Nuclear Fuel Performance

NE-533

Spring 2022

Notes

- Had some pushback on additional project
- This additional presentation project will be optional
- Default grading scheme is in the syllabus
- Opt-in grading scheme is outlined in last class
- You must contact me to opt-in to the additional presentation

- Distance students, please schedule your exam with me, even if it will take place during the class period
- On campus students, please let me know if you will take the test virtually

Last Time

- The average grain size in UO_2 impacts fuel behavior and performance
- The material wants to reduce its energy by having large grains grow at the expense of small grains
- Fuel densification is driven by reduction in surface area of pores – continuation of sintering process
- Five families of fission products which uniquely 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

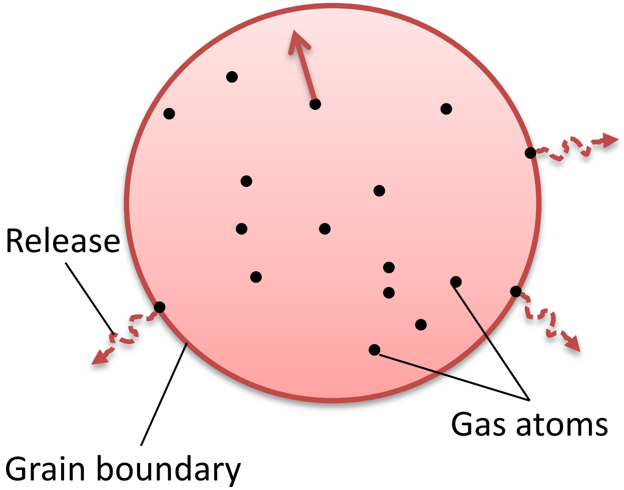
- Fission gas release models attempt to predict the rate at which gas is released from the fuel
- To model fission gas release, ideally, we must model all three stages of gas release
  - Diffusion of gas atoms to grain boundaries
  - Growth and interconnection of grain boundary bubbles
  - Transport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

