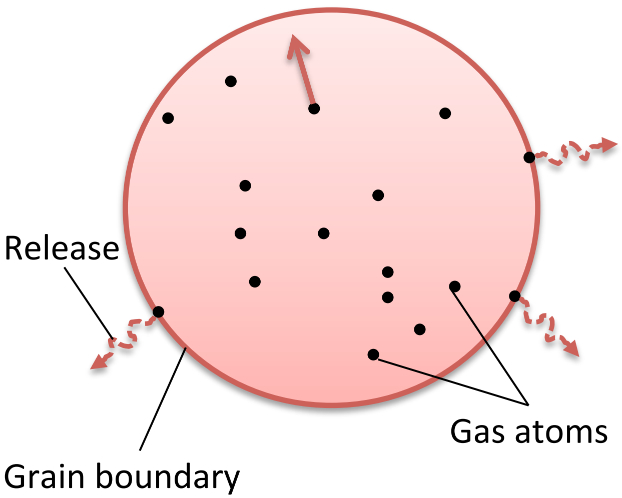
- To model fission gas release, ideally we must model all three stages of gas release
    1. Diffusion of gas atoms to grain boundaries
    2. Growth and interconnection of grain boundary bubbles
    3. Transport of gas atoms through interconnected bubbles to free surfaces
- The earliest models only considered Stage 1
- Most models now consider stage 1 and 2
- There are no models that consider all three stages

The Booth model is the earliest model of fission gas release and only considers stage 1

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

- 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

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



<u>ICs and BCs</u>

$c_g(r, 0) = 0$

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

$c_{g,r}(0, a) = 0$ (release)

First, we will model post-irradiation annealing

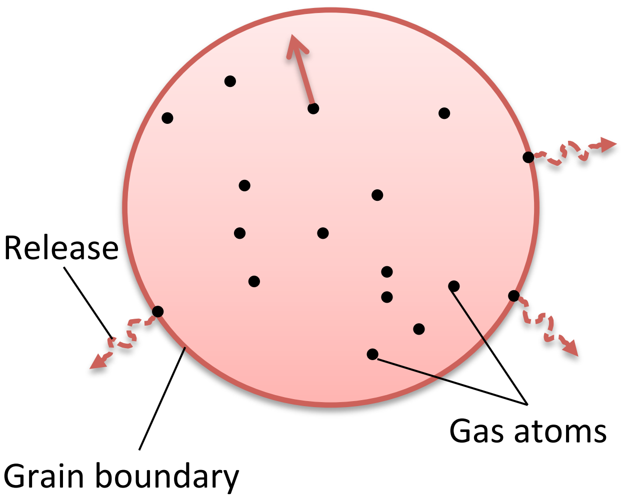
- The initial gas concentration is c_g^0
- No gas is produced

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

- 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

$$J_a = -D\left(\frac{\partial c_g}{\partial r}\right)_a$$

$$f = \frac{4\pi a^2 \int_0^t J_a dt}{4/3\pi a^3 c_g^0} = \frac{3}{a c_g^0}\int_0^t J_a dt$$



ICs and BCs

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

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

$c_{g,r}(0, a) = 0$ (release)

This equation is solved using a Laplace transform after nondimensionalization

- We nondimensionalize radius, time, and concentration
  - $\eta = r/a$
  - $\tau = D\,t/a^2$
  - $u = \eta\, c_g/c_g^0$
- The PDE becomes $\frac{\partial u}{\partial \tau} = \frac{\partial^2 u}{\partial \eta^2}$
- With IC and BC
  - $u(\eta, 0) = \eta$
  - $u(0, \tau) = 0$
  - $u(1, \tau) = 0$
- The final solution for the fraction becomes (see Olander's book for derivation)

$$f = 6\sqrt{\frac{\tau}{\pi}} - 3\tau$$

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

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

Now let's work a problem

- 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

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

  - f = 6*sqrt(8e-15*3600/(pi*(10e-4)^2)) – 3*8e-15*3600/(10e-4)^2 = 0.0181
- Now, let's try it without the last term

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

  - f = 6*sqrt(8e-15*3600/(pi*(10e-4)^2)) = 0.0182
  - The second term is not significant for small t

