

Chapter 7

Radiation damage

The structural materials of nuclear power plants interact with various radiations. The most interesting of these is the interaction with neutrons because of the remarkable degradation of the mechanical properties of the material, commonly known as “neutron irradiation embrittlement”.

7.1 Neutron-Nucleus interaction

7.1.1 Elastic scattering

Because neutrons are electrically neutral, they do not react well with electrons. The main reaction between neutrons and atoms begins when the neutron collides with the nucleus of an atom. If the energy of the incident neutron is E_i , and the energy of the neutron after collision with the nucleus is E_f , when Ω is defined as the solid angle at the time of collision, the collision probability is

$$\sigma_s(E_i, \Omega) = \int \sigma_s(E_i, E_f, \Omega) dE_f$$

The total scattering probability for neutrons of energy E_d is

$$\sigma_s(E_i) = \int \sigma_s(E_i, \Omega) d\Omega$$

Materials interact with various radiations. However, the content is vast and difficult to cover all in one semester. So boldly, in this lesson, we will only consider interactions with neutrons. There are mainly lab frame and center of mass frame to describe collision. The two frames do not conflict with each other and give the same result. But it's a matter of convenience. I think many of you have studied Reactor theory class. Here, I will simply write the results. If the mass of the neutron is 1 and the mass of the nucleus is A , applying momentum balance

$$v_c - V_c A = 0 \tag{7.1}$$

$$v'_c - V'_c A = 0$$

By energy balance,

$$\frac{1}{2}v_c^2 + \frac{A}{2}V_c^2 = \frac{1}{2}v_c'^2 + \frac{A}{2}V_c'^2$$

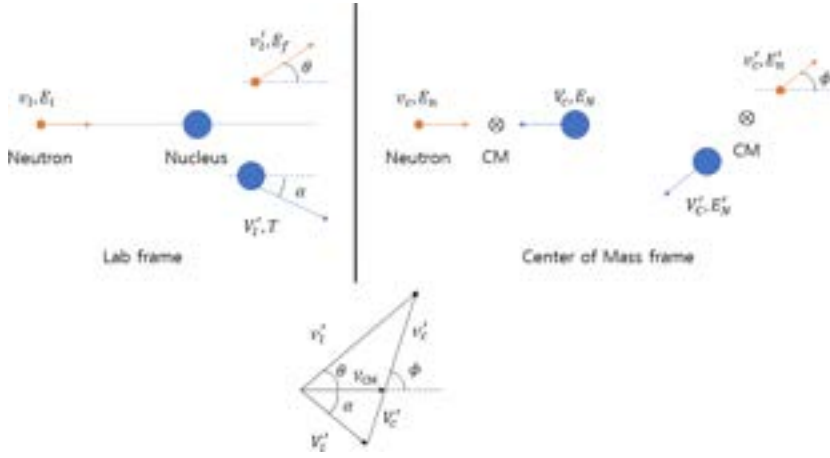


Figure 7.1: Lab frame and center of mass frame

Combined energy conservation, we have

$$v_c = v'_c$$

$$V_c = V'_c$$

which is the virtue of center of mass frame. Before collision, target nucleus at rest at laboratory frame and moves left direction in Fig. 7.1 with speed V_c in center of mass system. The relative velocity is

$$v_c = v_i - V_{CM} = v_i - V_c$$

Plug into Eq. 7.1,

$$V_{CM} = \left(\frac{1}{A+1} \right) v_i \quad (7.2)$$

Apply cosine law to diagram in Fig. 7.1,

$$V_l'^2 = V_{CM}^2 + V_c'^2 - 2V_{CM}V_c' \cos \phi$$

With the energy T transferred to the nucleus by the irradiated neutron,

$$V_l'^2 = \frac{2T}{A} \quad V_{CM}^2 = 2E_i \left(\frac{1}{A+1} \right)^2 \quad V_c'^2 = \frac{2}{A^2} E_n'$$

