Compositional patterning in concentrated binary alloys under irradiation

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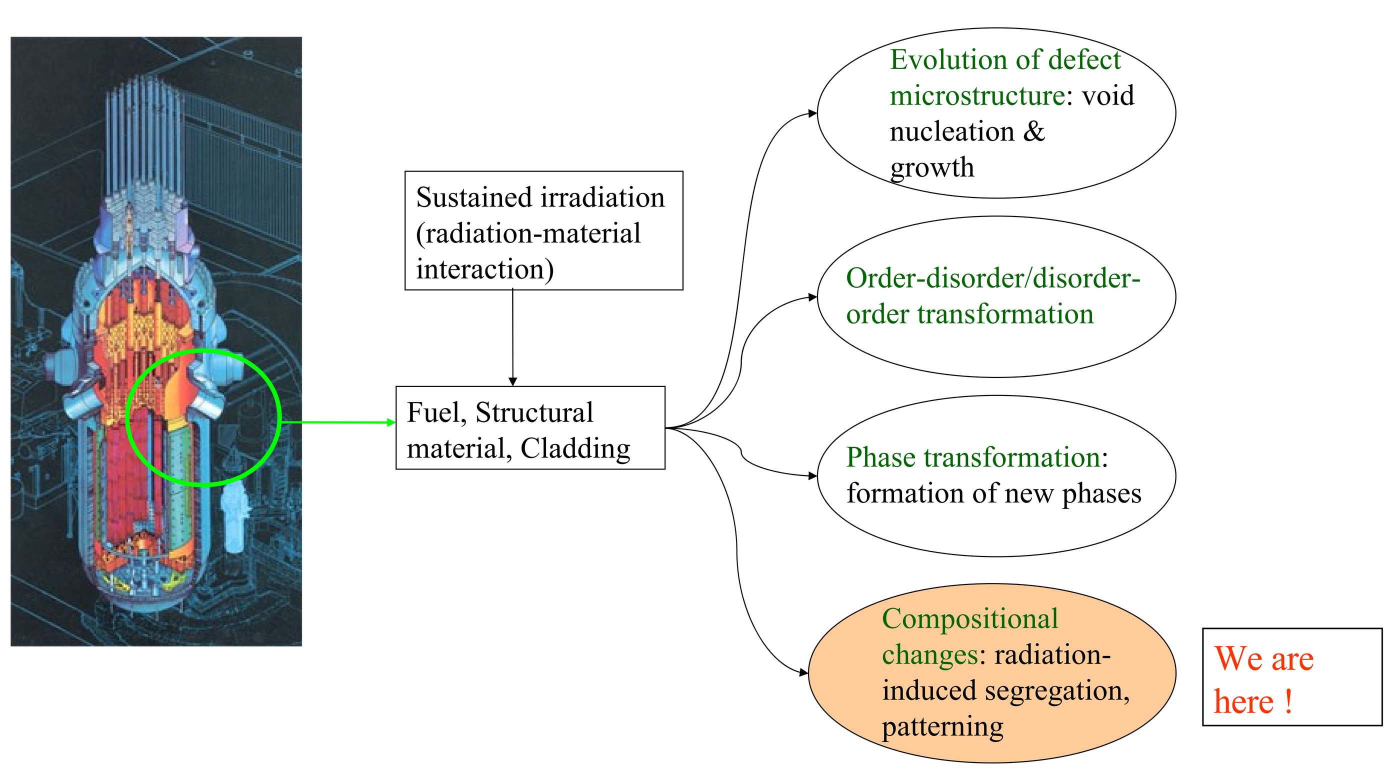
Abstract

A model to study the formation of compositional patterns in concentrated binary alloys under irradiation is presented. There are 6 species in the model: 2 regular lattice atoms (A and B), 3 interstitial dumbbells (AA, BB and AB) and vacancy. Dumbbell interstitials diffuse by interstitialcy mechanism and vacancies diffuse by vacancy mechanism. Diffusion of A and B atoms is coupled to the diffusion of defects. In addition to long range diffusion of the species, local defect-defect (vacancy-interstitial recombination) and defect-atom (dumbbell-atom leading to change in dumbbell type) reactions have also been considered. The model tracks the simultaneous evolution of all the species in reaction-diffusion formalism. The irradiation event is modeled as a stochastic point process which changes the concentration of all the species instantaneously in a random fashion. In each irradiation event, a spatial distribution of point defects (core-shell distribution: core dominated by vacancies and shell by dumbbell interstitials) has been introduced in the system which drives the kinetics of diffusion and reaction. The model has been non-dimensionalized with respect to intrinsic length and time scales and numerically solved using finite difference technique. Using this model in FCC CuAu alloy we have shown that depending upon the irradiation conditions and temperature, system selects specific wavelengths in steady state under sustained irradiation.