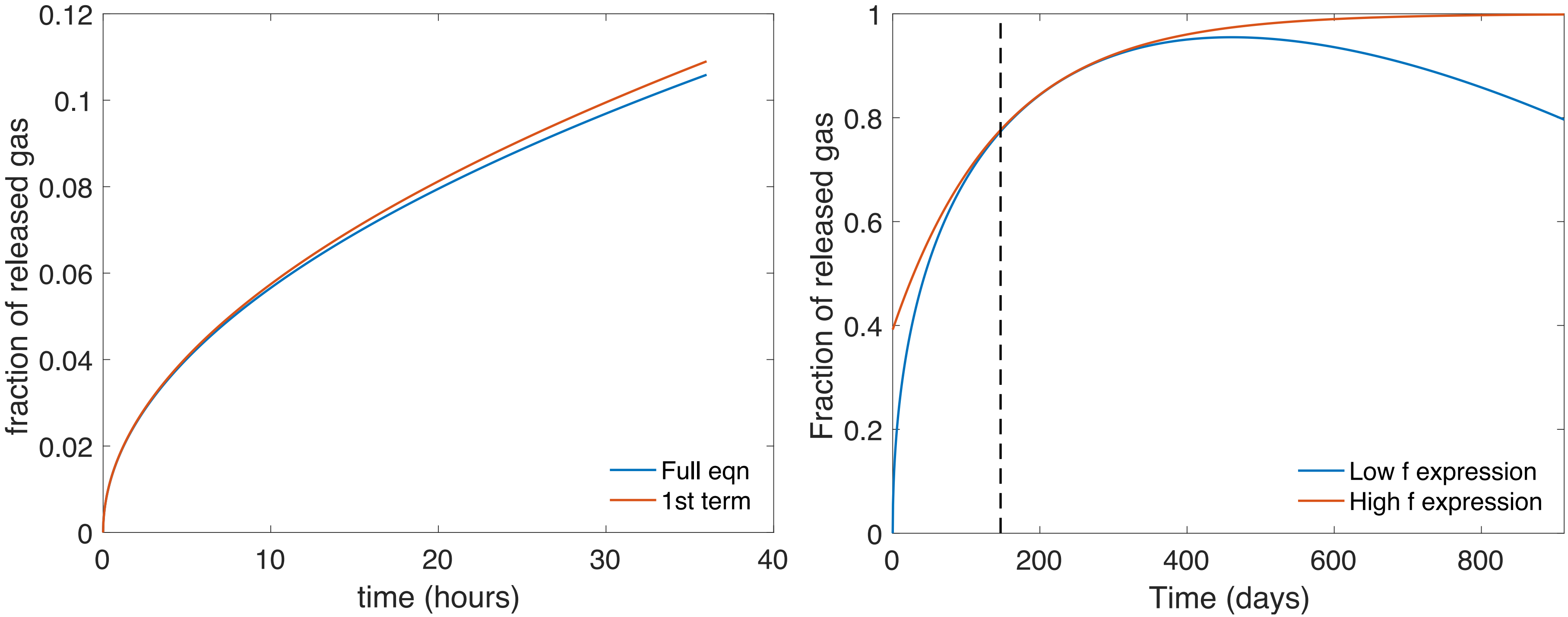
Now, here is a problem for you to try

- For a diffusion coefficient for Xe of D = 8e-15 cm^2/s and a 10 micron grain size, how long must you wait for 10% of the fission gas to escape? Only consider the first term of the equation.

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

- $f^2/36 = D\ t/(\pi\ a^2)$
- $t = \pi\ a^2\ f^2/(36*D)$
- t = pi*(10e-4)^2*(0.1)^2/(36*8e-15) = 1.09e5 s
- This is 30.3 hours

Leaving off the last term hurts the accuracy for longer times, and for really long times change expressions



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

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

Now, we will model 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

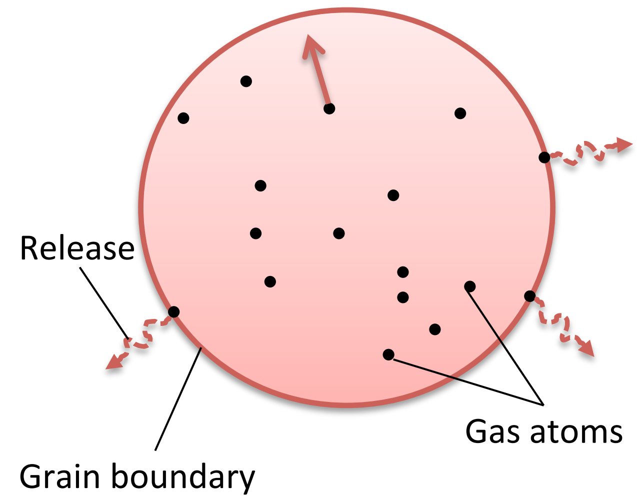
$$f = \frac{3}{ay\dot{F}t}\int_0^t J_a\,dt$$

- 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>

$c_g(r, 0) = 0$

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

$c_{g,r}(0, a) = 0$ (release)

Now let's solve an in-pile release problem

- 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

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

  - f = 4*sqrt(8e-15*3600/(pi*(10e-4)^2)) – 3/2*8e-15*3600/(10e-4)^2 = 0.0121
- Again, we can leave off the second term for short times

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

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

We have calculated the fraction, but how much gas was actually released?

