PennState
College of Engineering

NucE 497: Reactor Fuel Performance

Lecture 21: Fission products and fission gas

March 13, 2017
Michael R Tonks
Mechanical and Nuclear Engineering

Content taken from Ch 15 of Olander's book

Midsemester evaluation

My overall rating of the course is good.

Strongly disagree		0 %
Disagree		0 %
Somewhat disagree	1 respondents	3 %
Neither agree nor disagree		0 %
Somewhat agree	5 respondents	17 %
Agree	16 respondents	55 %
Strongly agree	7 respondents	24 %

My overall rating of this instructor is favorable.

Strongly disagree		0 %
Disagree		0 %
Somewhat disagree	1 respondents	3 %
Neither agree nor disagree		0 %
Somewhat agree	1 respondents	3 %
Agree	14 respondents	48 %
Strongly agree	13 respondents	45 %

The quizzes prepare me to better learn the material in a module

Strongly disagree	1 respondents	3 %
Disagree	5 respondents	17 %
Somewhat disagree	1 respondents	3 %
Neither agree nor disagree	5 respondents	17 %
Somewhat agree	9 respondents	31 %
Agree	7 respondents	24 %
Strongly agree	1 respondents	3 %

Homework assignments help me understand material.

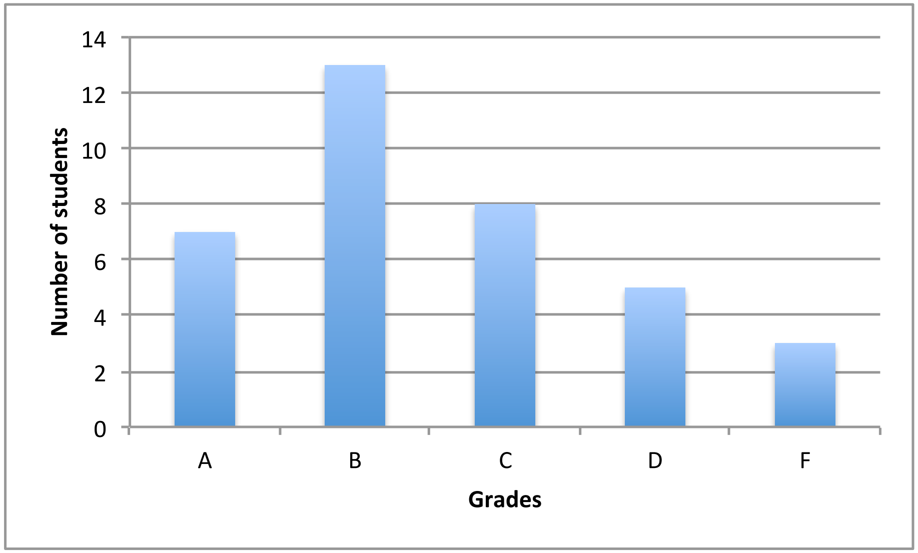
Strongly disagree	2 respondents	7 %
Disagree		0 %
Somewhat disagree	5 respondents	17 %
Neither agree nor disagree		0 %
Somewhat agree	8 respondents	28 %
Agree	8 respondents	28 %
Strongly agree	6 respondents	21 %

Here are some of your comments and my responses:

- Can you add the subject of the lectures to the file names on canvas?
  - **I will do so from now on**
- I would much prefer being quizzed on the material after we learn it.
  - **From now on the quizzes will be on material we have already covered.**
- The homework problems that involve changing your code are difficult
  - **The Matlab assignments where you modified my code were to prepare you for the project.**
  - **There will be no more assignments like that, besides the project**
  - **Note, that if you attempt a HW problem, you get half credit at the least. SO NEVER LEAVE A PROBLEM BLANK**
- Life lessons = very good
  - **I will give at least two more of these before the end of the semester**
  - **If you have a topic you would like me to talk about, let me know**

Exam 1 results

- Mean: 78.2; Std Dev: 14.9
  - Question 1: 24.8/30
  - Question 2: 30.3/35
  - Question 3: 23.2/35



Today we will discuss fission product and fission gas behavior in UO_2

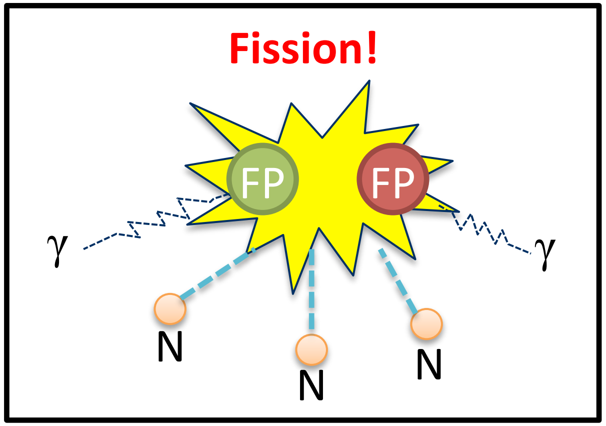
- Module 1: Fuel basics
- Module 2: Heat transport
- Module 3: Mechanical behavior
- Module 4: Materials issues in the fuel
  - Property evolution and Intro to materials science
  - Chemistry
  - Grain growth
  - **Fission products and fission gas**
  - Densification, swelling, and creep
  - HBS
  -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