Motivation



-Material employed in nuclear reactors undergo changes in their structural and material properties due to various kinds of changes brought about by sustained irradiation.

-Important to predict these changes beforehand to ensure safe functioning of nuclear reactors.

-Need realistic models which could deal with in-reactor complexities of various physical processes.

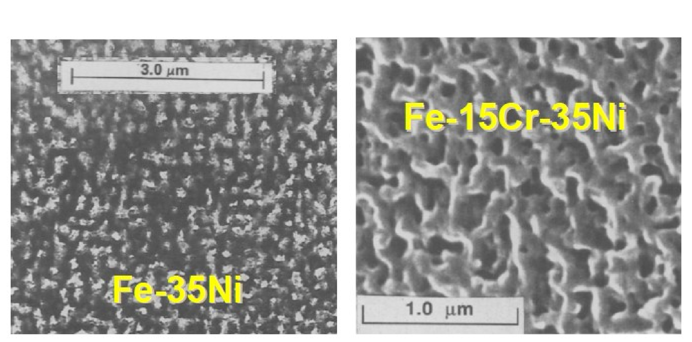
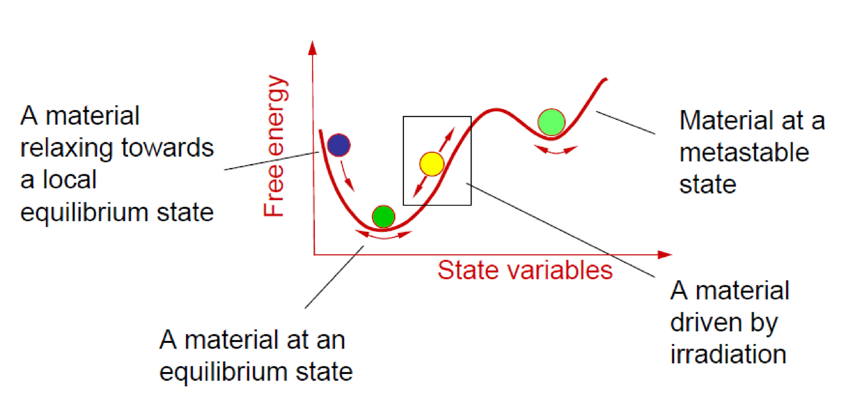


Figure 1: Compositional self-organization/patterning in invar type alloys

Recipe for irradiation studies

-A material under irradiation is far away from equilibrium. It may decay to a lower energy metastable state but not equilibrium.

-This is one of the fundamental reason behind irradiation-induced changes in the material.



Model

-We consider a concentrated, random solid solution AB in which irradiation produced interstitials form dumbbells.

-There are 6 species in the model: regular lattice atoms (A, B), dumbbell interstitials (AA,BB,AB) and vacancies.

-Diffusion of species by vacancy and interstitialcy mechanism.

-Solid solution is considered isotropic and correlation between defects jumps is neglected.

-Irradiation event is stochastic in space, time and strength. Each event introduces a spatial distribution of point defects in the material in core-shell configuration: core is dominated by vacancies and shell by interstitials.

-Irradiation produced defects drive the kinetics of diffusion and reaction of all the species, which result in macroscopic changes in material properties.