- Consider 5% enriched UO_2 with a thermal fission cross section of U-235, σ_{f235} = 550 barns or 5.5 × 10^{-22} cm^2 and a density of U, δ_U = 9.65 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?
  - First, we have to calculate the fission rate $\dot{F} = qN_U\sigma_{f235}\phi_{th}$
    - $N_U = N_a\ \delta_U/M_U$ = 6.022e23*9.65/238 = 2.44e22 atoms of U/cm^3
    - Fdot = 0.05*2.44e22*5.5e-22*3e13 = 2.01e13 fissions/(cm^3 s)
  - Next we calculate the total amount of fission gas produced
    - N_{FG} = y Fdot t = 0.3017*2.01e13*3600 = 2.18e16 fission gas atoms/cm^3
    - Vol = π r^2 h = pi*0.5^2*1.2 = 0.94 cm^3
    - NFG*Vol = 2.18e16*0.94 = 2.06e16 fission gas atoms
  - Finally we calculate the total amount of gas released using the fraction
    - f = 0.0121
    - Gas released = 0.0121*2.06e16 atoms produced = 2.49e14 atoms released

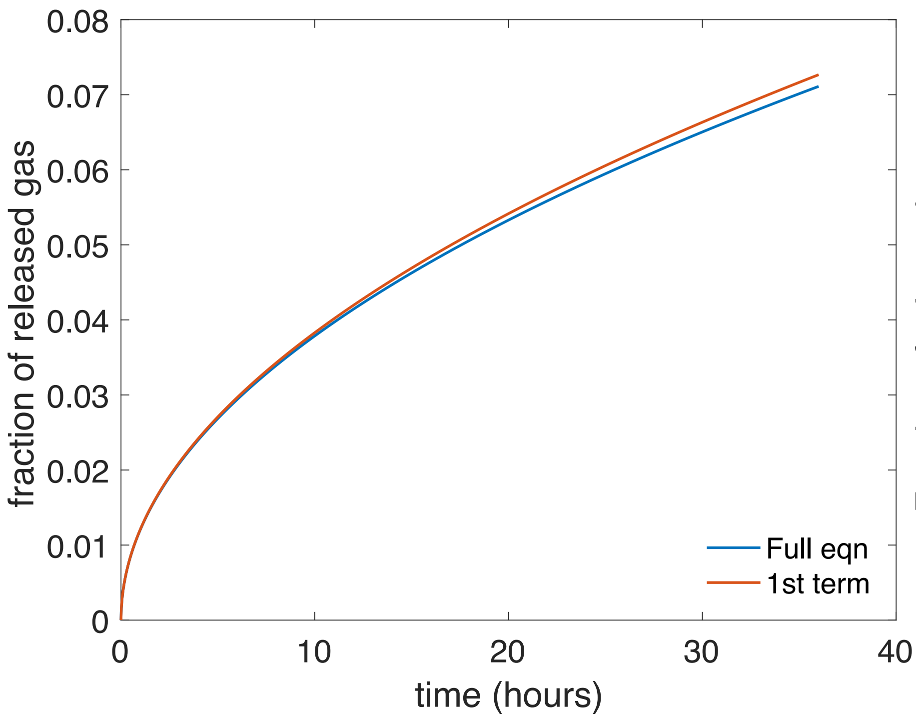
Now, you determine how much gas would be released if the fission rate was decreased by 50%

