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Numerical Coupling Radiation Induced Segregation and Compositional–Microstructural Evolution

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Abstract

A hybrid model of microstructural evolution of a coupled multi–field system that is subjected to ion irradiation is presented. Materials exposed to low energy ion irradiation experience a wide range of radiation effects, e.g. surface roughening and chemical segregation. The hybrid model combines Monte Carlo methods and a phase field model to simulate the kinetic and radiation−induced processes that lead to radiation induced chemical segregation with associated phase transformations of a binary system by differential diffusivity.

1 Introduction

Radiation events lead to the creation of over–saturated point defects, causing unique microstructural changes including radiation–induced segregation (RIS). It is well known that ion–irradiation can lead to chemical separation in a multi–component system, which is essentially an effect driven by the difference in diffusivities of interstitials and vacancies of the different components. This radiation effect has been observed in a wide variety of materials including binary [1] and multi–component [2] systems. Some of these materials also show interesting microstructural features forming like nano–cones [3] and nano–fibrous structures [4].

2 Model Development

A binary (AB) system with thermodynamic characteristics of $GaAs$ is studied. However, since diffusion of defects in $GaAs$ is inherently complex leading to disagreement on diffusion data interpretation[5], we do not claim to simulate $GaAs$. A hybrid model that couples continuum and statistical methods is used to simulate the generation of defects due to irradiation, diffusion of components A and B, phase transformation and microstructural evolution. The model is based on the hybrid Potts–phase field model developed by *Homer et al.* [6], but is further developed to simulate radiation damage and its effects on diffusion.

2.1 Microstructural and Composition Representation

We use continuous fields to represent the chemical composition and vacancy density, and discrete fields to represent the phases. The simulation space is digitized into 2D squares, called sites, where the discrete field in each square describes its phase state and the continuous fields have digitial values at that grid point. The starting microstructure consists of an AB ($GaAs$) solid in contact with vacuum, called void. The concentration of vacancies is zero (as the equilibrium vacancies is very low) and the chemical composition is $C_A = C_B = 0.5$ at all sites. Periodic boundary conditions are used in the x-direction only, hence, in effect, the solid is a semi–infinite solid with an irradiated surface in contact with a vacuum.

2.2 Radiation Damage

Radiation damage is accounted for in the form of vacancy defects. The calculation of the radiation damage is a three step process. These are, if a collision event takes place: (i) determine the amount of energy deposited in the pixel; (ii) calculate how many defects are created by the collision cascade corresponding to the amount of energy deposited; and (iii) calculate the number of defects that survive the quenching stage. This surviving defect concentration contributes to the microstructural evolution.
A more detailed description of the radiation damage can be found in Hernández–Rivera [7].

2.3 Chemical and Vacancy Evolution

The RIS model developed by *Wiedersich et al.* [1] couples point defect kinetic equations to track the evolution of these defects. In this hybrid model, we model this field by use of an adapted Cahn–Hilliard equation

$$\frac{\partial C_v}{\partial t} = \eta_s(\Delta t) + \nabla \cdot \left(M_{C_v} \nabla \cdot \left[\frac{\partial f_v}{\partial C_v} - \kappa_{C_v} \nabla^2 C_v \right] \right) \tag{1}$$

where $\eta_s(\Delta t)$ is the additional concentration of surviving defects during irradiation time interval Δt, M_{C_v} is the mobility of the vacancies, κ_{C_v} is an interfacial energy constant and f_v is the vacancy bulk free energy defined as by the regular solution equation

$$f_v = \frac{1}{\Omega}\big(E_v^f C_v + k_B T[C_v \ln(C_v) + (1 - C_v)\ln(1 - C_v)]\big) \tag{2}$$

where Ω is the unit site volume and E_v^f is the vacancy formation energy.
For the evolution of the chemical components, we use an approach similar to the one given by *Wiedersich*. The different point defect fluxes through an AB system are

$$\begin{aligned} J_i &= J_i^A + J_i^B \\ -J_v &= J_v^A + J_v^B \end{aligned} \tag{3}$$

where J_i^A is the flux of defect i through the site of A component. Since we are considering a binary system and using Fick's first law, Equation 3 could be simplified for a single component since $C_A + C_B = 1$ ($\nabla C_A = -\nabla C_B$). Now, using Fick's second law and setting the net flux to include diffusion due to chemical gradients and through the defect mechanisms just discussed, we have