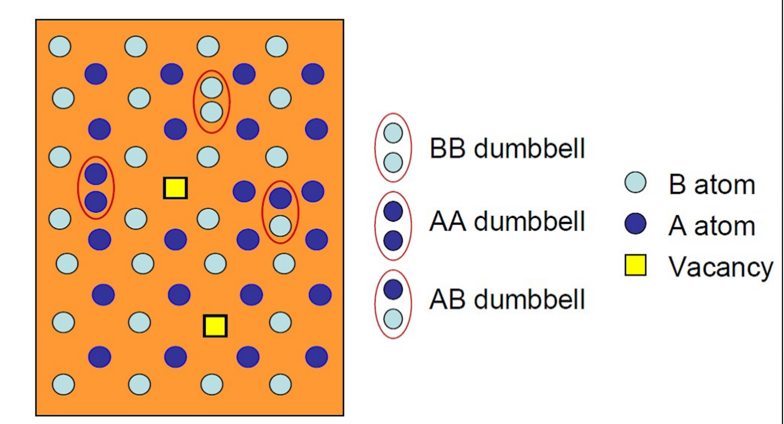


Figure 2: Schematic showing all the species in the model

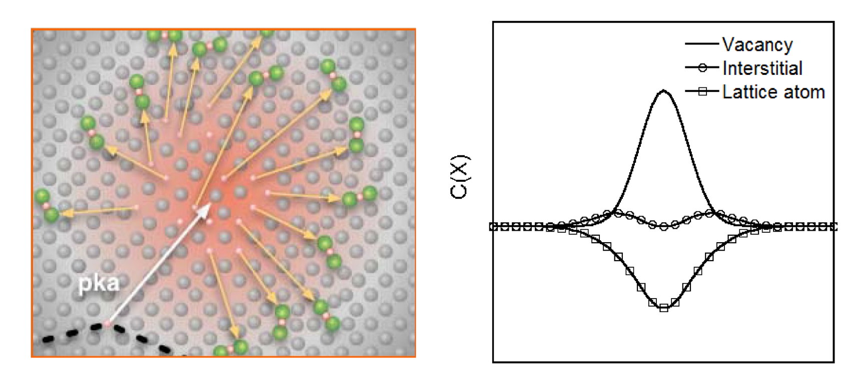


Figure 3: Schematic showing production (left) and spatial distribution of point defects in an irradiation event (right) in core-shell configuration.

Defect production model:

-Vacancy profile

$$\Delta C_v^{casc}(r) = C_v^{amp}\ exp\Big[-\Big(\frac{r-r_c}{\alpha_v}\Big)^2\Big]$$

-Interstitial profile

$$C_I^A(r) = C_v^{amp}\frac{1}{\pi\alpha_i^4}\Big\{\int_{\Omega_{cascade}} C_A(r)exp\Big[-\Big(\frac{r-r_c}{\alpha_v}\Big)^2\Big]d\Omega_{cascade}\Big\}(r-r_c)^2 exp\Big[-\Big(\frac{r-r_c}{\alpha_i}\Big)^2\Big]$$

$$C_I^B(r) = C_v^{amp}\frac{1}{\pi\alpha_i^4}\Big\{\int_{\Omega_{cascade}} C_B(r)exp\Big[-\Big(\frac{r-r_c}{\alpha_v}\Big)^2\Big]d\Omega_{cascade}\Big\}(r-r_c)^2 exp\Big[-\Big(\frac{r-r_c}{\alpha_i}\Big)^2\Big]$$

-Profile for rest of the species

$$\begin{aligned}
\Delta C_{AA}^{casc}(r) &= C_I^A(r)C_A^{post-casc}(r)\\
\Delta C_{BB}^{casc}(r) &= C_I^B(r)C_B^{post-casc}(r)\\
\Delta C_{AB}^{casc}(r) &= C_I^A(r)C_B^{post-casc}(r)\\
&\quad +C_I^B(r)C_A^{post-casc}(r)\\
\Delta C_{A}^{casc}(r) &= -C_A(r)\Delta C_v^{casc}(r) - \Delta C_{AA}^{casc}(r)\\
&\quad -C_I^B(r)C_A^{post-casc}(r)\\
\Delta C_{B}^{casc}(r) &= -C_B(r)\Delta C_v^{casc}(r) - \Delta C_{BB}^{casc}(r)\\
&\quad -C_I^A(r)C_B^{post-casc}(r)
\end{aligned}$$

where

α_v Full width at half maximum for vacancy profile

C_v^{amp} Amplitude of vacancy profile

α_i Full width at half maximum for interstitial profile

$$C_A^{post-casc}(r) = C_A(r)[1-C_v^{casc}(r)]$$
$$C_B^{post-casc}(r) = C_B(r)[1-C_v^{casc}(r)]$$

After each irradiation event, the concentration of all the species change instantaneously by the above amount.

Balance laws: diffusion and reaction processes

-All the concentrations evolve by simultaneous action of diffusion, reaction and irradiation damage.

-This leads to a set of coupled reaction-diffusion equations in all the concentration variables.

