Chapter 13
Swelling Due to Fission Gases

13.1 Introduction

Because of a.) insolubility,
b.) gaseous nature,
the fission gases tend to promote volume changes in fuel or they tend to change gas pressure (and composition) in the fuel pin.

Need to Know 3 Things

1.) Concentration of atomically dissolved Kr, Xe in fuel,

2.) Number and size distribution of bubbles in fuel and where they are distributed in fuel,

3.) How much gas gets out of fuel.

Void Columnar Zone Equiaxed Zone As Fab.

13.2 General Considerations

13.2.1 Rate of Fission Gas Production

See Table 13.3

Note: • thermal n's for ^{239}Pu,
• fast n's for ^{238}U

Ratio of $\left(\frac{Xe}{Kr}\right)_{Stable}$ ≈ 7.3 for U-38

≈ 13.6 for Pu -39

Yield of (Xe +Kr) ≈ 0.23 -0.25

13.2.2 Xenon Equation of State

- Noble gas can be treated without regard to compound formation

- Normally use VanderWaal's eq. to describe gas

$$P\left(\left[\frac{1}{\rho_g}\right]-B\right)=kT$$

Pressure of Xe in bubble due to one mole of gas	Atomic density of gas in bubble	Normally a Constant ≈ 85 Å^3/atom

13.2.3 Mechanical Force Balance on Bubbles

At equilibrium, the pressure inside the bubble must just balance the external stress.

Surface energy

+ = Comp.
- = Tension

$$P = \frac{2\gamma}{r} + \sigma$$

Pressure of gas pushing out **Hydrostatic Stress**

See Ch. 8

Bubbles grow by adding gas atoms to bubble which increases pressure so that:

$$P > \frac{2\gamma}{r} + \sigma$$

The compression around the bubble attracts vacancies which make the bubble grow, r increases, and pressure drops.

P drops as $\frac{1}{R3}$ **Eventually, equilibrium is reestablished**

$\frac{2\gamma}{r}$ **drops as** $\frac{1}{R}$

13.2.4 Number of gas atoms in a bubble.

Using $$P = \frac{2\gamma}{r} + \sigma$$ **we find,**

$$\left(\frac{2\gamma}{r} + \sigma\right) \bullet \left(\frac{1}{\rho_g}\right) = kT$$

Solving Vander Waal's Eq.,

$$\frac{1}{\rho_g} = B + \left(\frac{1}{\left\{\frac{2\gamma}{kT}\right\}\frac{1}{r} + \frac{\sigma}{kT}}\right)$$

γ **not well known but ≈ 1000** $\frac{\text{ergs}}{\text{cm}^2}$

See Figure 13.3

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Number of Gas Atoms in Bubble

$$m = \left(\frac{4\pi r^3}{3}\right)\rho_g \approx \left(\frac{4\pi r^3}{3}\right)\left(\frac{1}{B}\right) \qquad \text{for } r<10\ \text{Å}$$

$$m = \frac{\left(\frac{4\pi r^3}{3}\right)}{B + \left(\frac{kT}{2\gamma}\right)r} = \left(\frac{4\pi r^2}{3}\right)\left(\frac{2\gamma}{kT}\right) \qquad \text{for } r> 1000\ \text{Å}$$

13.2.5 Swelling Due to Gas Bubbles

Let N’ = Number of bubbles inserted into 1 cm^3
of originally solid material

Now we have 1 cm^3 + bubbles

Let N = Number of bubbles per cm^3 (of solid + bubbles)

$$N' = \frac{N}{1 - \left(\frac{4\pi R^3}{3}\right)N}$$

But now the volume increase is defined as;

bubbles

$$\frac{\Delta V}{V} = \left(\frac{4\pi R^3}{3}\right)N' = \frac{\left(\frac{4\pi R^3}{3}\right)N}{1 - \left(\frac{4\pi R^3}{3}\right)N}$$

Fresh (solid)
fuel = 1 cm^3

——————————————————————————

Not all bubbles are of the same size;

$$\frac{\Delta V}{V} = \frac{\left(\frac{4\pi}{3}\right)\sum R_i^3 N_i}{1-\left(\frac{4\pi}{3}\right)\sum R_i^3 N_i} \approx \left(\frac{4\pi}{3}\right)\int_0^\infty R^3 N(R)\,dR$$

13.2.6 Overall Gas Balance

$$Y_{Xe}\,\dot{F}\,t = C + \sum m\left(R_i\right)N\left(R_i\right)$$

dispersed **(in bubble)** **# of gas atoms in bubbles of size R**

Problem 13.6

13.3 Migration of Atomic Sized defects

13.3.1 Vacancies and Interstitials

from radiation Damage consideration, (Chapt. 17 - 18) we will show that the following sequence of events takes place:

Heavy or
Fission --------------> Displaced atoms Recombine (some)
Light FP's

Stable atoms moved from Lattice Site

Takes place in $\approx 10^{-12}$ s, then defects want to move.