- 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

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

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



<u>ICs and BCs</u>

$c_g(r, 0) = 0$

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

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

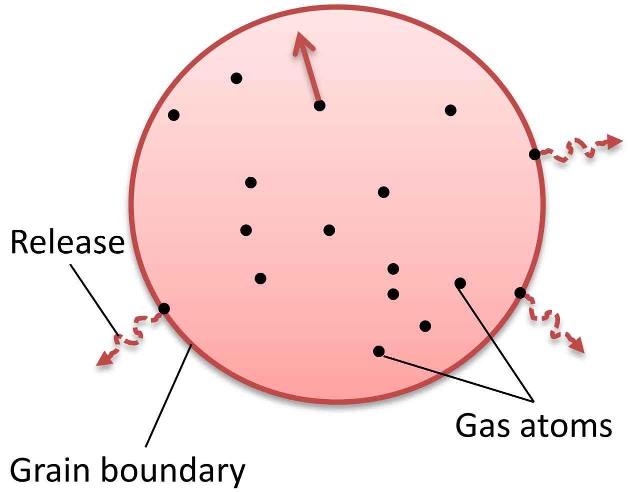
Modeling post-irradiation annealing

- 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

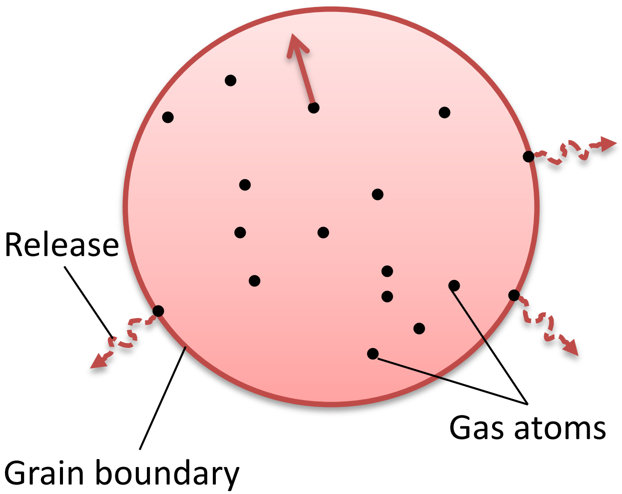
$c_g(r, 0) = c_g^0$

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

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

Solving the Booth Model

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



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

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

Booth Example

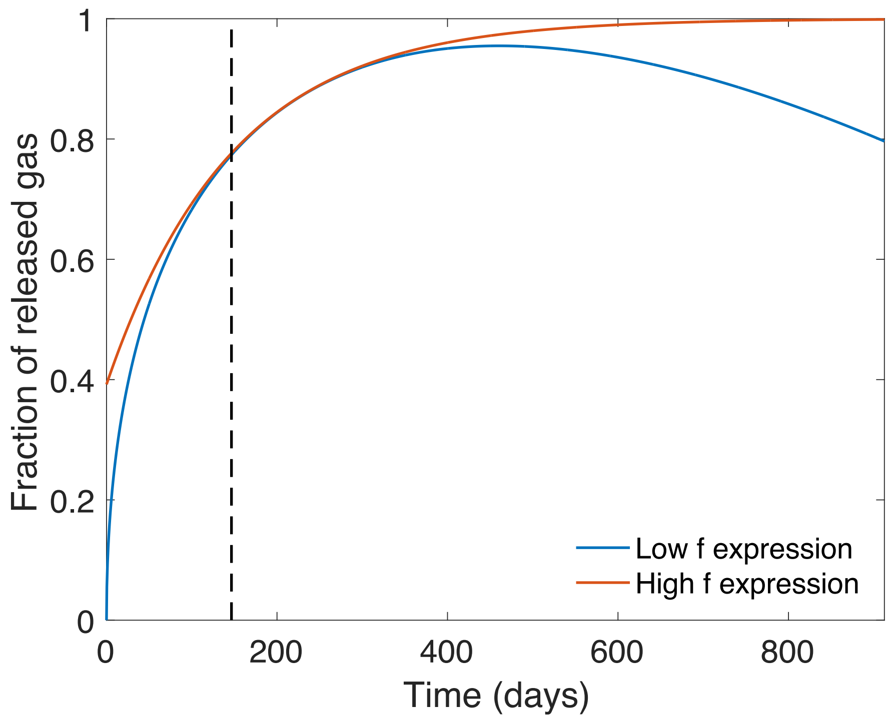
- For a diffusion coefficient for Xe of D = 8e-15 cm^2/s, what fraction of the fission gas trapped in an post-irradiation annealed fuel pellet has escaped after one hour? It has an average grain size of 10 microns
  - D = 8e-15 cm^2/s
  - a = 10e-4 cm
  - t = 3600 s
- Which f? $\tau = D \times t/a^2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

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

  - 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

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

- 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

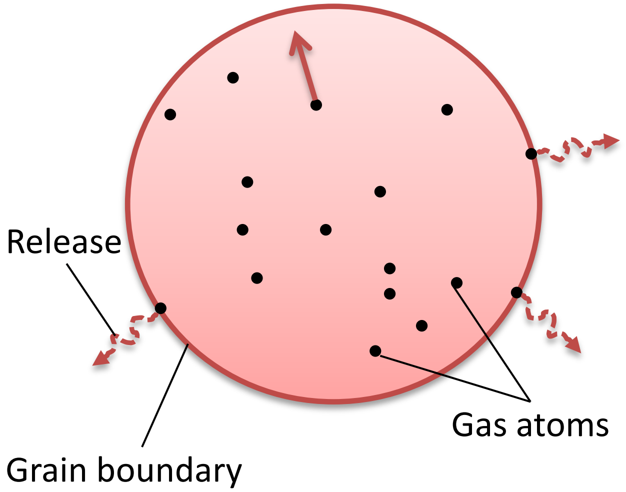
$$f = \frac{3}{ay\dot{F}t}\int_0^t J_a\,dt$$

- 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)