-Defects diffuse by vacancy and interstitialcy mechanism

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-Two types of reactions considered: vacancy-interstitial recombination and change in dumbbell type

$$\frac{\partial C_j}{\partial t} = \underbrace{-\nabla\cdot J_j}_{\text{Diffusion}} + \overbrace{R_j}^{\text{Reaction}} + \underbrace{S_j}_{\text{Irradiation damage}} \qquad j = A,B,AA,BB,AB,V$$

$$\begin{aligned}
\frac{\partial C_{AA}}{\partial t} &= -\nabla\cdot(\Omega J_{A^I}(AA)) - R_{AA}^v + \mathcal{R}_{AA} + \Delta C_{AA}^{casc}\\
\frac{\partial C_{BB}}{\partial t} &= -\nabla\cdot(\Omega J_{B^I}(BB)) - R_{BB}^v + \mathcal{R}_{BB} + \Delta C_{BB}^{casc}\\
\frac{\partial C_{AB}}{\partial t} &= -\nabla\cdot(\Omega[J_{A^I}(AB) + J_{B^I}(AB)]) - R_{AB}^v + \mathcal{R}_{AB}^{A^I} + \mathcal{R}_{AB}^{B^I} + \Delta C_{AB}^{casc}\\
\frac{\partial C_v}{\partial t} &= -\nabla\cdot(\Omega J_v) - R_{AA}^v - R_{BB}^v - R_{AB}^v + \Delta C_v^{casc}\\
\frac{\partial C_A}{\partial t} &= -\nabla\cdot(\Omega J_A) + 2R_{AA}^v + R_{AB}^v + \mathcal{R}_A + \Delta C_A^{casc}\\
\frac{\partial C_B}{\partial t} &= -\nabla\cdot(\Omega J_B) + 2R_{BB}^v + R_{AB}^v + \mathcal{R}_B + \Delta C_B^{casc}
\end{aligned}$$

Numerical solution

All the equations have been non-dimensionalized with respect to intrinsic time and length scales of the material. The non-dimensionalized equations have been solved using Finite difference technique; the spatial differential operators are approximated using centered difference and time integration is done with explicit Euler method.

Results

-Model is used to study compositional patterning in CuAu alloy. Cu has faster kinetics than Au.

-Our simulations are valid for solid solution region of the phase diagram.

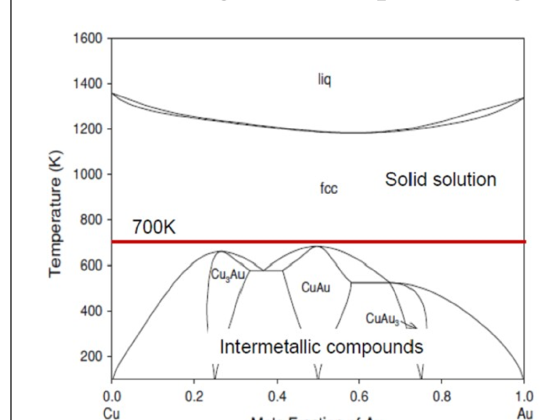

Figure 4: Phase diagram of CuAu

Simulation conditions:

Grid resolution (Δx, Δy)	4.0
Time step size (Δt)	0.3
Number of grid points	250
Initial Cu, Au concentration	0.5
Amplitude of vacancy distribution, C_v^{Amp}	0.2
Full width at half maximum of core distribution, α_v	2 Δx
Full width at half maximum of shell distribution, α_i	6 Δx