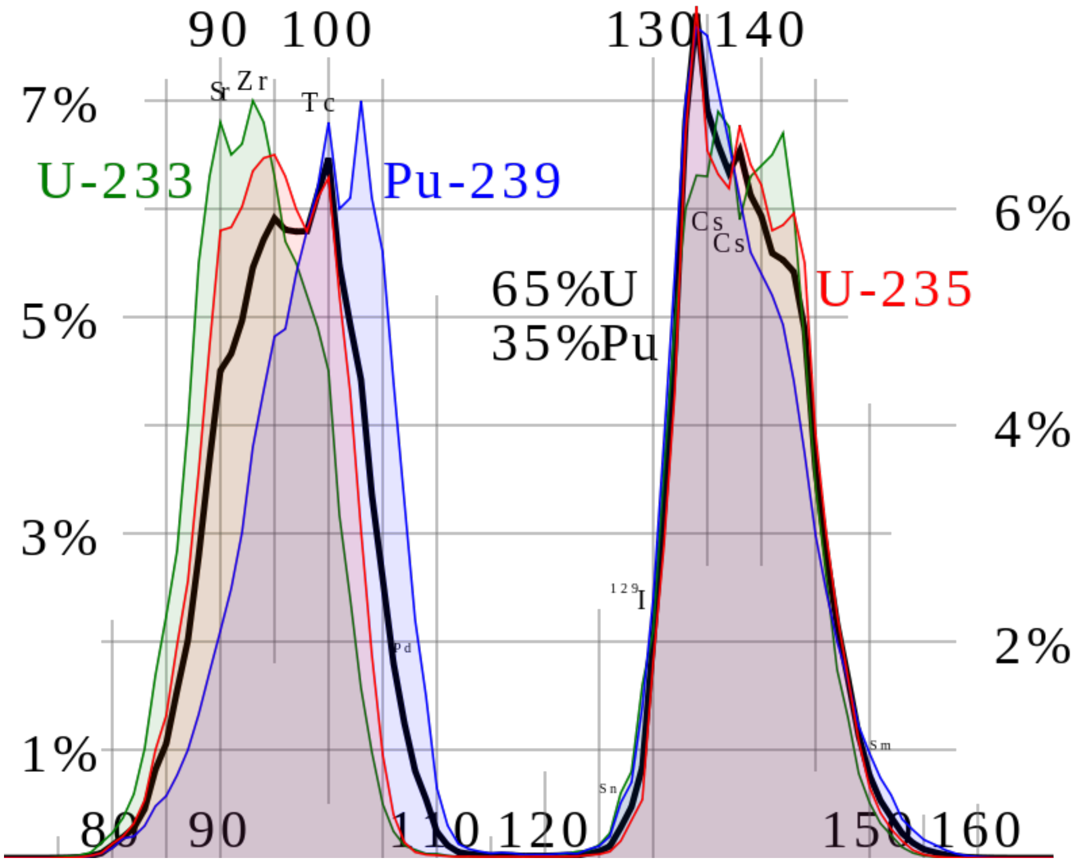
- In the hotter portions of the fuel pellet, the UO_2 grains
  a) Tend to shrink, reducing the average grain size
  b) Tend to grow, increasing the average grain size
  c) Subdivide, reducing the average grain size
  d) Don’t do anything
- Grain boundaries do not impact
  a) Cracking
  b) Fission gas release
  c) Creep
  d) None of the above