$$\frac{\partial C_B}{\partial t} = \nabla \cdot \big(D_B \nabla C_B - D_v^B \nabla C_v + D_i^B \nabla C_i\big) \tag{4}$$

where the first term is the flux due to a chemical gradient, and the second and third terms are fluxes through the vacancy and interstitial defects, respectively. Since only the creation of vacancies is considered, the third term in Equation 4's RHS is not included in our hybrid model. Lastly, we should point out that we assume that the B component diffuses faster than A through the vacancy mechanism, i.e. $D_A^v < D_B^v$.

2.4 Phase Transformations

We use a discrete integer value at each site to describe its phase state, which are: AB, A–rich, B–rich and liquid. These are defined by the free energy curves plotted in Figure 1. When a site's energy has been increased due to a collision event to that at or above another phase's energy, then

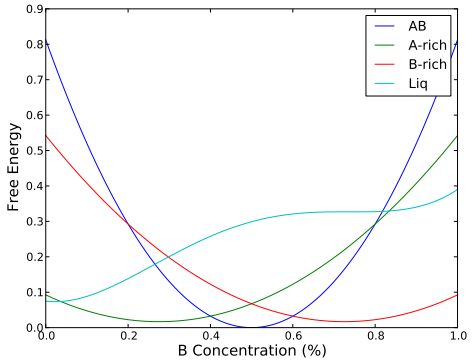

Figure 1: Isothermal chemical free energy curves for the two–component, four−phase system being modeled.

the site's phase is changed to that higher energy phase.
Phase transitions can also occur by each site attempting to nucleate another phase. If the change in energy calculated by Equation 5 lowers the chemical bulk free energy, f_A^{bulk}, the phase transition event occurs. These nuclei can grow and coalesce in a similar process by growing into the neighboring sites again with a transition probability given by Equation 5.

$$P_{\text{phase change}} = \begin{cases} \text{change}, & \Delta f_A^{bulk} \leq 0 \\ \text{no change}, & \Delta f_A^{bulk} > 0 \end{cases} \tag{5}$$

where Δf_A^{bulk} is the difference between the chemical bulk free energy (Figure 1). For a phase nucleation event we compare the difference in the chemical bulk free energies of the different phases.

2.5 Surface Diffusion

In addition to the surface diffusion described in Section 2.3, surface diffusion is also simulated by exchanging mass and void sites at the surface as described in *Holm et al.* [9], and implemented by *Homer*. This leads to roughening of the material surface. The probability that an exchange will happen, is given by

$$P_{\text{surf mig}} = \begin{cases} 1, & \Delta E_{int} \leq 0 \\ \exp\left(-\frac{\Delta E_{int}}{k_B T}\right), & \text{otherwise} \end{cases} \tag{6}$$

where ΔE_{int} is the difference between the interface energy for the i and j sites before and after the exchange, where E_{int} is the Potts interfacial energy[6, 9].

3 Results and Discussion

As the material is irradiated, the vacancy concentration increases through the sample depth as shown in Figure 2(left). It increases at the surface at a faster rate than deeper into the target, eventhough irradiation introduced more vacancies sub–surface than at the surface. This is a clear indication that vacancies are diffusing into the free surface, the only available sink. The vacancy profiles in Figure 2(left) are the average concentrations at that depth. The noise in the plots is due to roughening of the target surface by surface diffusion.

