Chapter 13
Swelling Due to Fission Gases

13.1 Introduction

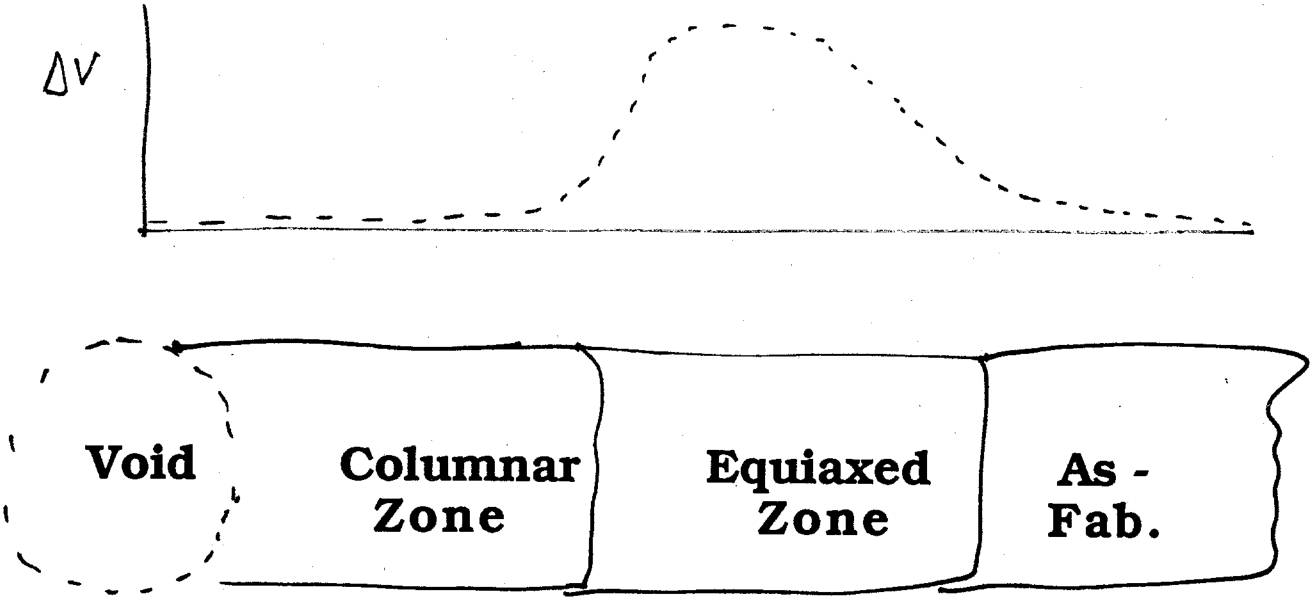
Because of a.) insolubility,
b.) gaseous nature,
the fission gases tend to promote volume changes in fuel or they tend to change gas pressure (and composition) in the fuel pin.

