Phase Field Modeling of Microstructure in Irradiated Materials

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Abstract

Reactor materials in nuclear power systems are subject to neutron irradiation conditions which results in intense point defect generation by virtue of atomic collision cascades. The diffusion and clustering of these point defects lead to a variety of microstructure evolution processes which can severely affect their in-service mechanical performance. Here we present a *phase field* framework for modeling microstructure of irradiated material. The model presented here focuses on the void microstructure evolution in irradiated materials, a technologically important problem which is responsible for void swelling of reactor materials. The traditional approach for modeling of void swelling considers the void nucleation and growth stages separately using point kinetic models which treat void nucleation and growth as uniform processes in space. In contrast, our phase field approach treats processes of void nucleation and growth simultaneously in a spatially resolved fashion. The defect fluxes and the defect density modulations are formulated using *Cahn-Hilliard* type description for the vacancy and interstitial concentration fields. The void growth dynamics is obtained in terms of the evolution of a non-conserved order parameter field, whose evolution is prescribed by a phenomenological *Allen-Cahn* type equation. The model also accounts for the effect of applied stress, cascade-induced and thermally-induced fluctuations, vacancy-interstitial recombination, and interaction of vacancies and interstitials with lattice sinks. Illustrative results of model capabilities are presented using the case of a pure metal as an example.

Introduction

- High energy particles (neutrons, ions, electrons or gamma rays) collide with lattice atoms thereby knocking them out of their lattice sites. The displaced atom leaves behind a vacant site (or *vacancy*) and eventually comes to rest in a location which is in between lattice sites, in the form of an *interstitial* atom.
- Under irradiation, cascade of point defects (both vacancy and interstitial) are created in an uncorrelated fashion in the material. These defects diffuse and interact over various timescales to yield interesting microstructural changes

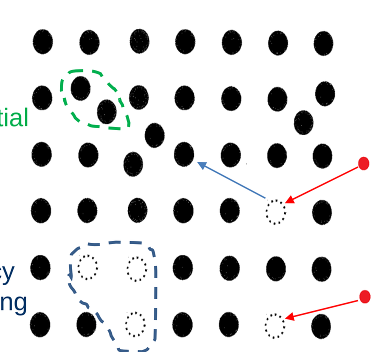

Figure 1: Schematic of radiation damage

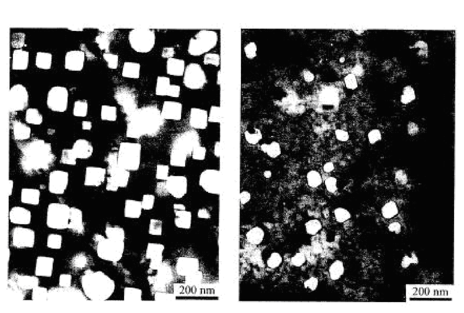

Figure 2: Micrographs of irradiation-induced voids in magnesium [Adda, 1972]

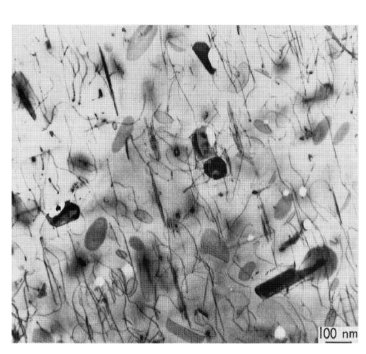

Figure 3: Dislocation loops and voids in a transmission electron micrograph (TEM) of a 300 series stainless steel irradiated at 500°C to a dose of 10 dpa [Mansur, 1994]

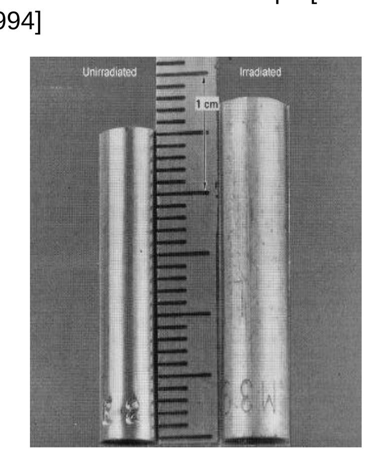

Figure 4: Photograph of 20% cold-worked 316 stainless steel rods before (left) and after (right) irradiation at 533°C to a fluence of 1.5×1023 neutrons m-2 in the EBR-11 reactor [Mansur, 1994]