Finally, we reach

$$T = \eta_1 \eta_2 E_i + \frac{\eta_1}{\eta_2} E_n' - 2\eta_1 \sqrt{E_i E_n'} \cos \phi \quad (7.3)$$

where

$$\eta_1 = \frac{1}{A+1} \quad \eta_2 = \frac{A}{A+1}$$

E_n' is given by

$$E_n' = \eta_2^2 E_i \quad (7.4)$$

By further handling,

$$T = \frac{\gamma}{2} E_i (1 - \cos \phi) \quad (7.5)$$

1 MeV n on C	$\gamma = 0.28$	$\bar{T} = 0.14$ MeV
1 MeV n on Fe	$\gamma = 0.069$	$\bar{T} = 0.035$ MeV
1 MeV n on U	$\gamma = 0.017$	$\bar{T} = 0.009$ MeV

where

$$\gamma = \frac{4A}{(A+1)^2}$$

and ϕ is the scattering angle. Within range $0 \leq \phi \leq \pi$, as ϕ increases, T increases accordingly. By definition of solid angle,

$$d\Omega = \frac{dA}{r^2} = \frac{rd\phi(2\pi r \sin \phi)}{r^2} = 2\pi \sin \phi d\phi$$

Since the scattering probability is also function of T or ϕ , we can write down

$$\sigma_s(E_i, T)dT = \sigma_s(E_i, \phi)d\Omega = 2\pi\sigma_s(E_i, \phi) \sin \phi d\phi \quad (7.6)$$

From Eq. 7.5,

$$dT = \frac{\gamma}{2} E_i \sin \phi d\phi \quad (7.7)$$

we also have

$$\sigma_s(E_i, T) = \frac{4\pi}{\gamma E_i} \sigma_s(E_i, \phi) \quad (7.8)$$

Since

$$\sigma_s(E_i) = \int \sigma_s(E_i, \phi)d\Omega = 2\pi \int \sigma_s(E_i, \phi) \sin \phi d\phi$$

When scattering is isotropic, we can write

$$\sigma_s(E_i) = 4\pi\sigma_s(E_i, \phi)$$

With Eq. 7.8,

$$\sigma_s(E_i, T) = \frac{\sigma_s(E_i)}{\gamma E_i}$$

which does not depend on T . The average recoil energy is

$$\bar{T} = \frac{\int_{\hat{T}}^{\check{T}} T \sigma_s(E_i, T)dT}{\int_{\hat{T}}^{\check{T}} \sigma_s(E_i, T)dT} = \frac{\hat{T} + \check{T}}{2} \simeq \frac{\hat{T}}{2} = \frac{\gamma E_i}{2} \quad (7.9)$$

7.2 The displacement of Atoms

The struck lattice atom energy T is referred to as a primary knock-on atom (PKA). An atom that collides with a neutron moves out of its place and collides with its neighboring atoms, causing consequent further escape. This large-scale additional displacement of neighboring atoms is called a collision cascade.

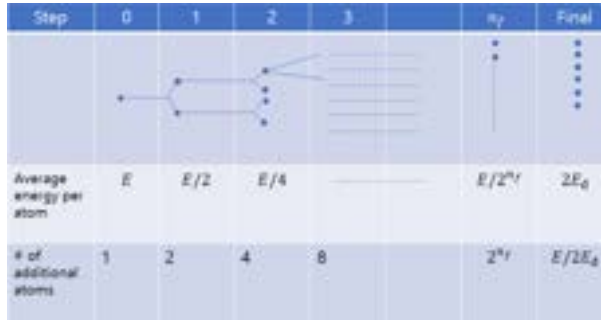


Figure 7.2: Illustration of Kinchin-Pease model

7.2.1 Kinchin-Pease(K-P) model

By Eq. 7.9, assume that the average kinetic energy of the atoms is halved with each collision. (Assuming collisions between atoms of the same mass) The figure proposed by Kinchin and Pease is shown in Fig. 7.2. The details of Kinchin-Pease model is when initial PKA

$$E > 2E_d$$

1. PKA causes further atomic displacements.
2. After the first collision, the number of displaced atoms is 2, their average energy

$$\bar{T}_1 = \frac{E}{2}$$

3. After the second collision, four atoms are involved.

$$\bar{T}_2 = \frac{E}{4}$$

4. In general, after n collisions, 2^n atoms are involved and

$$\bar{T}_n = \frac{E}{2^n}$$

5. The termination condition is

$$\bar{T}_n \leq 2E_d$$

6. When

$$E_d < \bar{T}_n < 2E_d$$

PKA can transfer enough energy to displace an atom, but the displaced atom is replaced by the PKA, so no new defects are produced.