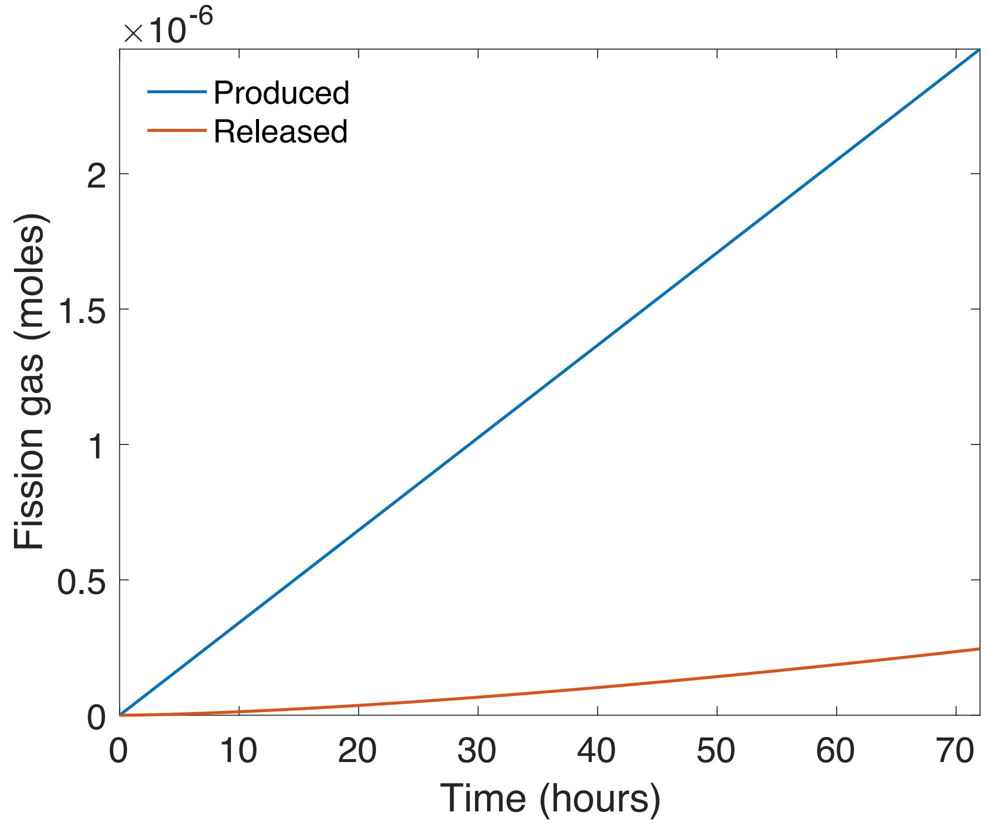
Example

- 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), so we can use:

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

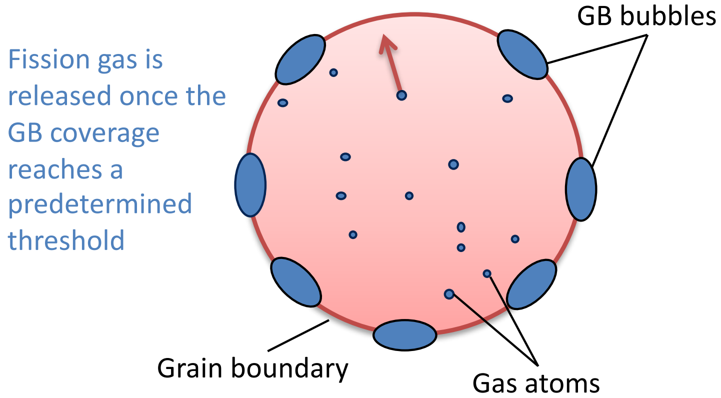
  - f = 4*sqrt(8e-15*3600/(pi*(10e-4)^2)) – 3/2*8e-15*3600/(10e-4)^2 = 0.0121

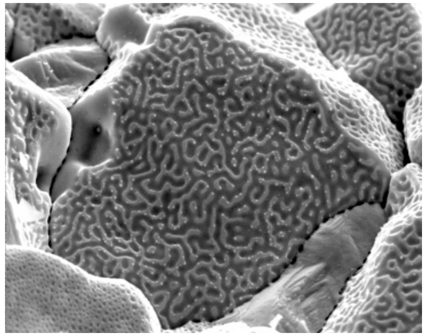
As time progresses, both the fraction released and the produced gas increase



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)





