Radiation-induced material redistribution in concentrated binary alloys

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Abstract

We studied irradiation-induced material redistribution in concentrated binary alloys, AB. When a material is irradiated, Frenkel pairs (vacancies and interstitials) are produced in concentrations much more above their thermal concentrations. Depending upon the crystal structure and size of the alloying elements, various defect configurations are formed. It has been seen that, if the solute size is such that they can fit in the interstices (C, H, Boron, etc), the mobile defect configurations are vacancies and interstitials. But, if the size of the solute is equal to bigger than the solvent atoms, formation of dumb-bell interstitial configurations is observed (FCC, BCC). In present study, we have considered a FCC binary alloy, AB, in which the size of B is almost equal to A. Therefore, we may have vacancies, AA, BB, AB (dumb-bells) as mobile defect configurations. In the presence of defect sinks, the defect move towards them resulting in depletion of faster diffusing alloying species and enrichment of slower diffusing species: Inverse Kirkendall Effect. This causes segregation of alloying elements near defect sinks, which may deteriorate various material properties. In the absence of defect sinks, we can see redistribution of alloying elements as a result of irradiation. Present study is done to create a conceptual framework for the actual problem: Radiation-induced microstructural and microchemical changes in concentrated binary alloys.

Introduction

In the present system we have 6 species: A, B atoms, vacancies, $AA,\ BB,\ AB$ dumb-bell interstitials. The material transport takes place by two diffusion mechanisms: Vacancy and interstitialcy mechanisms. In vacancy diffusion, an atom , hops on the lattice if a vacancy is present in its nearest neighbour. Interstitialcy mechanism, on the other hand, takes place when one of the atoms in the dumb-bell moves to a nearest neighbour atom and forms a dumb-bell there:

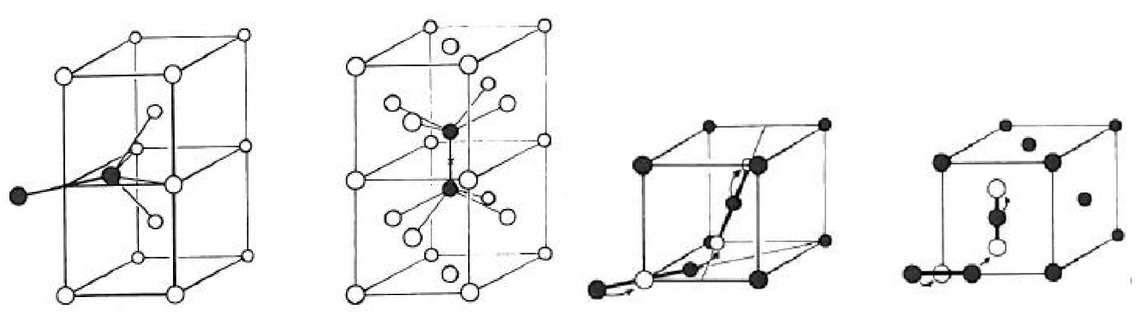


Figure 1: Dumb-bell interstitials in BCC(left most), FCC(2^{nd}), Jump of dumb-bells, BCC(3^{rd}) and FCC(4^{th})

In order to study redistribution of species in the alloy, correct kinetics has to be understood. This requirement necessitates the derivation of flux for each species in the system using vacancy and interstitialcy mechanism. Once kinetics of each species is established, we will be able to write reaction-diffusion equation, in the absence of sinks as:

$$\frac{\partial C_j}{\partial t} = -\nabla \cdot J_j + \text{Reactions} + \text{Source} + \text{Recombination}$$

where, $J_j = -\sum_{k=1}^{n} D_{jk} \nabla C_k$.

Theory

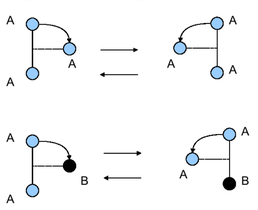
The flux of all the species consist of following:

- Vacancies: Vacancy flux due to exchange of vacancies with either A or B atoms by vacancy mechanism is:

$$\begin{aligned} J_v &= -(J_A^v + J_B^v) = (D_{Av} C_v \nabla C_A - D_{Av} C_A \nabla C_v + D_{Bv} C_v \nabla C_B - D_{Bv} C_B \nabla C_v) \\ &= [(D_{Av} \nabla C_A + D_{Bv} \nabla C_B) C_v - (D_{Av} C_A + D_{Bv} C_B) \nabla C_v] \end{aligned}$$

  where, $D_{\xi v} = d^2\, \nu^0_{\xi v}\, exp(-E^m_{\xi v}/kT), \xi = A,\ B$; d = distance between two lattice sites, ν^0 = vibration factor, E^m = migration energy.