Fission produces heat, but it also results in various fission fragments

- 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

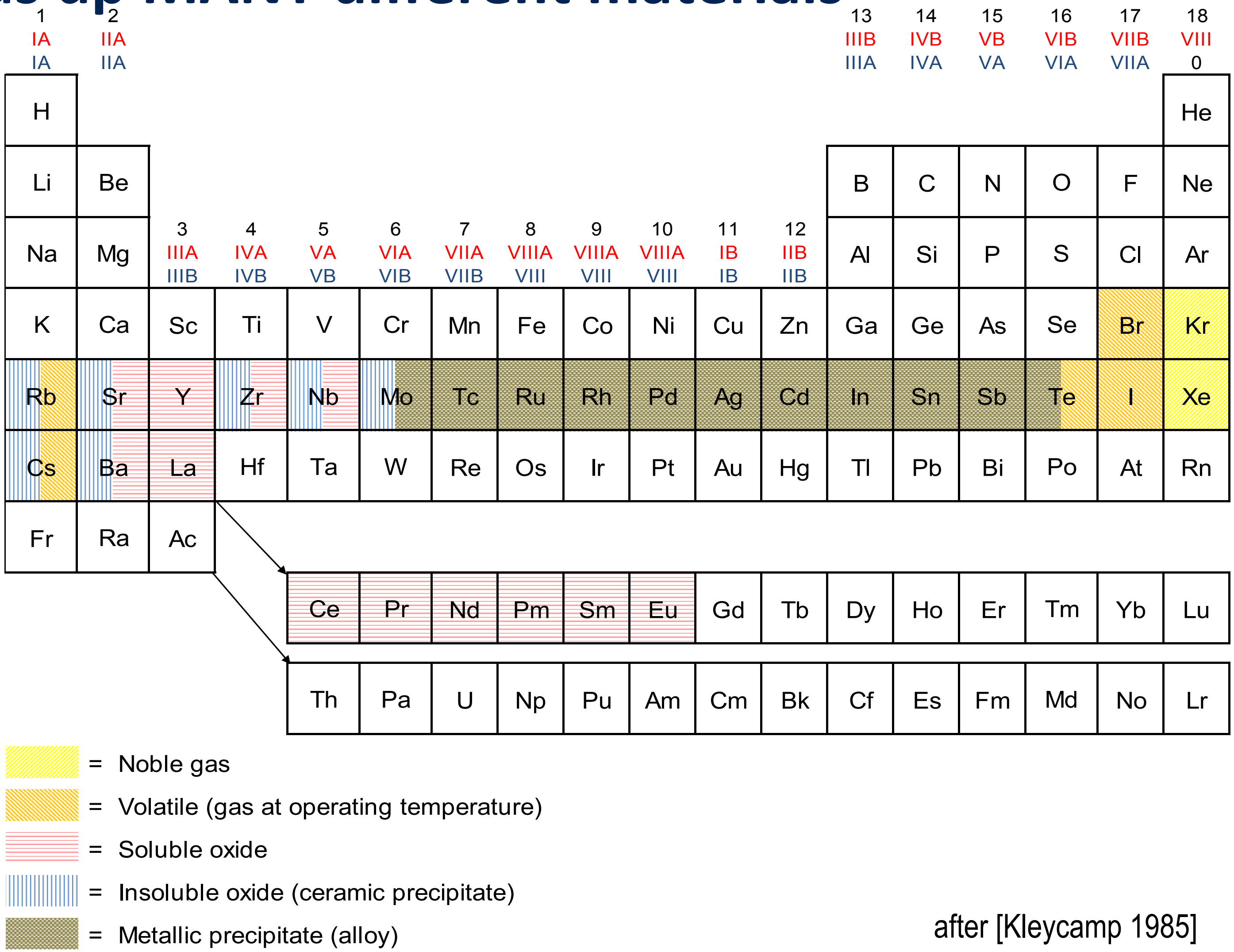


Every fission product that is produced is now in the crystal lattice of the fuel, changing the microstructure



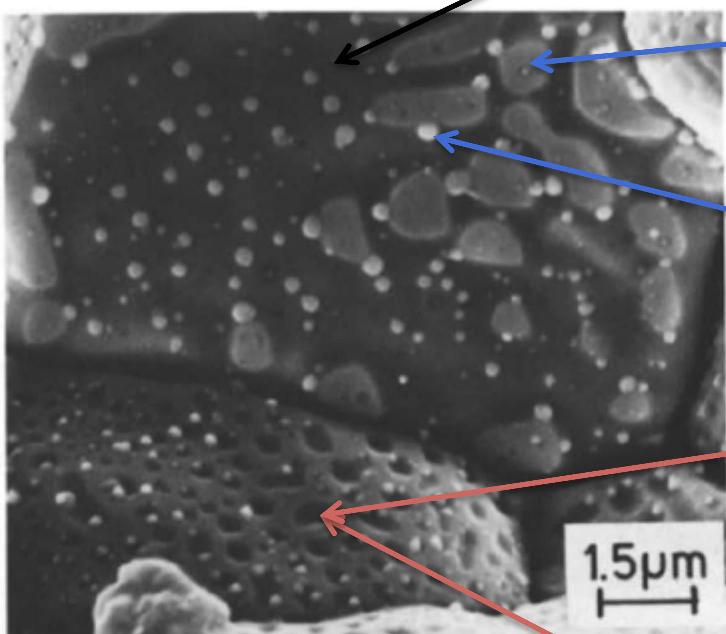
Fission product yields by mass for thermal neutron fission of U-235, Pu-239, a combination of the two typical of current nuclear power reactors, and U-233 used in the thorium cycle. (from Wikipedia)

While the fuel starts with just U and O, it quickly builds up MANY different materials



after [Kleycamp 1985]

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

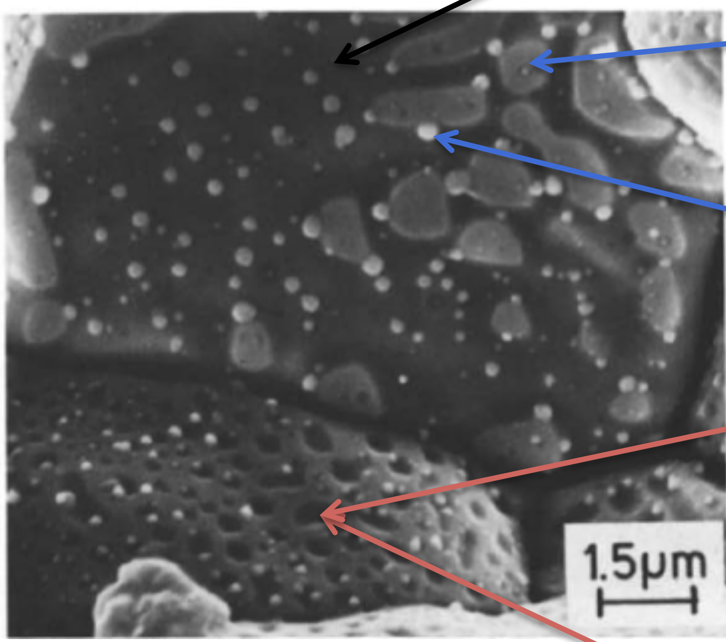
Insoluble oxides (Zr, Ba and Sr)
- Form insoluble oxides in the fluorite lattice