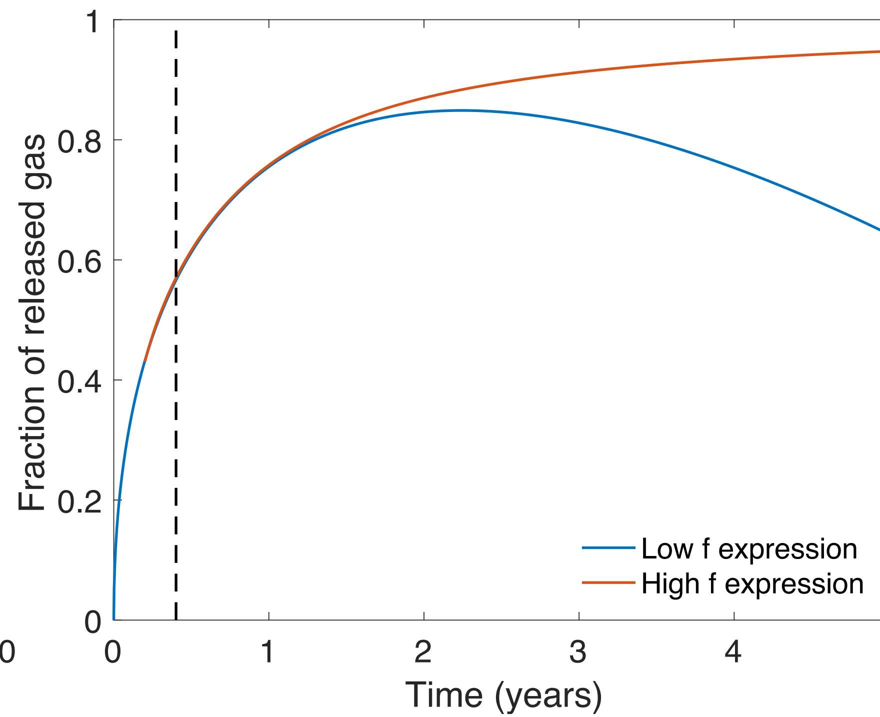
- With a fission rate of 3×10^{13} n/(cm^2 s), 2.49e14 atoms were released

$$\dot{F} = qN_U\sigma_{f235}\phi_{th} \qquad f = 4\sqrt{\frac{Dt}{\pi a^2}}$$

- Would the fraction of released gas change?
  - No, so f = 0.0121
- Would the total amount of gas produced change?
  - Yes, because Fdot would change
- Find the new amount of atoms that would be released
  - It would simply decrease by 50% like the fission rate
  - 0.5*2.49e14 = 1.245e14 atoms would be released

Again, leaving off the last term hurts the accuracy for longer times, and for really long times change expressions