Need to Know 3 Things

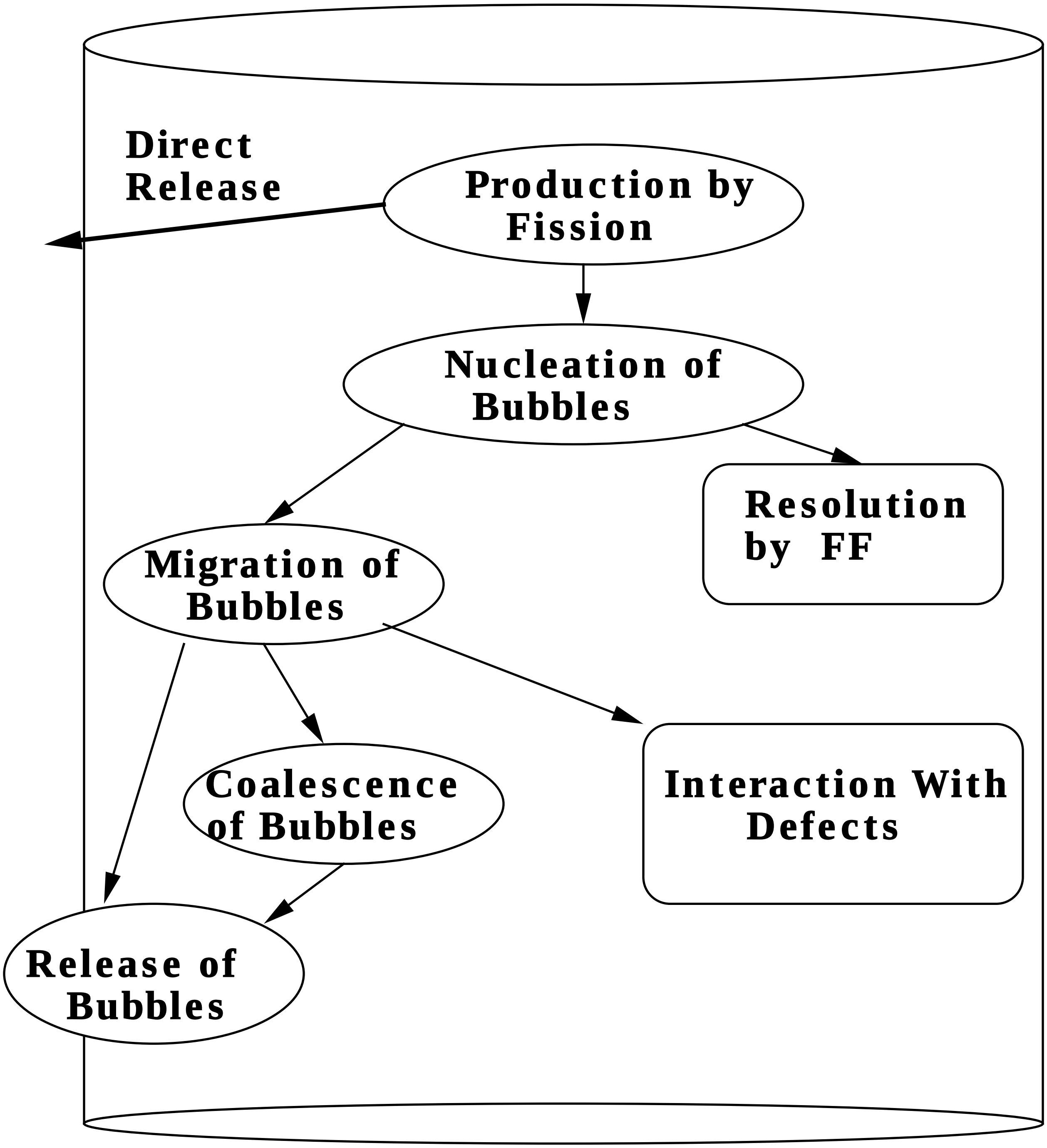
1.) Concentration of atomically dissolved Kr, Xe in fuel,

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

3.) How much gas gets out of fuel.



Behavior of Fission Gases in Fuel



See Table 13.2 for Varibles Affecting Fission Gas Behavior

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**

Table 13.3 Isotopes of Xenon and Krypton Released in Fission*

Isotope	Half-life	Percent yield ^{238}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

*From S. Katcoff, *Nucleonics*, 18: 201 (1960). The yields for the stable products represent chain yields; the yields of short-lived isotopes are cumulative yields up to the particular isotope. Plutonium-239 yields are for thermal neutrons, whereas those for ^{238}U are for fast neutrons.

†Short-lived fission product.

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 $\approx 1000\ \frac{\text{ergs}}{\text{cm}^2}$

See Figure 13.3

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Number of Gas Atoms in Bubble