7. n_f is final number of collisions

$$2E_d = \frac{E}{2^{n_f}}$$

Number of displacements when the projectile energy is E_i

$$\nu(E_i) = \underbrace{\nu(E_i - T)}_{\text{Recoil}} + \underbrace{\nu(T)}_{\text{Struck atom}} \quad (7.10)$$

The energy transfer cross section when $\gamma = 1$ is

$$\sigma_s(E_i, T) = \frac{\sigma_s(E_i)}{E_i}$$

The probability that a PKA of energy E_i transfers energy in the range $(T, T + dT)$ is

$$\frac{\sigma_s(E_i, T)dT}{\sigma_s(E_i)} = \frac{dT}{E_i}$$

With Eq. 7.10,

$$\begin{aligned} \nu(E_i) &= \frac{1}{E_i} \int_0^{E_i} [\nu(E_i - T) + \nu(T)] dT \\ &= \frac{1}{E_i} \left[\int_0^{E_i} \nu(E_i - T) dT + \int_0^{E_i} \nu(T) dT \right] \end{aligned}$$

Let

$$X = E_i - T$$

Then

$$\begin{aligned} \nu(E_i) &= \frac{1}{E_i} \left[\int_0^{E_i} \nu(X) dX + \int_0^{E_i} \nu(T) dT \right] \\ &= \frac{2}{E_i} \int_0^{E_i} \nu(T) dT \end{aligned}$$

We can easily know that

$$\begin{aligned} \nu(E_i) &= 0 \quad \text{for} \quad 0 < E_i < E_d \\ \nu(E_i) &= 1 \quad \text{for} \quad E_d \leq E_i < 2E_d \end{aligned}$$

Therefore, the integration can be partitioned by

$$\nu(E_i) = \frac{2}{E_i} \left[\int_0^{E_d} 0 dT + \int_{E_d}^{2E_d} 1 dT + \int_{2E_d}^{E_i} \nu(T) dT \right]$$

yielding

$$\nu(E_i) = \frac{2E_d}{E_i} + \frac{2}{E_i} \int_{2E_d}^{E_i} \nu(T) dT \quad E_i > 2E_d \quad (7.11)$$

Multiplying E_i and take derivative with respect to E_i we have

$$E_i \frac{d\nu(E_i)}{dE_i} = \nu(E_i)$$

The solution of ODE is

$$\nu(E_i) = CE_i$$

Put it back to Eq. 7.11, we can determine C

$$\nu(E_i) = \frac{E_i}{2E_d}$$

Another upper limit also presents E_c which does not increase displacement atoms any longer. In sum,

$$\nu(E_i) = \begin{cases} 0 & \text{for } E_i < E_d \\ 1 & \text{for } E_d \leq E_i < 2E_d \\ \frac{E_i}{2E_d} & \text{for } 2E_d \leq E_i < E_c \\ \frac{E_c}{2E_d} & \text{for } E_i \geq E_c \end{cases}$$

7.3 Displacement rates

The displacement rate is

$$R_d = \int_{\bar{E}}^{\hat{E}} N \phi(E_i) \sigma_d(E_i) dE_i \quad (7.12)$$

The unit of R_d is displacements/m³ · s and unit of N is atoms/m³. $\sigma_i(E_i)$ is the displacement cross section. Neglecting inelastic scattering and assume all scattering is isotropic,

$$\sigma_d(E_i) = \frac{\sigma_s(E_i)}{\gamma E_i} \int_{E_i}^{\gamma E_d} \nu(E_i) dE_i$$