Radiation effects in materials:

- Dimensional changes (~10%)
  - – Void formation and swelling
  - – Irradiation Growth (shape changes)
- Phase Instabilities
  Amorphization, disordering of ordered precipitates, precipitate nucleation, radiation induced segregation & crystal structural transformation.
- Mechanical Effects
  Increase in yield strength, decrease in ductility, reduction in strain hardening

Multiscale Paradigm of Phenomena

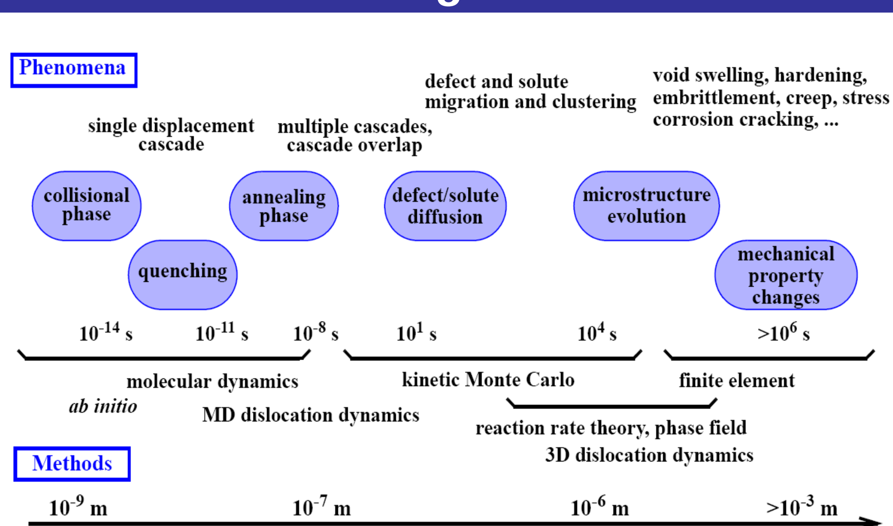

Figure 5: Schematic of the multiscale phenomena and the relevant methods [Stoller]

Phase Field Method for Microstructure Evolution

- *Microstructure:* spatial distribution of structural features (Phases of different compositions, Grains of different orientations, domains of different polarizations etc.)
- Phase field approach describes the microstructure using one of more auxiliary field variables (called *phase fields*)
- Phase fields are *continuous field* variables which are functions of mesoscale space and time.
- The phase field variables evolve with time their evolution is governed by a set of kinetic equations which are coupled in space and time, driven by the reduction of the free-energy of the system.

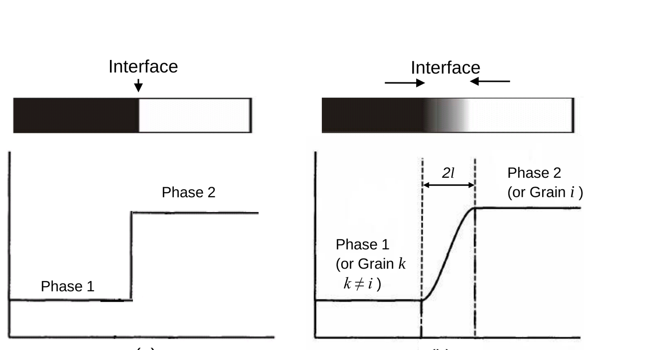

Figure 6: Illustration of interface between two phases (or domains): (a) sharp interface description and (b) diffuse-interface between two different phases or grains

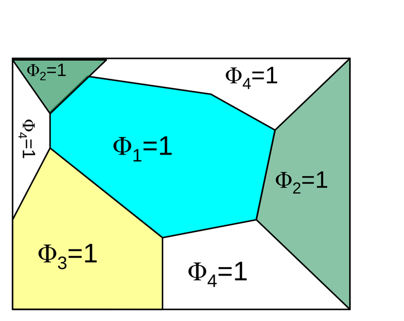

Figure 7: Schematic of microstructure showing phase field variable (η) corresponding to different domains

Phase field model for Void Formation

A diffuse-interface phase field model is developed to describe the evolution of void microstructure in metals under the effect of irradiation. The main features of the model discussed herein are :

- Void is treated as a cluster of vacancies. Void phases are obtained by precipitation of vacancies in a supersaturated system
- In order to distinguish, solid from void regions we use a long-range order parameter (η).
- Vacancies (characterized by their concentration field) are assumed to be the only contributing defect. Effect of interstitial is ignored for the present investigation (is a part of ongoing research by authors).
- Current model is tested for the case of single crystals of one component system (i.e., pure metals). Study of polycrystalline metals and alloys (i.e., two component systems) is a subject of ongoing research by the authors.