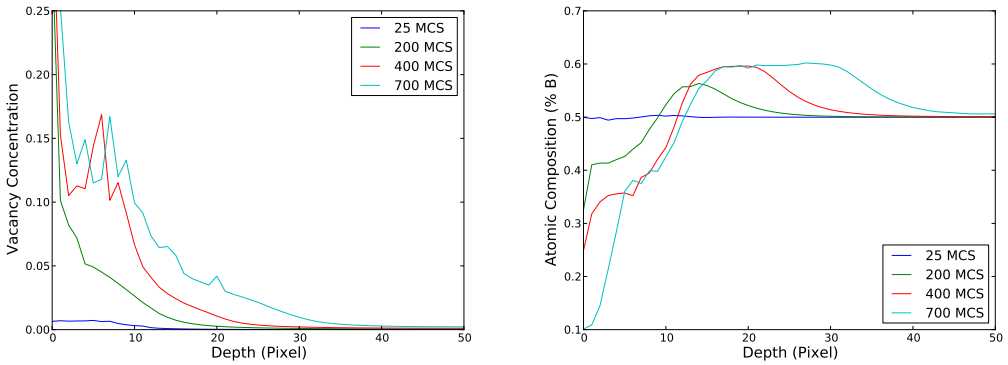

Figure 2: (left) Vacancy concentration profile as it evolves with irradiation time. (right) Chemical composition profile as it evolves with irradiation time

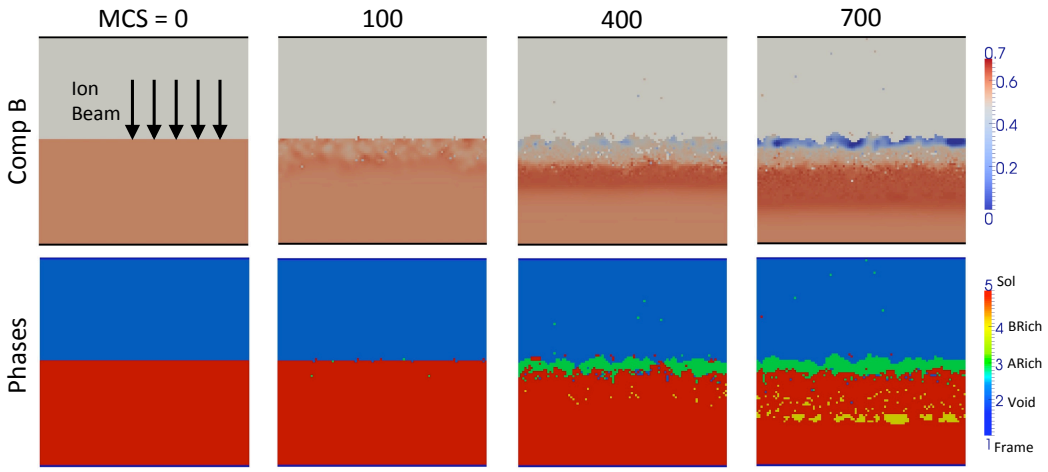

Figure 3: Evolution of the compositional and phase fields as the material is irradiated from the top. The creation and diffusion of the vacancies leads to chemical segregation and phase transformations.

The B–component concentration profiles as a function of target depth z at different irradiation times are plotted in Figure 2(right). The starting concentration of B is $C_B = 0.5$, but is depleted at the surface and enriched at increasing depth with irradiation time. Since the diffusion of B through the vacancy mechanism is faster than that for A (i.e. $D_B^v > D_A^v$), B is depleted near the surface. This is due to the fact that the movement of a vacancy is directly correlated to the movement of an atom in the opposite direction, as evident from Equation 3.

Chemical segregation is evident in Figure 3 which shows the composition and phase evolution. As irradiation proceeds and irradiation damage accumulates, the region closest to the surface starts to become enriched in A while the region just below becomes B–rich. As previously explained, this is because of the difference in diffusivities. Another interesting result is the phase transformation shown in Figure 3. As described in Section 2.4, when the chemical composition deviates considerably from the stoichiometric composition the material undergoes a phase transition. Once sufficient damage has been accumulated to create a stable A– or B–rich phase region, it can start nucleating and coalescing into a larger stable region.

4 Summary

One of the major advantages that this model provides is the coupling between composition and phase fields efficiently. Using the hybrid model we were able to accurately model the chemical segregation of an irradiated binary system. We show that even with the implementation of a single radiation–induced point defect type, the difference between diffusivities for each chemical component is sufficient to lead to segregation. Furthermore, it was shown that our model is able to simulate direct phase transformation due to compositional changes in the microstructure. Lastly, we see that this type of hybrid model that incorporates different processes into the evolution of a microstructure has great potential to simulate a wide range of complicated physics processes.

Statement of Contribution

The work presented is part of the student's dissertation research. Therefore, the student has been the primary contributor to the development of the model, simulation work and writing of this paper. The ideas presented here are the student's, with insightful guideance from Veena.

Acknowledgement

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