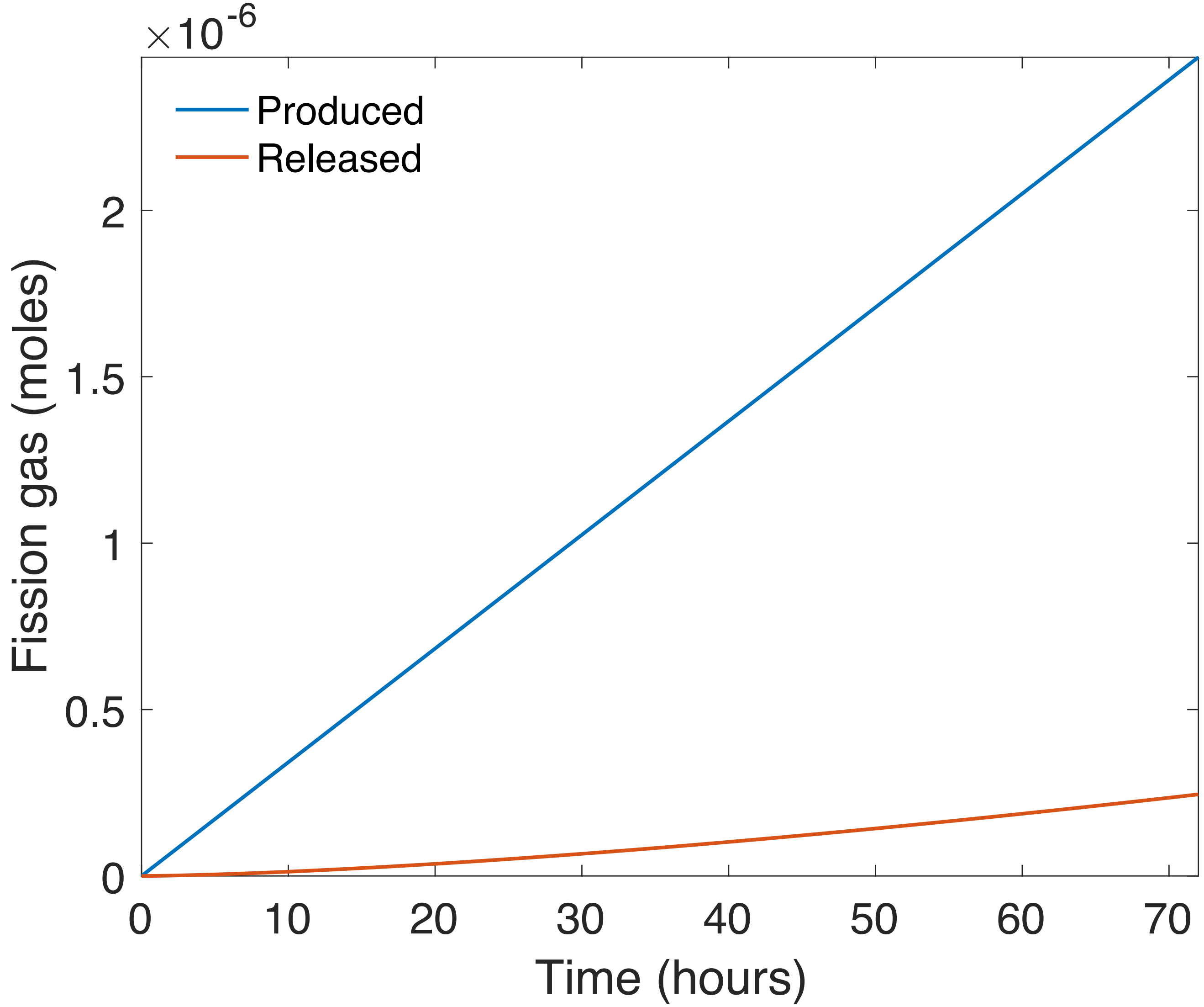




$$f = 4\sqrt{\frac{Dt}{\pi a^2}} - \frac{3}{2}\frac{Dt}{a^2} \qquad \tau < \pi^{-2}$$

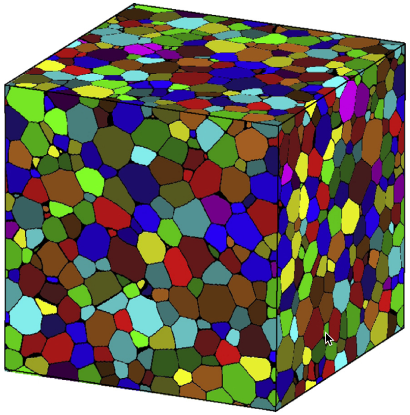
$$f = 1 - \frac{0.0662}{\frac{Dt}{a^2}}\left(1 - 0.93e^{-\pi^2 \frac{Dt}{a^2}}\right) \qquad \tau \geq \pi^{-2}$$

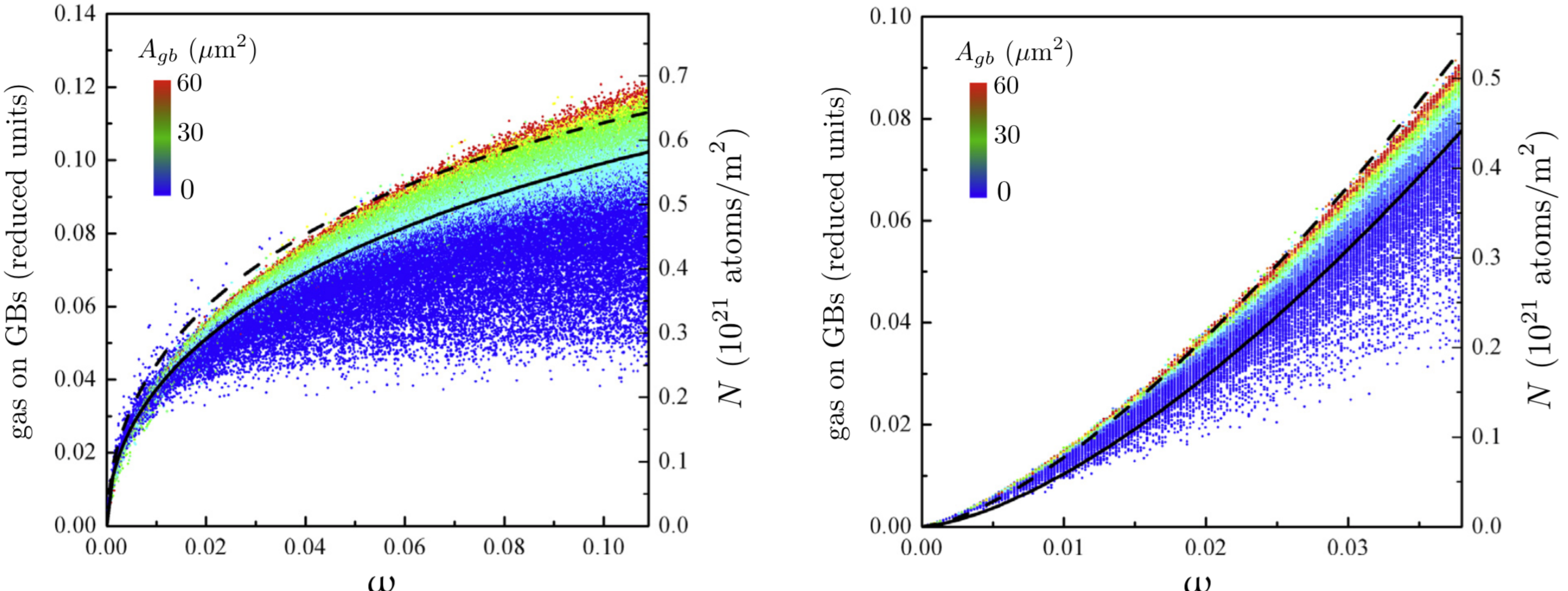
As time progresses, both the fraction and the produced gas increase