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

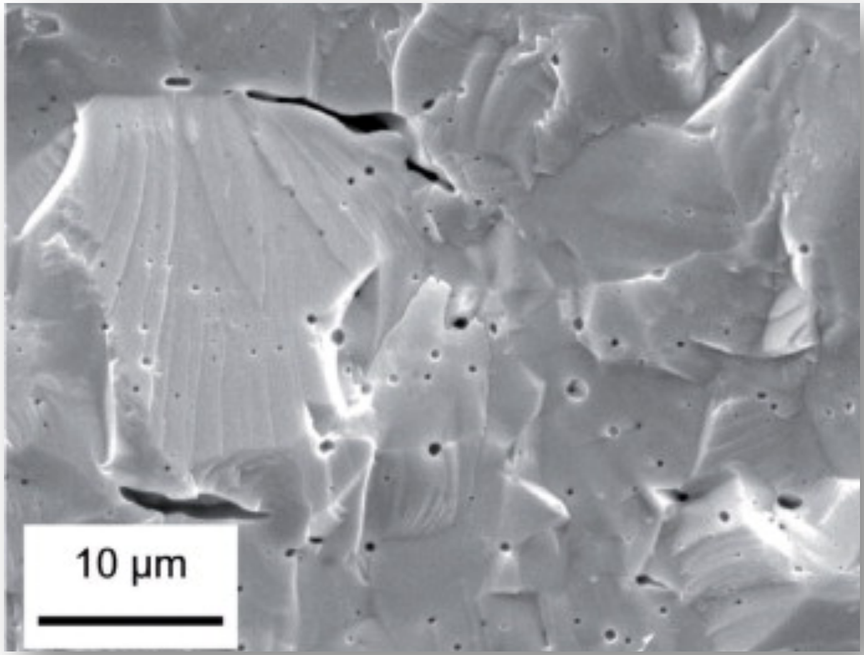
Noble gases (Xe, Kr)
- Cause swelling
- Decrease thermal conductivity
- After release, raise gap pressure and lower thermal conductivity

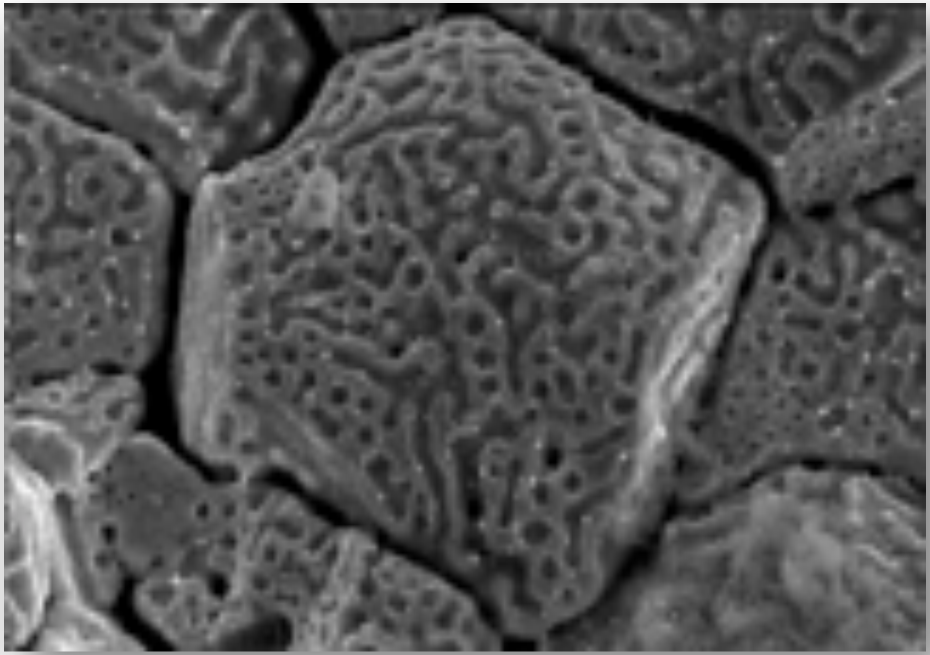
Fission gas is primarily composed of Xe and Kr

Table 13.3 Isotopes of Xenon and Krypton Released in Fission*

Isotope	Half-life	Percent yield ^{235}U	Percent yield ^{239}Pu
^{83}Kr	Stable	0.4	0.3
^{84}Kr	Stable	0.85	0.5
^{85}Kr	Stable (10.6 years)	0.15	0.13
^{85m}Kr†	4.4 hr	1.3	
^{86}Kr	Stable	1.4	0.8
^{87}Kr†	78 min	2.5	
^{88}Kr†	2.8 hr	3.5	
Total stable krypton yields		2.8	1.7
^{131}Xe	Stable	3.2	3.8
^{132}Xe	Stable	4.7	5.3
^{133}Xe†	5.3 day	6.6	6.9
^{134}Xe	Stable	6.6	7.5
^{135}Xe†	9.2 hr	5.5	
^{136}Xe	Stable	5.9	6.6
Total stable xenon yields		20.4	23.2