If $\gamma E_d > E_c$ with Kinchin-Pease model,

$$\begin{aligned} \sigma_d(E_i) &= \frac{\sigma_s(E_i)}{\gamma E_i} \left[\int_{E_d}^{2E_d} dE_i + \int_{2E_d}^{E_c} \frac{E_i}{2E_d} dE_i + \int_{E_c}^{\gamma E_i} \frac{E_c}{2E_d} dE_i \right] \\ &= \frac{\sigma_s(E_i)}{2\gamma E_i E_d} \left[\gamma E_i E_c - \frac{E_c^2}{2} \right] \end{aligned}$$

When $\gamma E_i \sim E_c$

$$\sigma_d(E_i) \simeq \left(\frac{\gamma E_i}{4E_d} \right) \sigma_s(E_i)$$

Eq. 7.12 becomes

$$\begin{aligned} R_d &= \frac{N\gamma}{4E_d} \int_{E_d/\gamma}^{\infty} \sigma_s(E_i) E_i \phi(E_i) dE_i \\ &= N \sigma_s \left(\frac{\gamma \bar{E}_i}{4E_d} \right) \phi \end{aligned}$$

where \bar{E}_i is an average neutron energy and ϕ is the total neutron flux above energy E_d/γ , and the term in brackets is the number of displacements (Frenkel pairs) produced per neutron.

For example, 0.5 MeV neutrons in Fe in a fast flux,

$$N = 0.85 \times 10^{23} \text{ atoms/cm}^3 \quad \sigma_s = 3 \times 10^{-24} \text{ cm}^2$$

$$\phi = 10^{15} \text{ neutrons/cm}^{-2} \text{ s}^{-1} \quad \frac{\gamma \bar{E}_i}{4E_d} = 350 \text{ displaced atoms/neutron}$$

then

$$R_d = 9 \times 10^{16} \text{ displaced atoms/cm}^3 \cdot \text{s}$$

or

$$\frac{R_d}{N} = 10^{-6} \text{ dpa/s}$$

or

$$\frac{R_d}{N} = 32 \text{ dpa/year}$$

7.4 Rate theory

When neutrons are irradiated into the solid, the atom hit by the neutron sometimes leaves its own site (PKA, self-interstitial atom), and the original site becomes empty (vacancy). These (self) interstitial and vacancy are called point defects in the solid, and interstitial and vacancy are collectively called Frenkel defect.

These Frenkel pairs cause collective chaos of millions of atoms around the neutron-irradiated region, which is called a cascade. Numerous Frenkel defects are formed here, and the formed defects interact with each other. However, of course, although it depends on the irradiated neutron energy and flux and the properties of the material, the natural always tends to find its equilibrium state, and after several tens of ps, most of the Frenkel pairs disappear. Atoms that leave the original site return to where they should be. However, very few of them cannot find their home and become 'defect' or 'damage'.

Let's discuss the situation after Cascade a little more deeply.

7.4.1 Damage after the cascade

There are largely four main processes after the cascade, in terms of defect.

1. Production
2. Recombination
3. Absorption at sinks
4. Migration

As with all balances in the world, the point defect (vacancy, interstitial) balance based on the Fick's 2nd law is given by the Eqs. 7.13 and 7.14.

$$\frac{dC_v(\mathbf{r}, t)}{dt} = \nabla \cdot D_v \nabla C_v + G_v - L_v \quad (7.13)$$

$$\frac{dC_i(\mathbf{r}, t)}{dt} = \nabla \cdot D_i \nabla C_i + G_i - L_i \quad (7.14)$$

where C_v and C_i are concentrations of vacancy and interstitial and G_v and L_v are gain and loss rate of vacancies and G_i and L_i are gain and loss rate of interstitials. The possible gain terms of the defects are

1. Displacement production
2. Reaction production

The possible loss terms are

1. Recombination
2. Loss to sinks. The possible sinks are
 - (a) Dislocations
 - (b) Cavity
 - (c) etc.

For simplicity, we do not consider grain of defects by reaction production in this lecture. We assume that defect production is function of neutron flux and it is constant during the irradiation, K_0 .