The Booth model assumes every grain is spherical and the same size, which impacts accuracy

- We conducted a diffusion simulation in a polycrystal and kept track of gas that arrived at the grain boundary

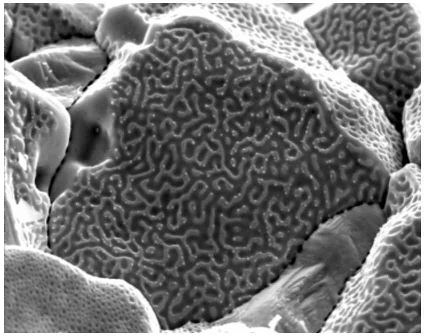




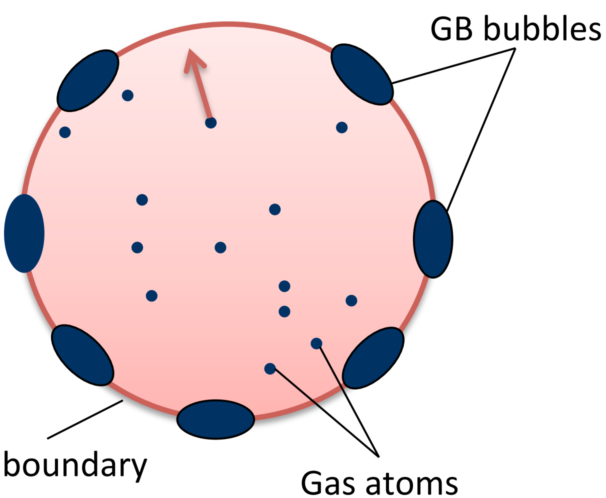
Millett, Zhang, Tonks, and Biner, JNM 440 (2013) 435–439

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



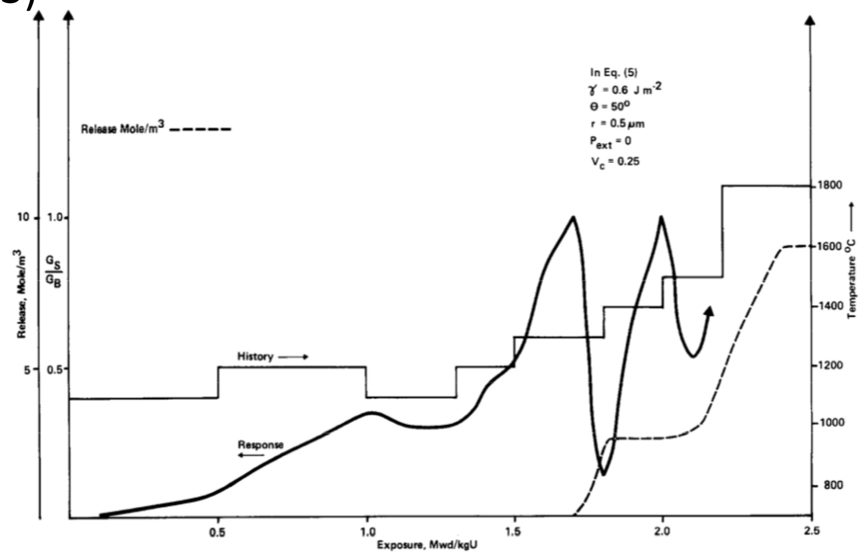
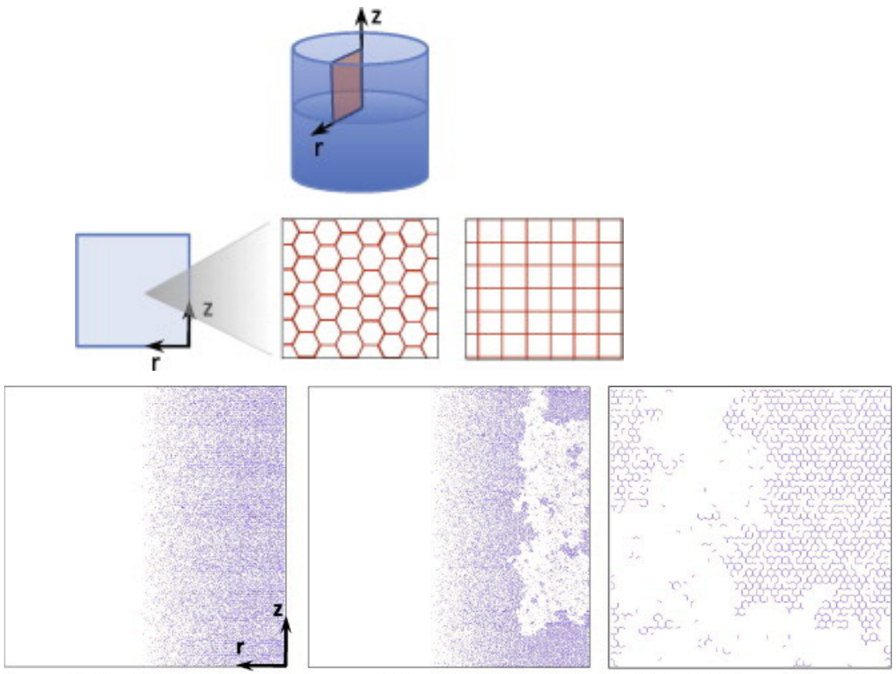