Fission gases are released in a process composed of three stages

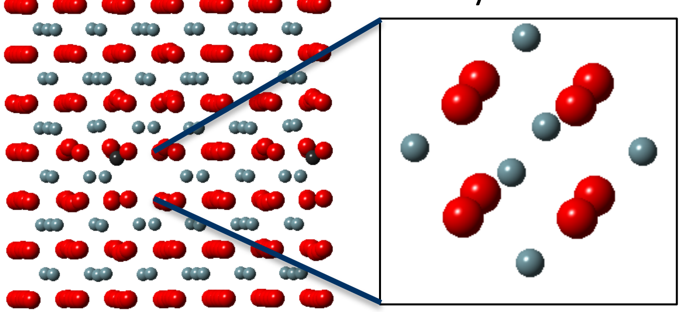


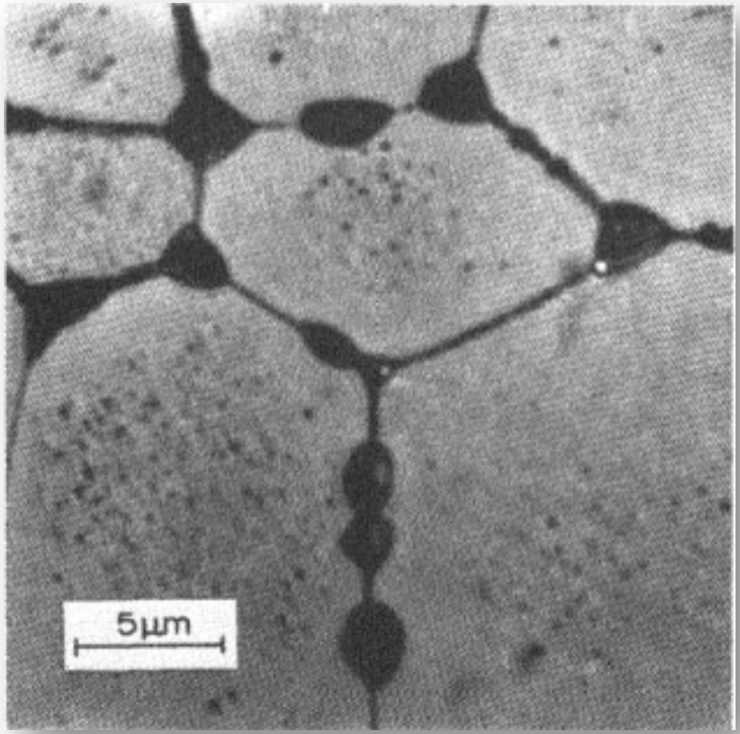


Stage 1: Gas atoms are produced throughout the fuel due to fission and diffuse towards grain boundaries

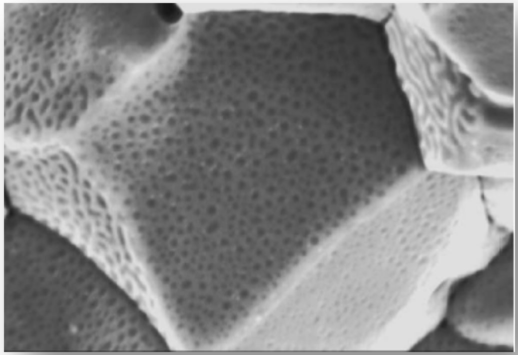
- Small intragranular bubbles form within the grains, but never get larger than a few nm radius due to resolution from energized particles
- Gas atoms that don't get trapped within the intragranular bubbles migrate to grain boundaries

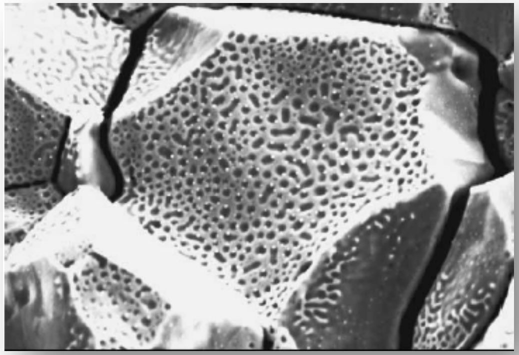
Los Alamos
NATIONAL LABORATORY
EST. 1943