Now, we take care about recombination term, a sort of defect loss, which means defect annihilation by interaction of vacancies and interstitials.

$$\left. \frac{dC_v(\mathbf{r}, t)}{dt} \right|_{\text{recombination}} = \left. \frac{dC_i(\mathbf{r}, t)}{dt} \right|_{\text{recombination}} = -K_{iv}C_vC_i$$

Another loss term by sinks, we have to estimate sink strengths depending on the type of sinks.

$$\begin{aligned} \left. \frac{dC_v(\mathbf{r}, t)}{dt} \right|_{\text{sink}} &= -k_v^2 D_v C_v \\ \left. \frac{dC_i(\mathbf{r}, t)}{dt} \right|_{\text{sink}} &= -k_i^2 D_i C_i \end{aligned}$$

We consider two sinks, cavity and dislocations,

$$k_v^2 = z_v \rho_d + 4\pi r_c N_c \quad (7.15)$$

$$k_i^2 = z_i \rho_d + 4\pi r_c N_c \quad (7.16)$$

where r_c and N_c are cavity radius and number density of cavities, and ρ_d is dislocation density and z_v and z_i are bias factor for vacancies and interstitials. Eqs. 7.13 and 7.14 becomes

$$\frac{dC_v(\mathbf{r}, t)}{dt} = \nabla \cdot D_v \nabla C_v + K_0 - K_{iv}C_vC_i - k_v^2 D_v C_v \quad (7.17)$$

$$\frac{dC_i(\mathbf{r}, t)}{dt} = \nabla \cdot D_i \nabla C_i + K_0 - K_{iv}C_vC_i - k_i^2 D_i C_i \quad (7.18)$$

where

$$K_{iv} = \frac{Z_{iv}\nu(D_i + D_v)}{\lambda^2} \simeq \frac{Z_{iv}\nu D_i}{\lambda^2}$$

and λ is jumping distance and Z_{iv} is the recombination number. For simplicity, we neglect the spatial distribution of defect at this moment. Therefore,

$$\frac{dC_v(t)}{dt} = K_0 - K_{iv}C_vC_i - k_v^2 D_v C_v \quad (7.19)$$

$$\frac{dC_i(t)}{dt} = K_0 - K_{iv}C_vC_i - k_i^2 D_i C_i \quad (7.20)$$

Under steady state, since

$$\frac{dC_i(t)}{dt} = 0 \quad \frac{dC_v(t)}{dt} = 0$$

Eq. 7.20 becomes

$$K_0 - K_{iv}C_vC_i - k_i^2 D_i C_i = 0$$

Therefore,

$$C_i = \frac{K_0}{K_{iv}C_v + k_i^2 D_i}$$

Eq. 7.19 becomes

$$C_v^2 + \frac{k_i^2}{K_{iv}} D_i C_v - \frac{K_0 k_i^2 D_i}{K_{iv} k_v^2 D_v} = 0$$

The solution is

$$C_v = \frac{k_i^2 D_i}{2K_{iv}} (\sqrt{1 + \xi} - 1) \quad (7.21)$$

$$C_i = \frac{k_v^2 D_v}{2K_{iv}} (\sqrt{1 + \xi} - 1) \quad (7.22)$$

where

$$\xi = \frac{4K_0 K_{iv}}{k_i^2 k_v^2 D_i D_v} \quad (7.23)$$

When

$$K_{iv} \gg k_i^2 k_v^2 D_i D_v$$

recombination is dominant,

$$\xi \gg 1 \rightarrow \sqrt{1 + \xi} - 1 \simeq \sqrt{\xi}$$

therefore,

$$C_v \simeq \frac{k_i^2 D_i}{K_{iv}} \sqrt{\xi} = \sqrt{\frac{k_i^2 D_i K_0}{k_v^2 D_v K_{iv}}}$$

$$C_i \simeq \frac{k_v^2 D_v}{K_{iv}} \sqrt{\xi} = \sqrt{\frac{k_v^2 D_v K_0}{k_i^2 D_i K_{iv}}}$$