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

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

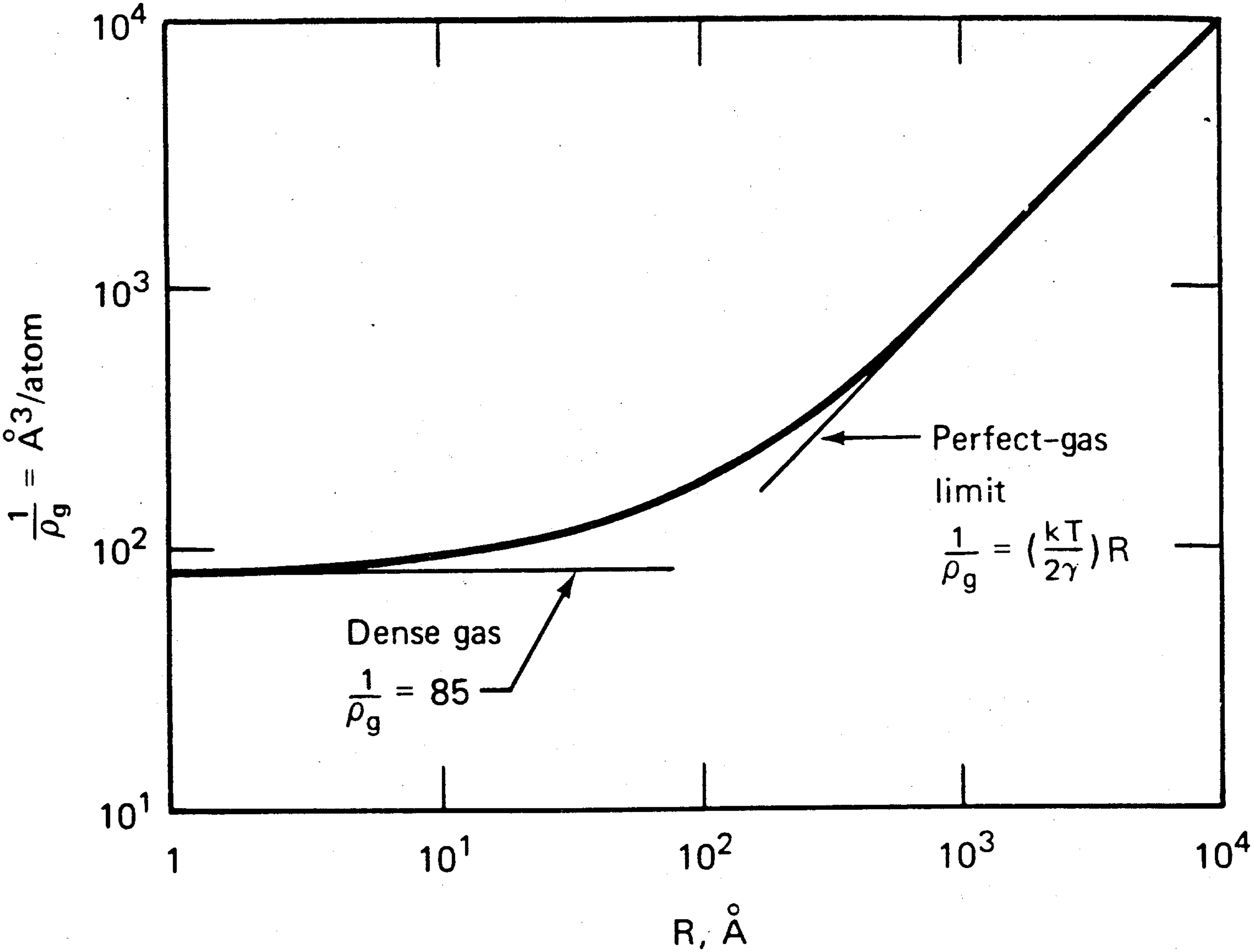

Fig. 13.3 Density of xenon gas in a spherical bubble imbedded in a stress-free solid of surface tension of 1000 dynes/cm.

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 ≈ 10^{-12} s, then defects want to move.

Problem 13.6

- In a fuel pellet we have N bubbles/cm^3 of radius R plus one bubble of radius R*
- Both bubble sizes are in equilibrium and large enough to use perfect gas laws.
- When R* exceeds R_c*, the large bubble can gobble up small bubbles and grow spontaneously.
- Determine R_c* (critical radius)
- If R_c*= 10R, what is swelling at breakaway?

Condition of mechanical equilibrium means

$$p = \frac{2\gamma}{r} \quad \text{and} \quad p\left(\frac{4\pi r^3}{3}\right) = mkT$$

or,

$$m = \left[\frac{8\pi\gamma}{3kT}\right] r^2 = Cr^2 \qquad 1)$$

If the large bubble expands by dR*, it sweeps out a volume $4\pi R^{*2}\, dR^{*}$.

This volume contains N bubbles of radius R per cc and each buble contains

$$m = CR^2 \quad \text{atoms}$$

Therefore, the number of additional gas atoms acquired by the large bubble as a result of expansion by dR* is;

$$dm^{*} = 4\pi R^{*^{2}} \bullet dR^{*} \bullet N \bullet C \bullet R^{2} \qquad 2)$$

However, to maintain mechanical equilibrium according to eq. 1), the number of additional gas atoms required is given by;

$$dm^{*'} = \frac{d\left(CR^{*^{2}}\right)}{dR^{*}} dR^{*}$$

$$= 2CR^{*} dR^{*} \qquad 3)$$

The bubble will grow spontaneously if

$$dm^{*} > dm^{*'}$$

or if $4\pi R^{*^{2}} \bullet N \bullet C \bullet R^{2} > 2CR^{*}$

or, $R^{*} \geq \dfrac{1}{2\pi NR^{2}} \qquad 4)$

Now if $\frac{R^{*}}{R} = 10$**, then from eq. 4**

$$N = \frac{1}{2\pi \bullet 10 \bullet R^{3}}$$

But

$$V_{bubble} = \frac{4\pi R^3}{3}$$

$$NV_{bub} = \Delta V = \frac{\frac{4\pi R^3}{3}}{2\pi \bullet 10 \bullet R^3} = \frac{2}{30} = 0.0667$$