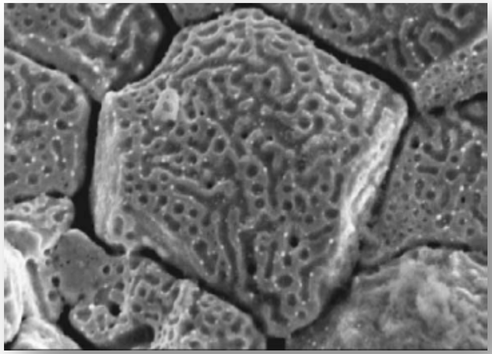


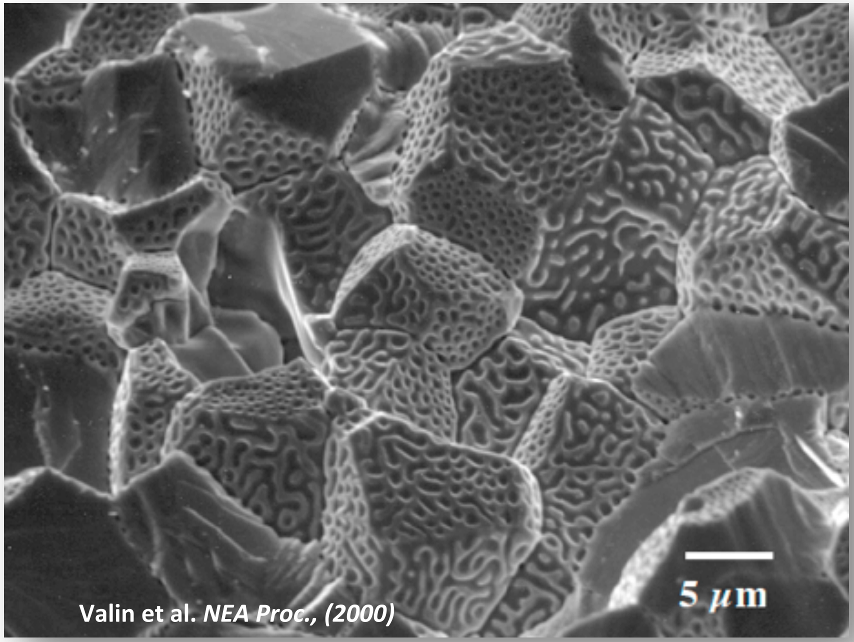


Stage 2: Gas bubbles nucleate on grain boundaries, growing and interconnecting


Nucleation

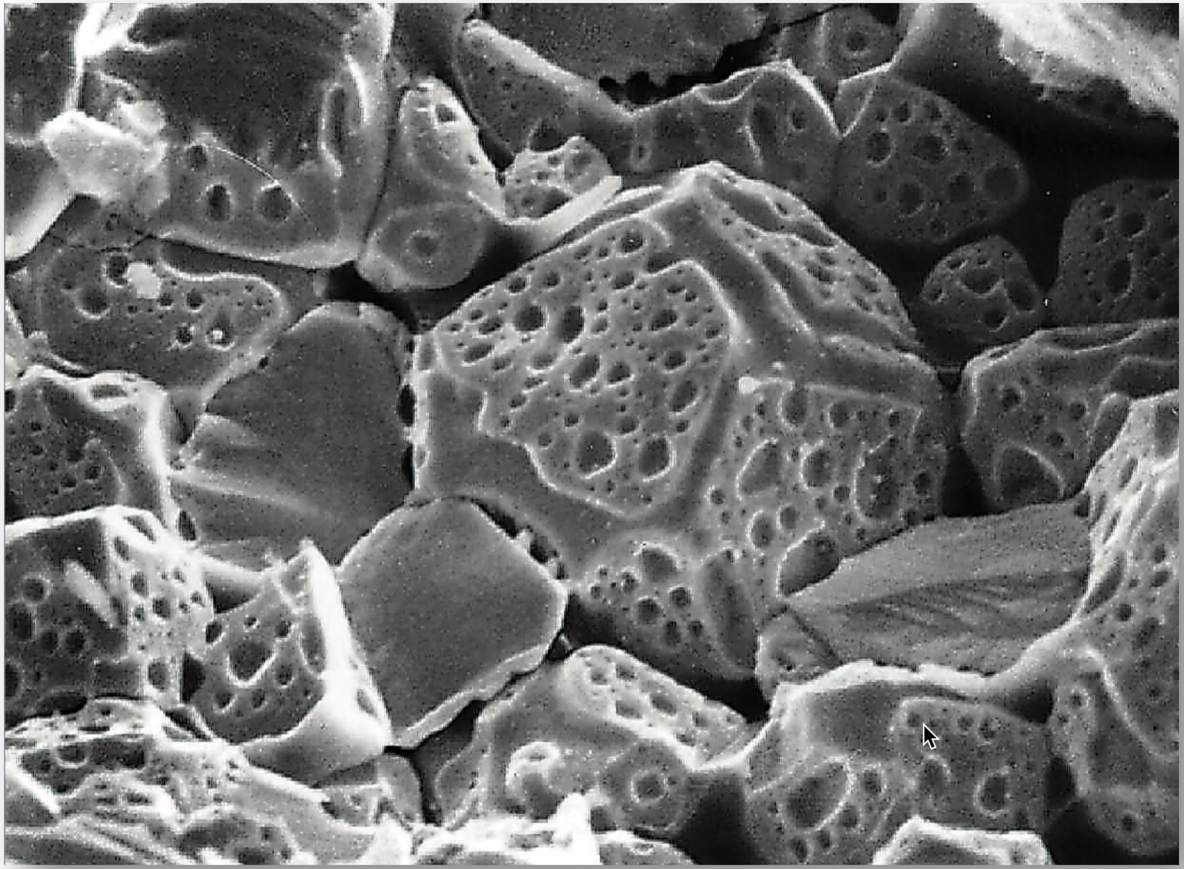

Growth

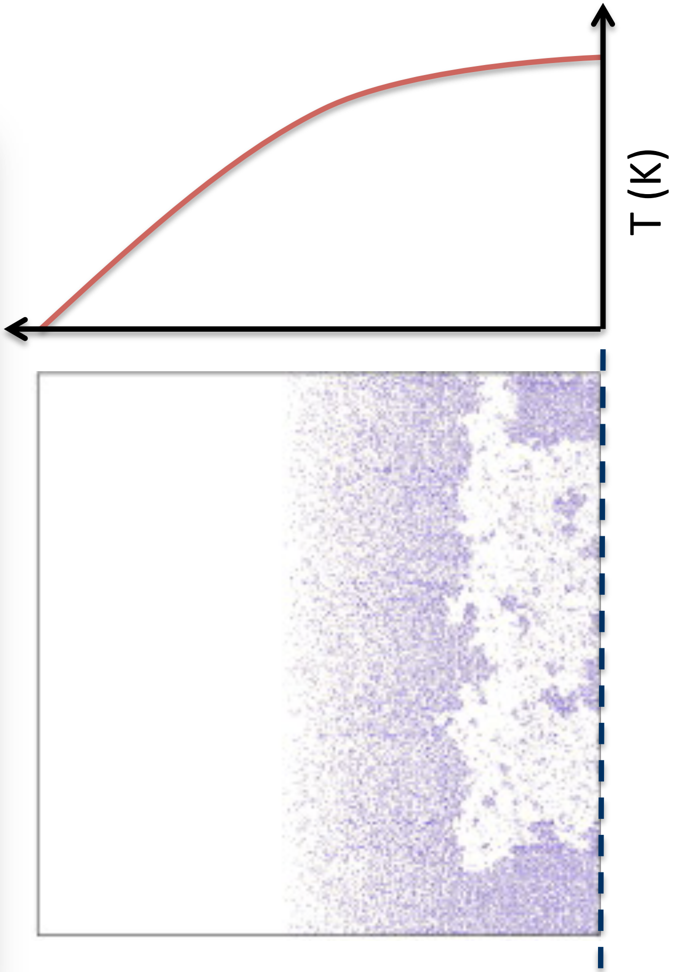

Percolation



Valin et al. *NEA Proc., (2000)*

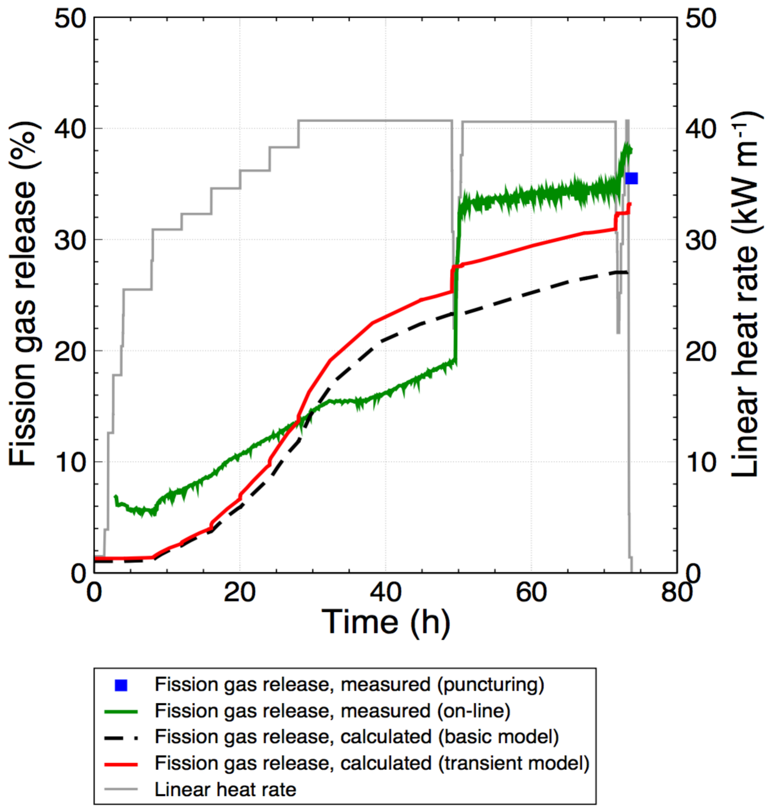