- Dumb-bell interstitials: Flux of dumb-bells due to Interstitialcy mechanism is calculated by considering following transitions (e.g. for AA dumb-bell):



$$J_{AA} = -D_{AA}(C_A \nabla C_{AA} - C_{AA} \nabla C_A) - \frac{1}{2} D(AA \to AB)(C_B \nabla C_{AA} - C_{AA} \nabla C_B) - \frac{1}{2} D(AB \to AA)(C_A \nabla C_{AB} - C_{AB} \nabla C_A)$$

  Similarly, fluxes of BB and AB dumb-bells can be obtained.

- A and B atom flux: Flux of alloying elements will depend on both vacancy and interstitialcy mechanisms:

$$J_A = J_A^v + J_A^I = \underbrace{J_A^v}_{vac.mech.} - \underbrace{[J_{AA} + J_{AB}^B]}_{interstitialcy}$$
$$J_B = J_B^v + J_B^I = \underbrace{J_B^v}_{vac.mech.} - \underbrace{[J_{BB} + J_{AB}^A]}_{interstitialcy}$$

  J_{AB}^B = flux of AB dumb-bell when B atom jumps leaving behind A atom on the lattice;
  J_{AB}^A = flux of AB dumb-bell when A atom jumps leaving behind B atom on the lattice.

Similarly, we can derive reactions which bring about the change in the concentration of all the species locally(flux-less). For example, the change in AA dumb-bell concentration can be brought about by the following reactions:

$$\text{Reactions}_{AA} = -K_2(AA \to AB) C_{AA} C_B + K_2(AB \to AA) C_A C_{AB}$$

where, $K_2(AA \to AB)$ = reaction rate coeffecient = $f_2 \nu^0_{AA} exp(-E^c(AA \to AB)/kT)$, f_2 = geometric factor, $E^c(AA \to AB)$ = conversion energy from AA to AB dumb-bell.

Model

We have 6 reaction-diffusion equations to solve to see the evolution of each concentration field under irradiation. A typical reaction-diffusion equation has the following form:

$$\frac{\partial C_j}{\partial t} = -\nabla \cdot J_j + \text{Reactions} + \text{Source} + \text{Recombination}$$

where, $J_j (j = A,\ B,\ V,\ AA,\ BB,\ AB)$ is defined as above.
For example, the evolution of AA dumb-bell interstitial is given by following equation:

$$\begin{aligned} \frac{\partial C_{AA}}{\partial t} &= \nabla \cdot \Big[D_{AA}(C_A \nabla C_{AA} - C_{AA} \nabla C_A) + \frac{1}{2} D(AA \to AB)(C_B \nabla C_{AA} - C_{AA} \nabla C_B) \\ &+ \frac{1}{2} D(AB \to AA)(C_A \nabla C_{AB} - C_{AB} \nabla C_A) \Big] - \Big[K_{1AA} C_A + K_1(AA \to AB) C_B \\ &+ K_{1vA} C_A + K_{1vB} C_B \Big] C_{AA} C_v - K_2(AA \to AB) C_{AA} C_B + K_2(AB \to AA) C_{AB} C_A \\ &+ \text{Source} \end{aligned}$$

We have applied source term using a core-shell model. It is applied at random in space but at a known time:

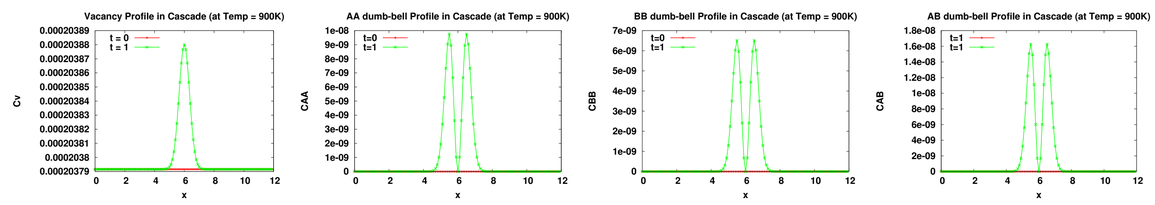


Figure 2: Cascade line profile: Vacancy(left most), AA interstitial (2^{nd}), BB interstitial (3^{rd}) and AB interstitial(4^{th})

Non-dimensionalization

To avoid dealing with intrinsic length and time scales, the reaction-diffusion equations have been non-dimensionalized by defining $x \to l\tilde{x} \Rightarrow \frac{\partial}{\partial x} \to \frac{1}{l}\frac{\partial}{\partial \tilde{x}}$ and $t \to \tau \tilde{t}$, $\tau = \frac{1}{K_{max}}$ and $l = \sqrt{\frac{D_{max}}{K_{max}}}$, where, K is reaction rate constant and D is the diffusivity.