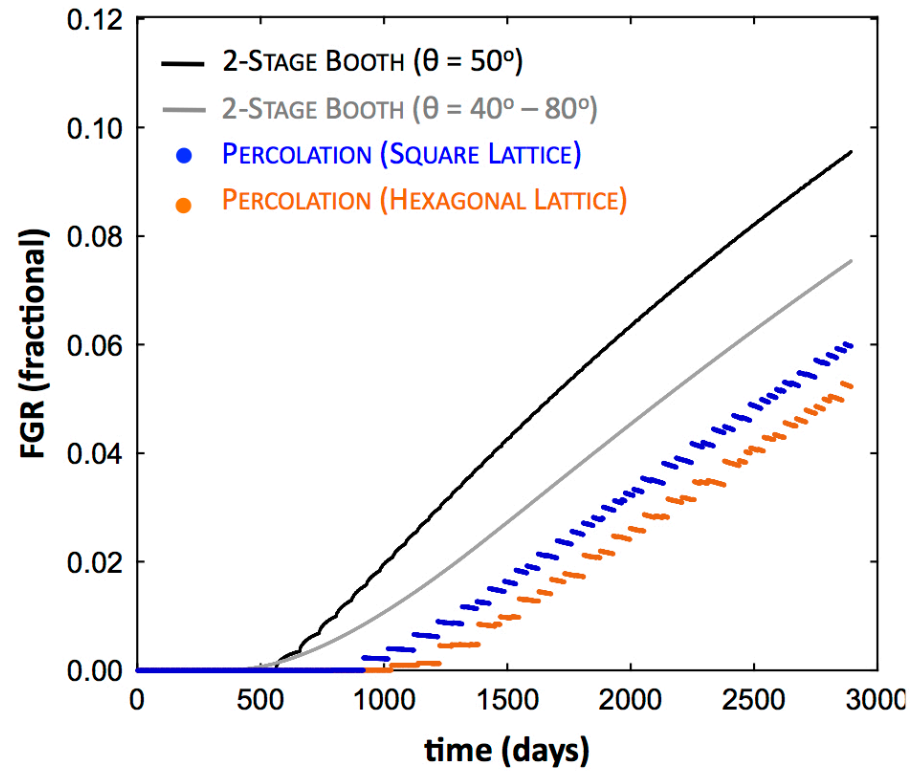
Fig. 1. Fraction of gas atoms on grain boundary, G_s/G_B, as a function of exposure for downward fuel cascading temperature history. γ is the bubble surface tension, 2θ is the angle where two free surfaces meet at a grain boundary, r is average bubble radius, V_c is the fractional coverage of the grain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

Forsberg & Massih, J. Nucl. Mater. 135 (1985) 140-148

The 2 stage Booth model over-predicts fission gas release, because it ignores stage 3.