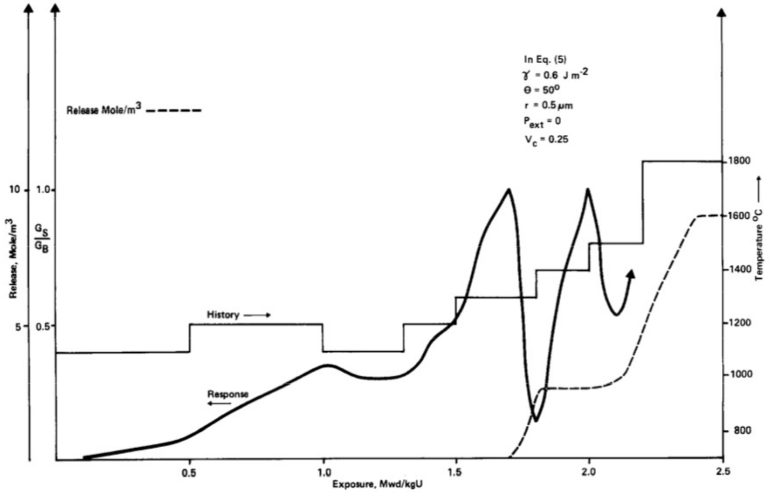
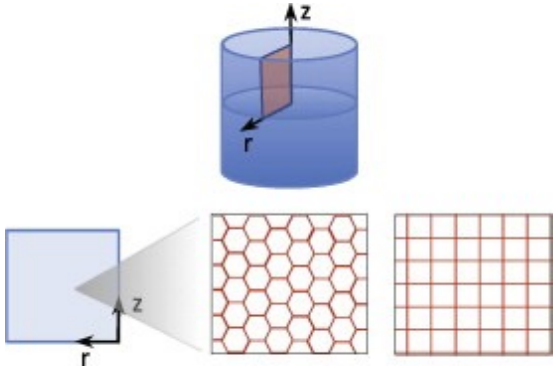
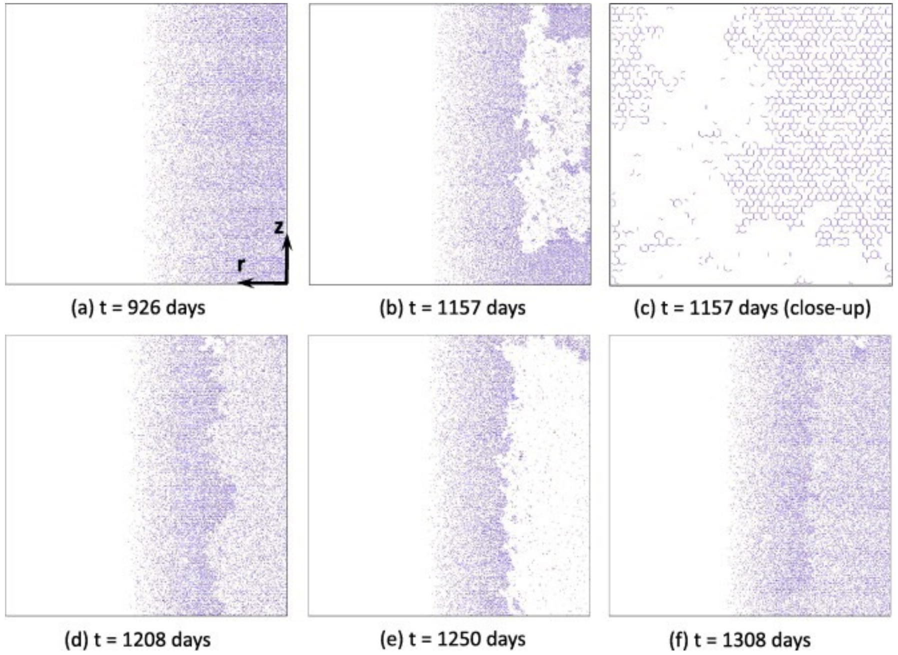


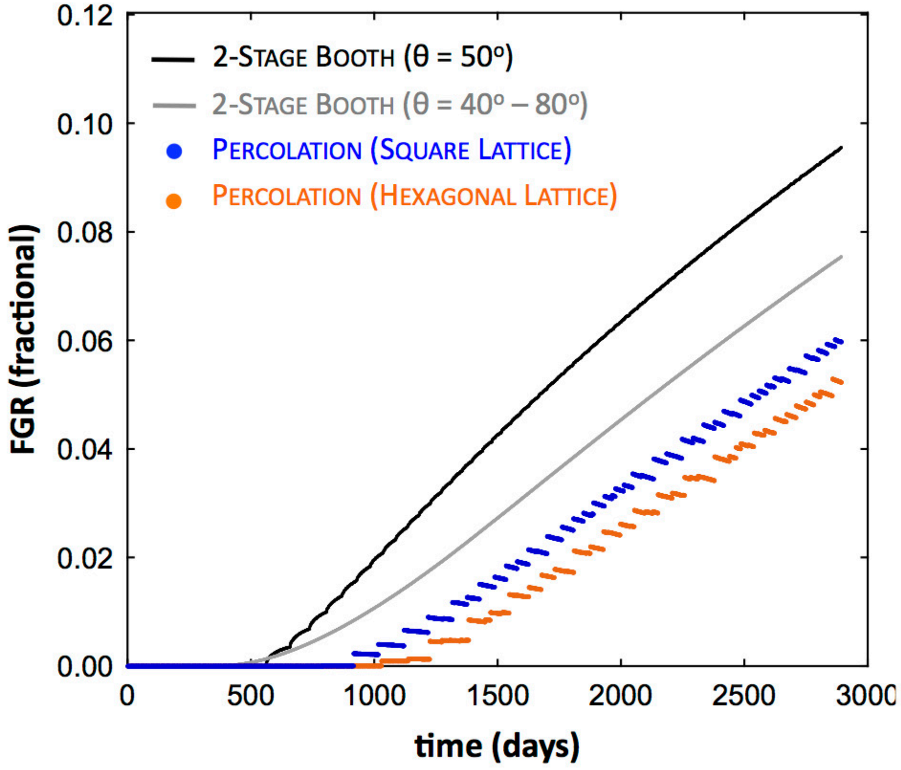
Fig. 1. Fraction of gas atoms on grain boundary, G_s/G_B, as a function of exposure for downward fuel cascading temperature history. γ is the bubble surface tension, 2θ is the angle where two free surfaces meet at a grain boundary, r is average bubble radius, V_c is the fractional coverage of the grain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