Swelling $\dfrac{\Delta V}{V} = \dfrac{0.0667}{1 - 0.0667} = \dfrac{0.0667}{0.9333} = 7.2\%$

- *≈ 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;

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 a
particular site

$x_v = C_V \Omega$

$\frac{\#}{\text{unit vol.}}$

vacancy site fraction

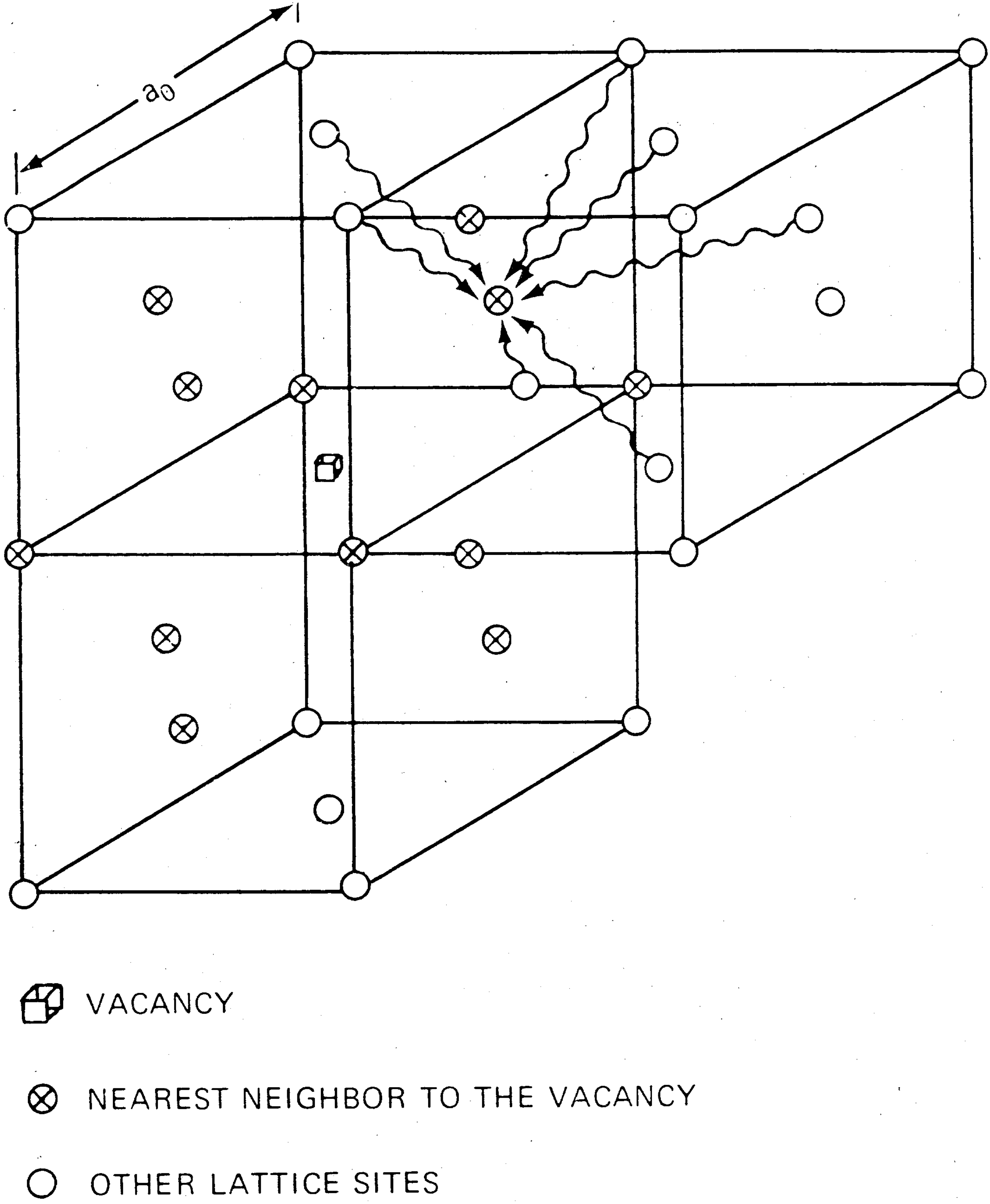

Fig. 13.4 Diagram for computing the rate of divacancy formation in a face-centered cubic lattice.