When

$$K_{iv} \ll k_i^2 k_v^2 D_i D_v$$

sink(annihilation) is dominant,

$$\xi \ll 1 \rightarrow \sqrt{1 + \xi} - 1 \simeq \frac{\xi}{2}$$

therefore,

$$C_v \simeq \frac{k_i^2 D_i}{2K_{iv}} \frac{\xi}{2} = \frac{K_0}{k_v^2 D_v}$$

$$C_i \simeq \frac{k_v^2 D_v}{2K_{iv}} \frac{\xi}{2} = \frac{K_0}{k_i^2 D_i}$$

7.4.2 Transient solutions - Recombination dominant

The transient solution is mainly derived by R. Sizmann. Reference: Sizmann, Rudolf. "The effect of radiation upon diffusion in metals." *Journal of Nuclear Materials* 69 (1978): 386-412.

Usually, at low temperature, recombination is more dominant than sink because, defects have to diffuse to interact with sink sources and as temperature decreases, diffusivity of the defects decreases significantly as well.

Stage 1

At the beginning, point defects are created and not much enough to interacts each other. During the stage, we can evaluate

$$C_v = C_i = K_0 t$$

When recombination is dominant, one can assume that defect generation rate is balanced by recombination rate, that is

$$K_0 = K_{iv}C_iC_v$$

Since

$$C = C_i = C_v$$

$$C = K_{iv}C^2t_1 \rightarrow t_1 = \left(\frac{1}{K_0K_{iv}} \right)^{1/2}$$

After t_1 , since the production and recombination is balanced,

$$C_i = C_v = K_0t_1 = \left(\frac{K_0}{K_{iv}} \right)^{1/2}$$

for a while.

Stage 2

Now, after some time, defects start arriving at the defect sinks. From now on, it corresponds to the sink dominant regime. Since the diffusivity of interstitial is much bigger than that of vacancy, we assume that interstitial arrive at sinks first.

$$\frac{dC_i(t)}{dt} = (-k_i^2D_i)C_i$$

The characteristic time for the process is

$$t_2 = \frac{1}{k_i^2D_i}$$

At $t = t_2$,

$$C_iC_v = \frac{K_0}{K_{iv}}$$

and

$$C_i = C_v$$

Since concentration of interstitial decreases, recombination rate decreases, therefore, vacancy concentration increases at this regime. Therefore,

$$C_v(t) = \left[\frac{K_0(k_i^2D_i)t}{K_{iv}} \right]^{1/2} \quad (7.24)$$

where

$$C_v(t_2) = \left[\frac{K_0}{K_{iv}} \right]^{1/2}$$

Consequently,

$$C_i(t) = \left[\frac{K_0}{K_{iv}(k_i^2D_i)t} \right]^{1/2}$$

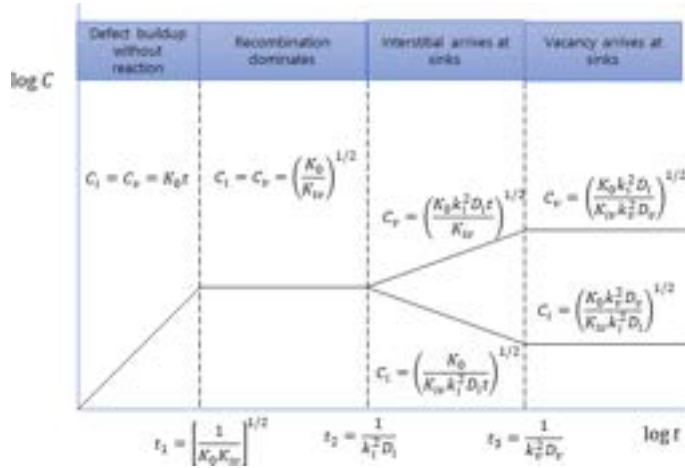


Figure 7.3: Defect concentrations as function of time in a recombination dominated regime.

Stage 3

After for a while, vacancies start arriving at the sinks. The characteristic time is

$$t_3 = \frac{1}{k_v^2 D_v}$$

Put it to Eq. 7.24,

$$C_v = \left[\frac{K_0 k_i^2 D_i}{K_{iv} k_v^2 D_v} \right]^{1/2}$$

Consistently,

$$C_i = \left[\frac{K_0 k_v^2 D_v}{K_{iv} k_i^2 D_i} \right]^{1/2}$$

The defect concentrations in recombination dominated regime is plotted in Fig. 7.3.