Free Energy Functional

To predict the phenomenological evolution of microstructure we describe the solid using its *Helmholtz free energy* functional. Helmholtz free energy for a crystalline solid comprising vacancies, voids and regular solid regions as a function of vacancy concentration $c_v(x,t)$ and artificial phase field $\eta(x,t)$ is obtained as:

$$F(c_v,\eta) = \int_V \left[h(\eta)\left(f_o^m(c_v)\right) + w(c_v,\eta) + \kappa_v \left|\nabla c_v\right|^2 + \kappa_\eta \left|\nabla \eta\right|^2 + F^{elastic} \right] dV$$

Free energy density due to point defects
F^{bulk} Bulk free energy of a material volume
Landau energy term causing bi-stability in the system
$F^{interfacial}$ gradient energy due inhomogeneities in the field (interfaces)
$F^{elastic}$ elastic strain energy (due to defects and any long-range interactions between defects)

Here, the local free energy density of the system is derived in terms of the enthalpic and entropic contributions of point defects:

$$f_o^m(c_v) = \left(E_v^f c_v + k_B T\left(c_v \ln(c_v) + (1-c_v)\ln(1-c_v)\right)\right)$$

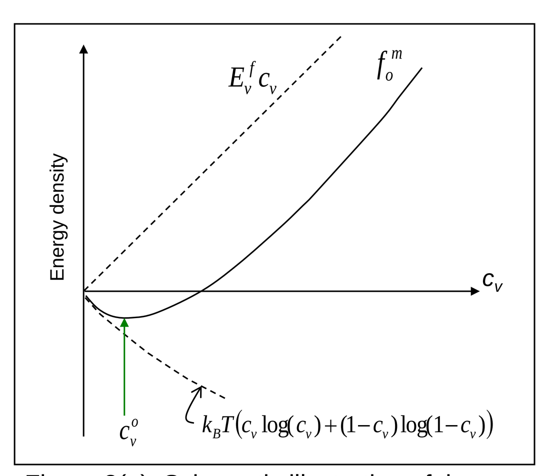

Figure 8(a): Schematic illustration of the terms in the expression free energy density f_o^m

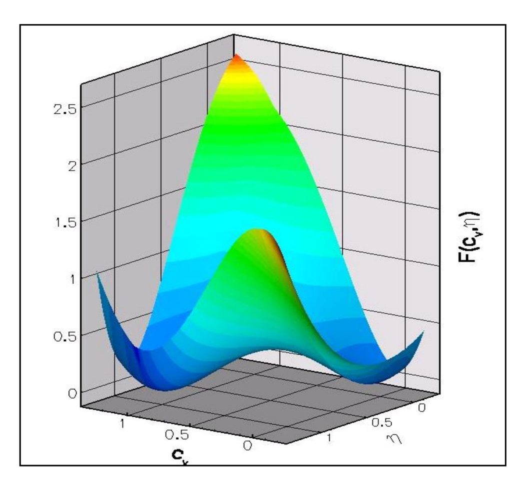

Figure 8(b): Free energy landscape of bulk energy terms F^{bulk}

Two stable wells:

$\eta = 0$, $c_v = c_v^o$ (Solid/matrix phase)

$\eta = 1$, $c_v = 1$ (Void phase)

Kinetic Evolution Equations

Following the standard procedure in phase field approach, the kinetic equations for the space and time evolution of the phase field variables can be obtained as below:

$$\frac{\partial c_v}{\partial t} = \nabla \cdot M_v \nabla \frac{1}{N}\frac{\delta F}{\delta c_v} + \psi_v(\mathbf{x},t) + \xi_v(\mathbf{x},t)$$

Generalized diffusion equation (modified Cahn-Hilliard)

$$\frac{\partial \eta}{\partial t} = -L\frac{\delta F}{\delta \eta} + \xi_\eta(\mathbf{x},t)$$