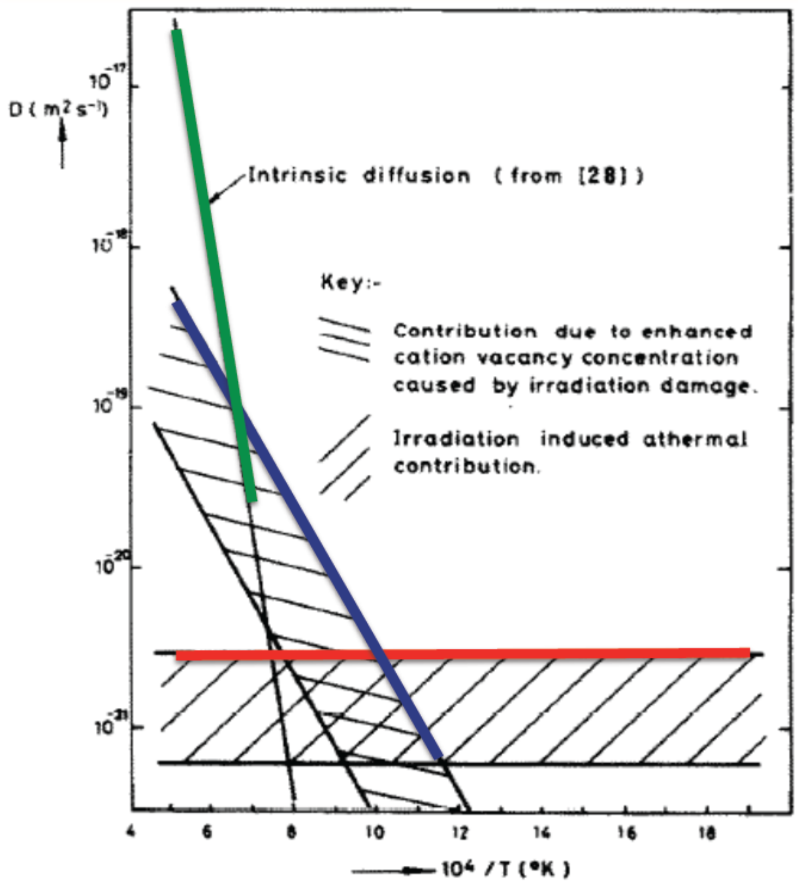
(a) t = 926 days
(b) t = 1157 days
(c) t = 1157 days (close-up)
(d) t = 1208 days
(e) t = 1250 days
(f) t = 1308 days



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

The diffusivity of the fission gas depends on temperature and on irradiation

- Experimental data shows three different regimes for the diffusivity

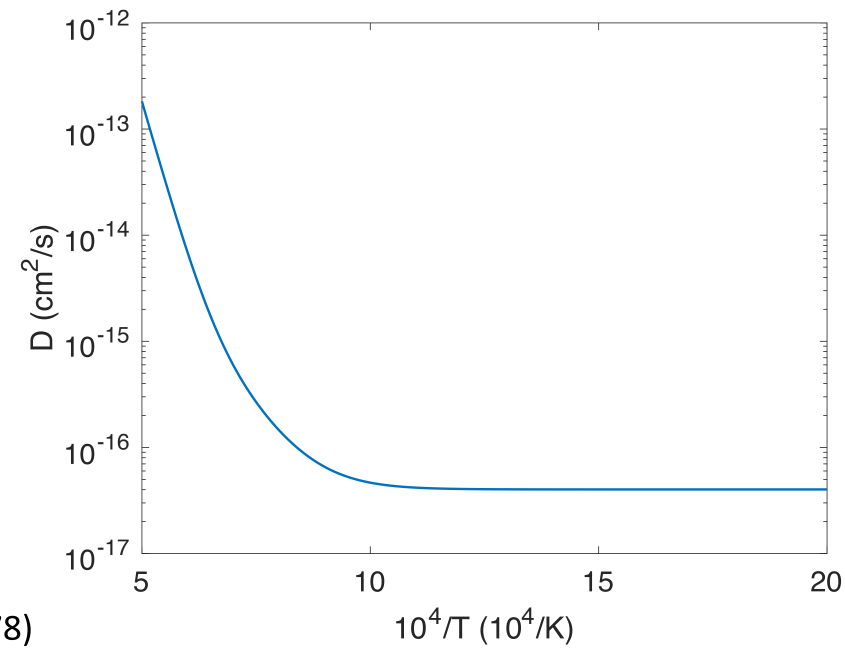


$$D = D_1 + D_2 + D_3 \text{ cm}^2/\text{s}$$

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

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

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



J. A. Turnbull and C.A. Friskney, J. Nucl. Mater. **71**, 238 (1978)

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 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