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

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

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

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

13.4.3 Interaction between Migrating Fission Gas Atoms

Assume that both gas atoms are mobile

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

do not 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 very important

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

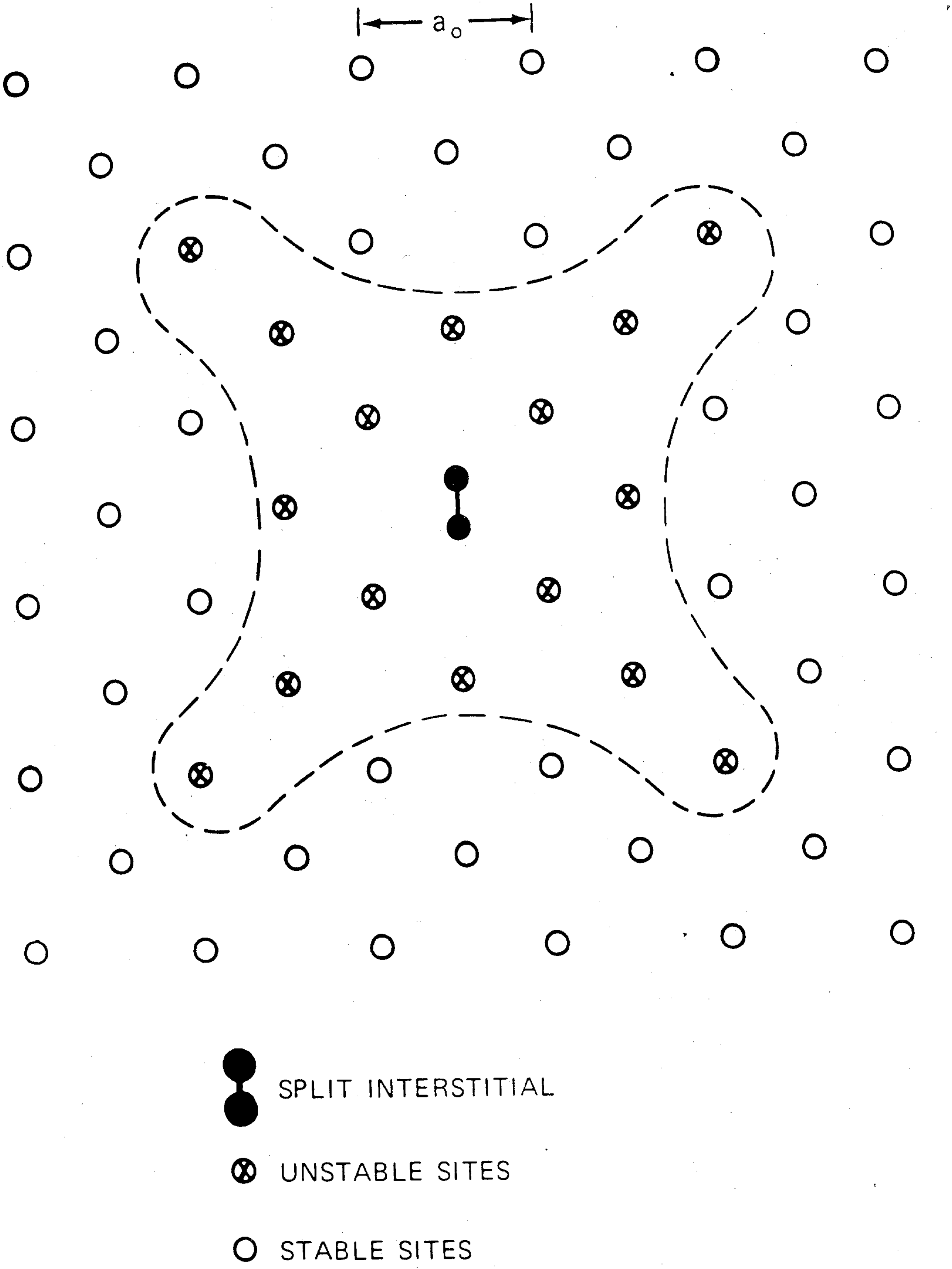

Fig. 13.5 Stability of Frenkel pairs in the (100) plane of copper. The dashed line separates stable from unstable vacancy sites. [After Gibson et al., *Phys. Rev.*, 120: 1229 (1960).]