Stage 3: Gas travels through interconnected bubbles to a free surface

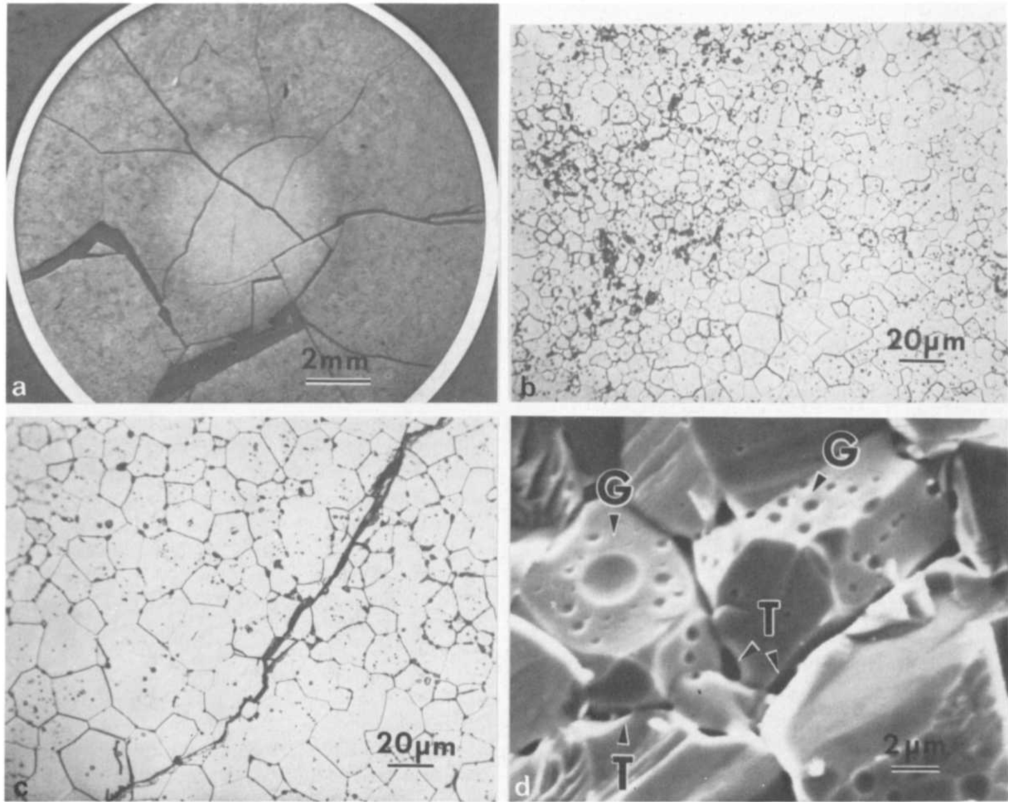




Fission gas release also occurs due to mechanisms that don't depend on diffusion

- Release can occur to particle recoil and knockout at low temperature
- It can occur due to fracture during rapid transients