Allen-Cahn equation (time dependent Ginzburg-Landau)

Here,

$\xi_v(\mathbf{x},t), \xi_\eta(\mathbf{x},t)$ - stochastic terms accounting for thermal fluctuations

$\psi_v(\mathbf{x},t)$ - amount of vacancies entering the system due to radiation damage is given by, $\psi_v(\mathbf{x},t) = G(\mathbf{x},t) - Rc_v c_i - k_i^2 D_v(t) c_v(t)$

Non-dimensionalized Evolution Equations

Substituting the variational derivatives, ignoring elastic effects , and under the assumption of mobility, $M_v = D_v/k_B T$ we get the temporal evolution equations below:

Using transformation: $t \to \tau\,\tilde{t}$ $\quad \mathbf{x} \to l\,\tilde{\mathbf{x}}$ $\quad \nabla = \frac{1}{l}\tilde{\nabla}$

Vacancy field evolution (Cahn-Hilliard equation)

$$\frac{\partial c_v}{\partial \tilde{t}} = \tilde{\nabla}^2 \left[h(\eta)\left(\tilde{E}_v^f + \ln\left(\frac{c_v}{1-c_v}\right)\right) + \frac{\partial \tilde{w}(c_v,\eta)}{\partial c_v} - 2\left(\frac{\kappa_v}{\kappa_\eta}\right)\tilde{\nabla}^2 c_v \right] + \psi_v(\tilde{\mathbf{x}},\tilde{t})$$

Evolution of non-conserved phase field (Allen-Cahn equation)

$$\frac{\partial \eta}{\partial \tilde{t}} = -\tilde{L}\left[h'(\eta)\tilde{f}_o^m(c_v,\eta) + \frac{\partial \tilde{w}}{\partial \eta} - 2\tilde{\nabla}^2\eta \right]$$

With length scale $l = \sqrt{\kappa_\eta / k_B T}$ and time scale $\tau = l^2/D_v$.

$\tilde{E}_v^f = E_v^f/k_B T$, $\tilde{f}_o^m = \tilde{f}_o^m / k_B T$, $\tilde{w} = w/k_B T$ - normalized (dimensionless) energy terms

Numerical Discretization of the Evolution Equations

- Coupled PDEs of the evolution equations are solved using finite differences on a 2D domain with periodic boundary conditions.
- We use explicit finite differences in space and forward Euler time marching scheme.

Concentration field

$$c_v\big|_{i,j}^{n+1} = c_v\big|_{i,j}^{n} + \frac{\Delta t}{(\Delta x)^2}\left(f_c\big|_{i+1,j}^{n} + f_c\big|_{i-1,j}^{n} + f_c\big|_{i,j+1}^{n} + f_c\big|_{i,j-1}^{n} - 4 f_c\big|_{i,j}^{n} \right) + \psi_v(\tilde{x},\tilde{t})$$

Void phase field

$$\eta\big|_{i,j}^{n+1} = \eta\big|_{i,j}^{n} - \Delta t\,\tilde{L}\left(f_\eta\big|_{i,j}^{n}\right)$$

Where $f_c\big|_{i,j}^{n}$ and $f_\eta\big|_{i,j}^{n}$ are functional derivatives evaluated at time step *n* and point (x_i, y_j)

Results and Discussion

Numerical simulations are performed on a 2D square grid with periodic boundary conditions under the assumption of constant temperature.

Void growth and shrinkage dynamics

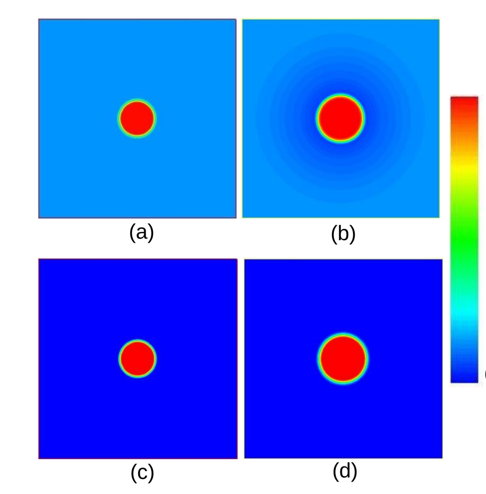

Figure 9: Evolution of fields in a supersaturated system: (a) $c_v(\tilde{\mathbf{x}},0)$, (b) $c_v(\tilde{\mathbf{x}},250)$, (c) $\eta(\tilde{\mathbf{x}},0)$, (d) $\eta(\tilde{\mathbf{x}},250)$