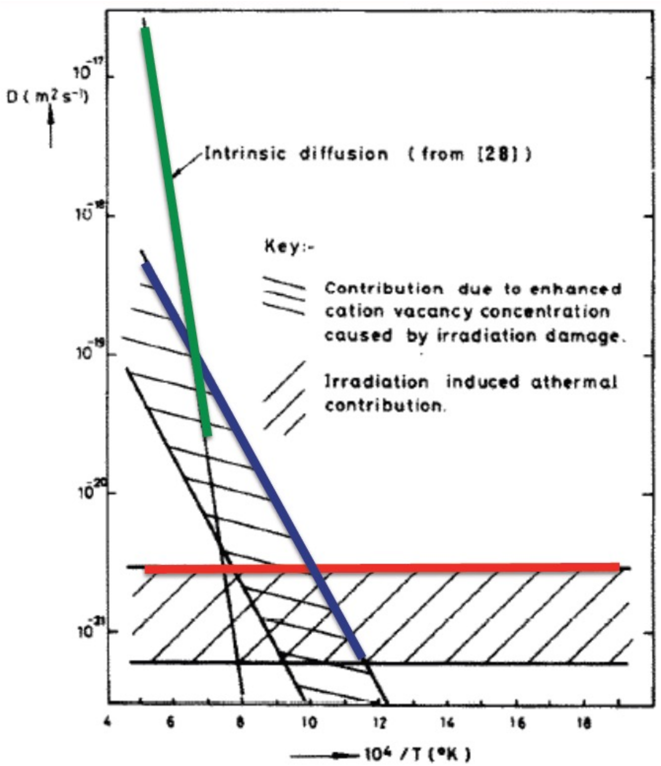




Millett, Tonks, and Biner. JNM, 424.1 (2012): 176-182.

Gas diffusion

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

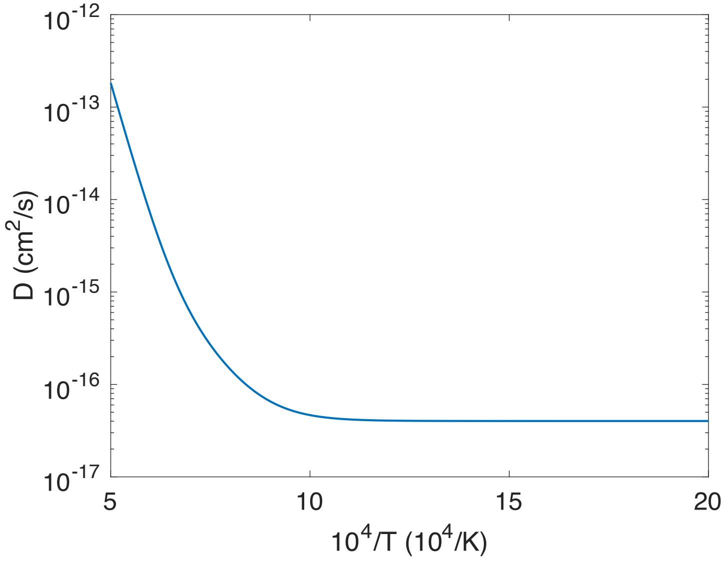


$$D = D_1 + D_2 + D_3 \text{ cm}^2/\text{s}$$

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

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

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



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 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

Notes

- Exam next Thursday
- Open book
- Exams are testing your conceptual knowledge and ability to implement the equations derived/provided
- Conceptual knowledge questions should be able to be answered without consulting notes
- Tests can be completed in the allotted time