- ***≈ 10,000 vacant lattice site per FP***
- ***≈ 10,000 atom stuck in nonequilibrium positions***
- ***≈ 0.25 Xe atom / fission***

Diffusion coefficients;

$$D_v = \lambda_v^2 \nu_v \exp\left(\frac{S_v^*}{R}\right)\exp\left(-\frac{\varepsilon_v^*}{RT}\right)$$

jump distances	**Freq.**	**Entropy of Motion**	**Energy of Motion**

$$D_i = \lambda_i^2 \nu_i \exp\left(\frac{S_i^*}{R}\right)\exp\left(-\frac{\varepsilon_i^*}{RT}\right)$$

Usually $\lambda \approx a_o \approx 3$ **Å**

$\nu \approx 10^{13}\ s^{-1}$

S, ε for ceramics are not well known, so we use the values found for metals

$\varepsilon_v^* = 80$ **kJ/mole** $S_v^* \approx 40$ **J / mole °K**

$\varepsilon_i^* = 20$ **kJ/mole** $S_i^* \approx 0$

interstitials much more mobile than vacancies

13.3.2 Xenon in UO_2

Xe does not seem to migrate by;

- **a.) interstitial mechanisms**
- **b.) simple vacancy mechanism**

It is found that Xe diffuses by a combination of 1 U vacancy and 2 Oxygen vacancies.

If Xe did diffuse by vacancies;

$$D_{Xe} = \lambda^2_{Xe} x_v \omega$$

Frac. of vacancies
on sublattice used

$$\nu_v \exp\left(\frac{S^*_{Xe}}{R}\right)\exp\left(-\frac{\varepsilon^*_{Xe}}{RT}\right)$$

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One could control the diffusion by controlling the vacancy concentration through chemical additions (see Chapter 11 -12)

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With the mechanism uncertain, have to rely on empirically determined diffusion coefficients

13.4 Reaction Rates of Atomic Size Defects

With species A and B in the matrix;

$$\text{reaction rate A, B} = k_{AB}\, C_A\, C_B$$

C_A, C_B are volumetric concentrations of A, B

- A can be mobile or stationary
- B can be mobile or stationary

Two forms of reactions;

1.) *Diffusion (mobility) controlled*
2.) *Reaction Rate Limited*

13.4.1 Vacancy -Vacancy Interaction

$$V + V \xrightarrow{k_{vv}} V_2$$

See Figure 13.4

(Dienes and Damask were the first to treat this problem)

$$\frac{\text{Rate of Divacancy Formation}}{\text{cm}^3} = P_{vv}C_v$$

$P_{vv} = 12\ P_x$ (see fig. 13.4)

prob/s that another
vacancy jumps into
a site that is nearest
neighbor to a particular
vacancy $P_x = 7\ x_v\omega$

rate at which
vacancy jumps
to $x_v = C_V\ \Omega$ a
particular site

$\frac{\#}{\text{unit vol.}}$ vacancy site fraction

$$\frac{\textbf{Rate of divacancy formation}}{\mathrm{cm}^3} = 84\,\omega\,\Omega\,C_V^2$$

This gives $$k_{vv} = 84\,\omega\Omega = \frac{84\Omega D_V}{a_0^2}$$

$$\omega = \frac{D_v}{a_o^2}$$

Remember:

This is for a fcc lattice only, it would be different for a bcc structure because would get a different combinatorial number. If both vacancies were mobile, k_{vv} ***should be multiplied by 2 .***

13.4.2 Vacancy - Interstitial Recombination

Analogous to the divacancy case

$$V + i \xrightarrow{k_{vi}} \textit{Nothing}$$

immobile mobile

which gives;

$$k_{vi} = \frac{48\Omega D_i}{a_0^2}$$

12 x 4

nearest interstitial

neighbors positions

Reason that this is not right is that interstitial atoms do not occupy nearest neighbor positions .... and ..... recombination volume is a lot larger

Figure 13.5

get $$k_{vi} = \frac{Z_{iv}\Omega D_i}{a_0^2}$$

more like
several hundred

<u>13.4.3</u> <u>Interaction</u> <u>between</u> <u>Migrating</u> <u>Fission</u> <u>Gas</u> <u>Atoms</u>

Assume that both gas atoms are mobile

$$g + g \xrightarrow{k_{11}} g_2$$

do <u>not</u> consider reverse reaction now

Analogous to the vac - int problem

$$k_{11} = \frac{Z_{11}\Omega D_{Xe}}{a_0^2}$$

much larger than 84

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the formation of a diatomic cluster is probably a prelude to nucleation and hence is <u>very</u> important

rate of formation of diatomic clusters $= k_{11}C^2$

13.4.4. Interaction Between Xenon Atoms and Atomic Sized Defects

Gas atoms are not the only defects that other gas atoms can interact with, they can find point defects as well