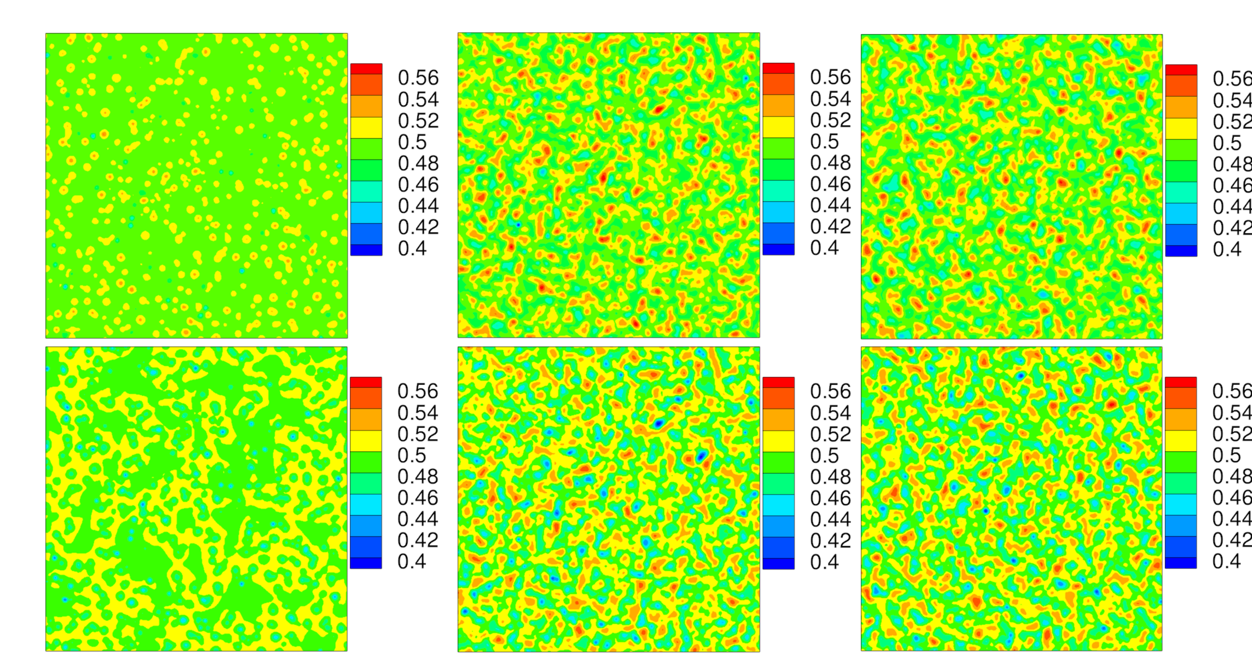

Figure 5: Snapshots of evolution in Cu (top) and Au (bottom) concentration after 2 x 10^5 (left), 2.9 x 10^6 (center) and 5.7 x 10^6 (right) time steps.

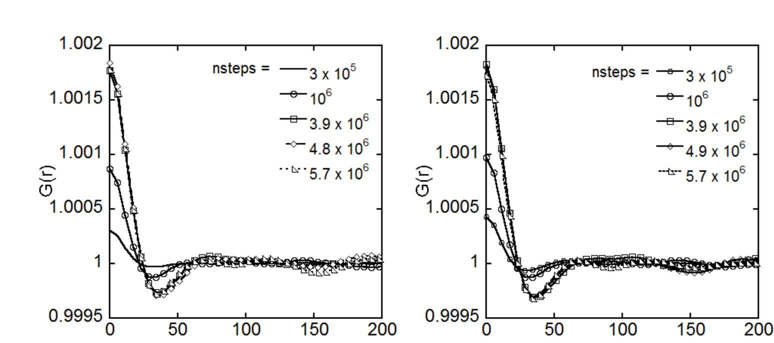

Figure 6: Evolution of radial pair correlation for Cu (left) and Au (right)

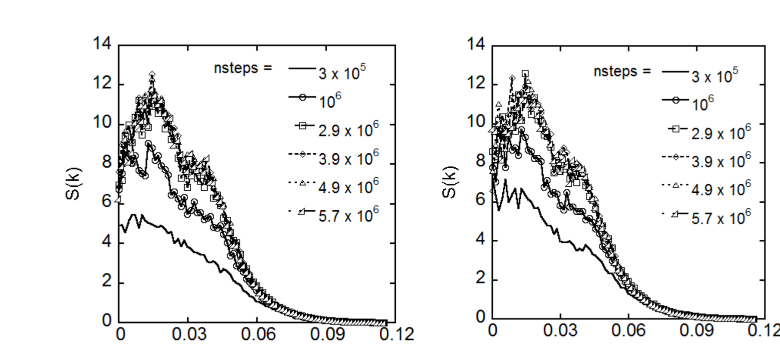

Figure 7: Evolution of structure factor for Cu (left) and Au (right).

Conclusion and future work

-The results show that even though the size and spatial distribution of patterns do not attain steady state, their statistical properties do attain steady state. This is clear from the radial pair correlation and structure factor evolution profiles.

-Currently, this model is being used to study segregation and morphological evolution at free surfaces.