7.4.3 Transient solutions - Sink dominant

Stage 1

At the beginning, point defects are created and buildup.

$$C_v = C_i = K_0 t$$

After than, the sink is dominant rather than recombination, the characteristic time for interstitial sink is

$$t_2 = \frac{1}{k_i^2 D_i}$$

come first. Then

$$C_i = \frac{K_0}{k_i^2 D_i}$$

After than, the vacancy start to arrive at sinks and their characteristic time is

$$t_3 = \frac{1}{k_v^2 D_v}$$

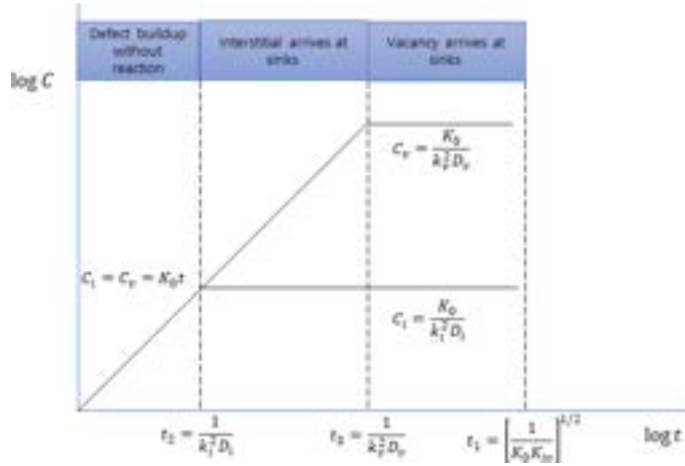


Figure 7.4: Defect concentrations as function of time in a sink dominated regime.

then

$$C_v = \frac{K_0}{k_v^2 D_v}$$

Typical case: $T = 298\text{ K}$ neutron is irradiated to bcc Fe (lattice parameter is 0.28 nm). The dislocation density is 10^8 cm^{-2} . The vacancy and interstitial migration energies are 1.5 eV and 0.65 eV . The recombination number is 200 and displacement rate is 10^{-7} dpa/s . The vibration frequency is 10^{13} s^{-1} . Consider only sink by dislocation and $z_i = 1.02$ and $z_v = 1$. We can evaluate

$$D_i = \frac{8}{6} \times (2.44 \times 10^{-8})^2 \times 10^{13} \times \exp\left(-\frac{0.65}{8.62 \times 10^{-5} \times 298}\right) = 8.134 \times 10^{-14}\text{ cm}^2/\text{s}$$

$$D_v = \frac{8}{6} \times (2.44 \times 10^{-8})^2 \times 10^{13} \times \exp\left(-\frac{1.5}{8.62 \times 10^{-5} \times 298}\right) = 3.464 \times 10^{-28}\text{ cm}^2/\text{s}$$

with jump distance

$$\frac{\sqrt{3}}{2} a_0 = 0.244\text{ nm}$$

The recombination rate constant is

$$K_{iv} = 200 \times 10^{13} \times \exp\left(-\frac{0.65}{8.62 \times 10^{-5} \times 298}\right) = 2.0 \times 10^4$$

Then

$$t_1 = \frac{1}{\sqrt{K_0 K_{iv}}} = 22.4\text{ s}$$

Since

$$k_i^2 = z_d \rho_i = 1.02 \times 10^8$$

therefore,

$$t_2 = \frac{1}{k_i^2 D_i} = \frac{1}{1.02 \times 10^8 \times 8.134 \times 10^{-14}} = 1.21 \times 10^5\text{ s} = 33.5\text{ h}$$

Since

$$k_v^2 = z_v \rho_i = 1 \times 10^8$$

therefore,

$$t_3 = \frac{1}{k_v^2 D_v} = \frac{1}{1 \times 10^8 \times 3.464 \times 10^{-28}} = 2.89 \times 10^{19} \text{ s} = 33.5 \text{ h} = 9.15 \times 10^{11} \text{ years}$$