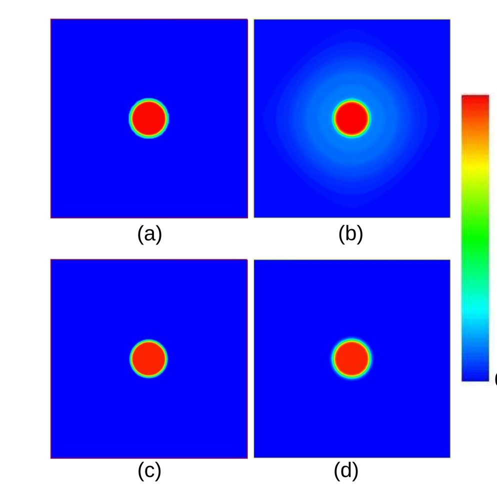

Figure 10: Evolution of fields in a under-saturated system: (a) $c_v(\tilde{\mathbf{x}},0)$, (b) $c_v(\tilde{\mathbf{x}},250)$, (c) $\eta(\tilde{\mathbf{x}},0)$, (d) $\eta(\tilde{\mathbf{x}},250)$

Void phase grows by absorbing vacancies from the supersaturated matrix (figure 9). If the matrix surrounding void is under-saturated, the void shrinks by ejecting vacancies into the system (figure 10).

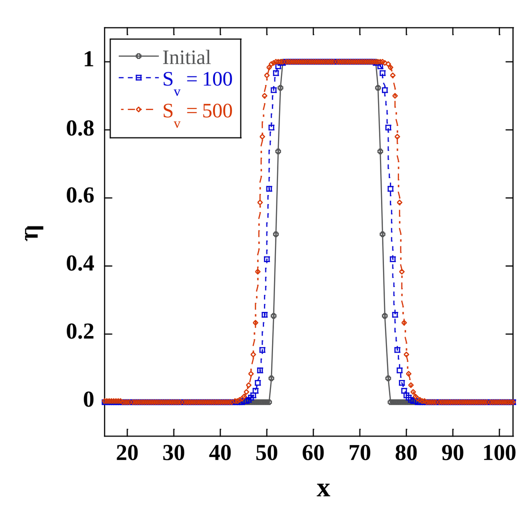

Figure 11: Line profile of $\eta(\tilde{\mathbf{x}},\tilde{t})$ through the center of the void at, initial and at $\tilde{t} = 50$

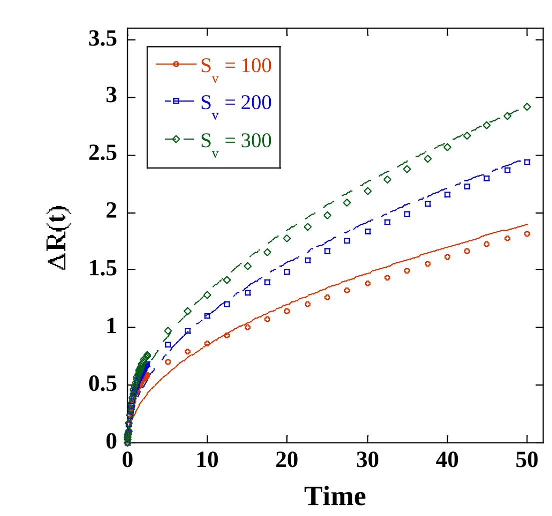

Figure 12: Void growth as a function of time in supersaturated systems (with the power law fits)

Void-void interaction

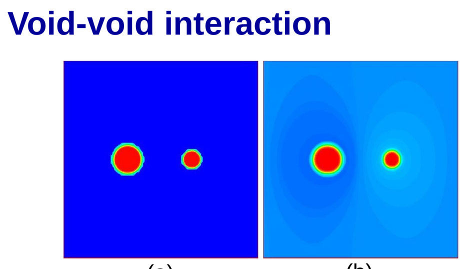

Figure 13: Evolution of concentration field around two interacting voids (a) $c_v(\tilde{\mathbf{x}},0)$, (b) $c_v(\tilde{\mathbf{x}},50)$

- Void growth is found to be dependent on the vacancy supersaturation and it follows power law $\Delta R(t) = \lambda t^n$,where $n = 0.5$
- Voids interact such that the larger voids grow at the expense of smaller voids.