Rate of trapping of fission gas atoms/cm^3 = $k_{gtr}\,C_t\,C$

traps gas atoms

as before,

$$k_{gtr} = \frac{Z_{gtr}\Omega D_{Xe}}{a_0^2}$$

$$\frac{\text{Rate of Gas Atom Trapping}}{cm^3} = \frac{D_{Xe}C}{L^2}$$

Trapping length

L^2 = j a_o^2

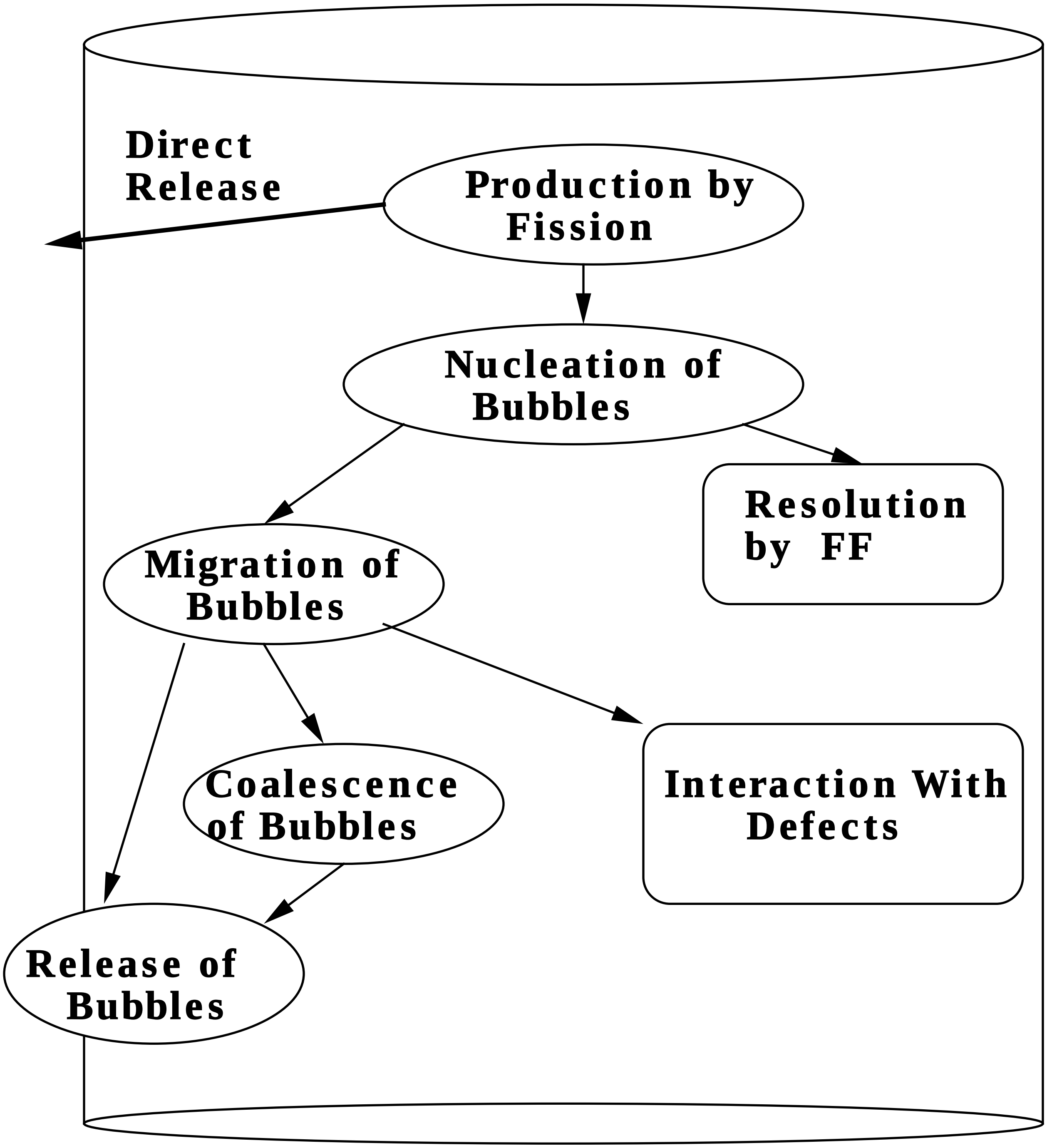
random walk theory

# of jumps to get to trap

lattice spacing

$$j = \frac{1}{Z_{gt}C_t\Omega}$$

Behavior of Fission Gases in Fuel



Direct Release

Production by Fission

Nucleation of Bubbles

Resolution by FF

Migration of Bubbles

Coalescence of Bubbles

Interaction With Defects

Release of Bubbles

See Table 13.2 for Varibles Affecting Fission Gas Behavior