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

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 into the gap

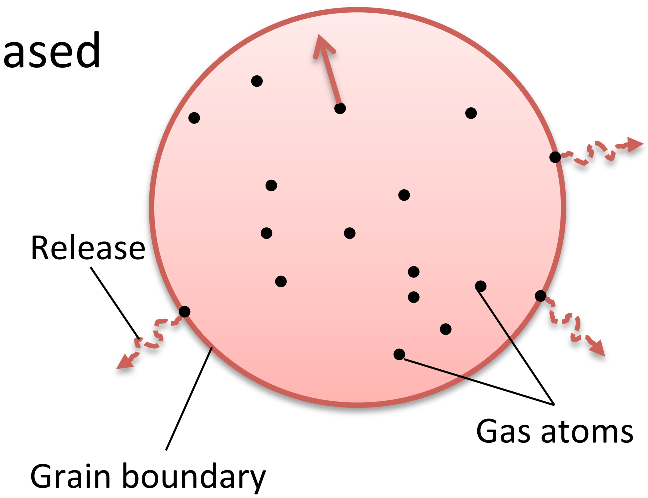
- To model fission gas release, ideally we must model all three stages of gas release
  1. Diffusion of gas atoms to grain boundaries
  2. Growth and interconnection of grain boundary bubbles
  3. Transport of gas atoms through interconnected bubbles to free surfaces
- The earliest models only considered 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

- A grain is considered as a simple sphere
- Once they reach the grain boundary, they are released
- 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

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

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



<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

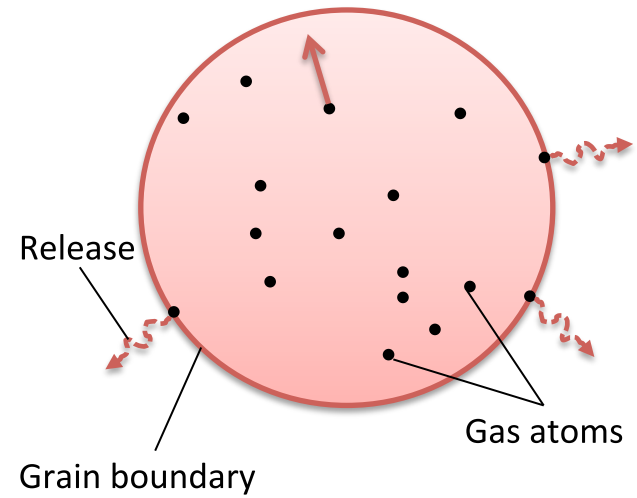
- The initial gas concentration is c_g^0
- No gas is produced

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

- 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

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

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



<u>ICs and BCs</u>

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

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

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

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

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

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 irradiated 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
  - Except for large t, the second term is not significant

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 50% of the fission gas to escape?

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

- $f^2/36 = D\, t/(\pi\, a^2)$
- $t = \pi\, a^2\, f^2/(36{*}D)$
- t = pi*(10e-4)^2*(0.5)^2/(36*8e-15) = 2.7e6 s
- This is 31.25 days

Now, we will model in-pile release

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

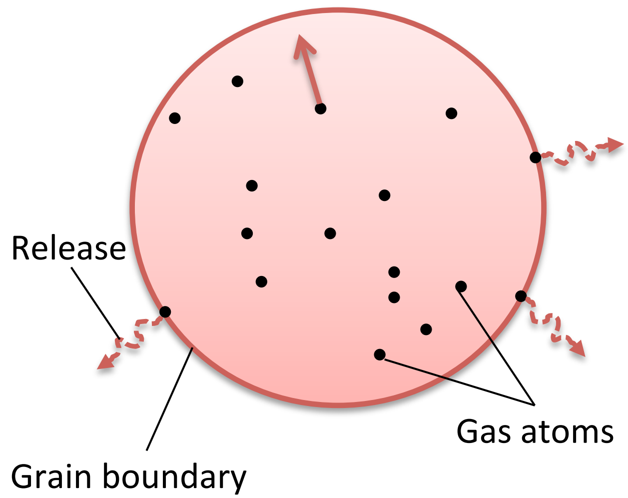
- The initial gas concentration is 0
- Gas is produced due to fission, where y is the chain yield (y = 0.3017 for Xe and Kr) and the fission rate $\dot{F} = qN_U\sigma_{f235}\phi_{th}$
- Gas can also decay, where λ is the decay constant
  - If we only consider stable stable products, λ = 0
- For in pile release, the fraction is equal to

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

- After solving with with a Laplace transform

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

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



ICs and BCs
$c_g(r, 0) = 0$
$c_{g,r}(0, t) = 0$
$c_{g,r}(0, a) = 0$ (release)

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 in pile experiments
- Spherical grain models predict stage one