Numerical Scheme

We solve the equations simultaneously in 2D using periodic boundary conditions. Keeping in mind the CFL condition $\left(\frac{\Delta t}{(\Delta x)^2} < C\right)$, we used Trapezoidal Method (improved Euler, $O(2)$) for marching along time and centered finite difference ($O(2)$) for approximating ∇^2.

Results

A single cascade is applied at the center of the domain. We see the evolution of various fields:
The diffusivities in the model are related to each other as:
$D_{AA} = D(AA \to AB) > D_{BB} = D(BB \to AB) > D_{AB} > D(AB \to BB) > D_{Av} > D_{Bv}$. Evolution of all fields at temperature $900K$ is given below:

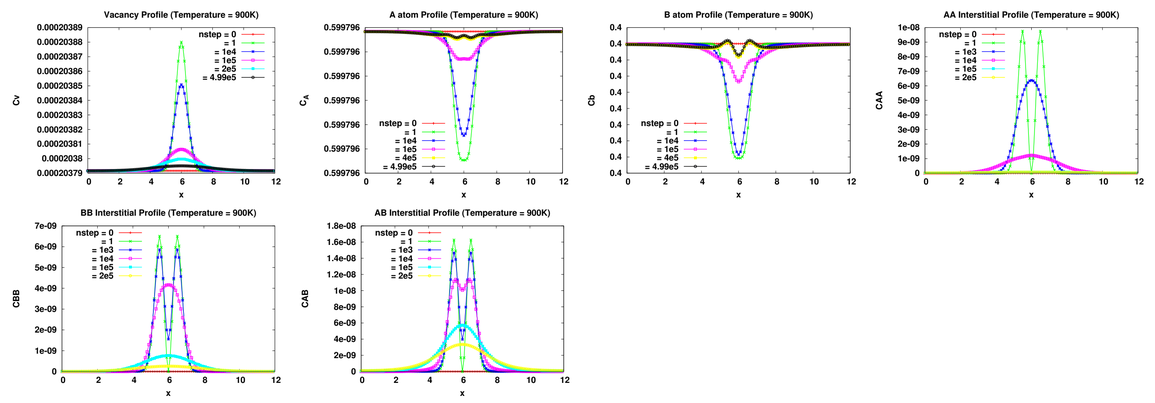


Figure 3: Vacancy(left most), A (2^{nd}), B (3^{rd}), AA interstitial(4^{th}), BB interstitial (bottom left), AB interstitial (bottom right)

Future Scope

Upto now we do not have microstructure in the model, which are the most important features in a material. We will do this in Phase field formalism. Following are the salient features of Phase field formalism:

- It is a material modeling approach at continuum scale using two types of field variables: conserved and non-conserved.
- It is based on the idea of Diffuse Interface(finite thickness, smooth variation of fields, no explicit tracking of interface).
- It involves simultaneous solution of Cahn-Hilliard and Allen-Cahn equations coupled by the free energy of system:

$$\frac{\partial C_i(r,t)}{\partial t} = \nabla \cdot \sum_j L_{ij} \nabla \left(\frac{\delta F}{\delta C_j(r,t)} \right) \quad \text{Cahn-Hilliard}$$
$$\frac{\partial \eta_p(r,t)}{\partial t} = -L_{pq} \frac{\delta F}{\delta \eta_q(r,t)} \quad \text{Allen-Cahn}$$

To get L (chemical mobility), we will use Onsager formulation, which relates fluxes of a species to different driving forces present in the system. In crystalline solids,

$$J_i = -\sum_{j=1}^{n} L_{ij} \nabla \mu_j = -\sum_{k=1}^{n} \sum_{j=1}^{n} L_{ij} \frac{\partial \mu_j}{\partial C_k} \nabla C_k$$

Comparing with generalized Fick's law, $J_i = -\sum_{k=1}^{n} D_{ik} \nabla C_k$, we get: $D_{ik} = \sum_j L_{ij} \frac{\partial \mu_j}{\partial C_k} \Rightarrow$ We need to solve $\mathbb{D} = \mathbb{L}\mathbb{Q}$, $\mathbb{Q} = \frac{\partial^2 F}{\partial C_j \partial C_k}$ to get $\mathbb{L}$.