Void nucleation and growth under irradiation

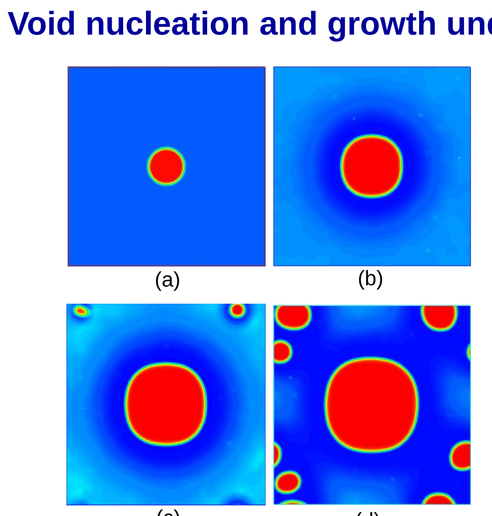

Figure 14: Vacancy field illustrating growth and nucleation of voids in a supersaturated system containing initial void (under irradiation) (a) $\tilde{t} = 0$, (b) $\tilde{t} = 100$, (c) $\tilde{t} = 190$,(d) $\tilde{t} = 250$

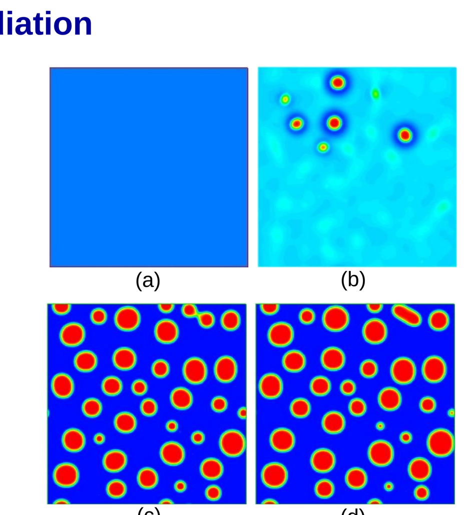

Figure 15: Vacancy field illustrating nucleation and growth of voids in a supersaturated system under irradiation,(a) $\tilde{t} = 0$, (b) $\tilde{t} = 225$, (c) $\tilde{t} = 275$, (d) $\tilde{t} = 300$

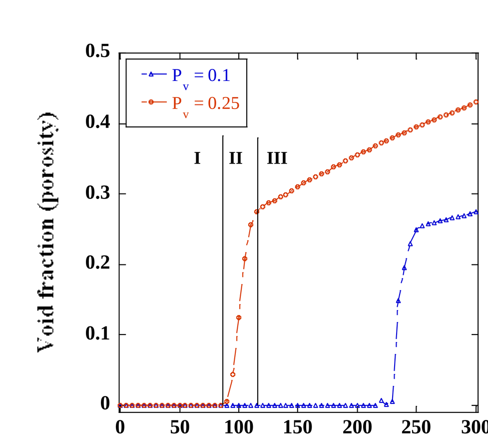

Figure 16: . Different stages of void formation observed by analysis of the void volume fraction as a function of time: stage I - incubation; stage II,- nucleation; stage III - growth

Void phase dynamics can be characterized into a series of stages:

Stage I : Incubation period
(no voids are formed)

Stage II : Nucleation regime
(at enough supersaturation multiple stable nuclei are formed to reduce the free energy of the system)

Stage III: Growth regime
(existing voids grow by absorbing vacancies from the solid, and larger voids grow at the expense of smaller ones – Ostwald Ripening).

Summary

A phase field model for tracking void growth dynamics in irradiated materials is presented. The free energy of the system is obtained in terms of point defect energies and gradient energy formulations of the heterogeneous system (based on Cahn-Hilliard and Allen-Cahn). The vacancies and voids in the system are characterized by vacancy concentration field and an order parameter, which evolve as per Cahn-Hilliard equation and Allen-Cahn equation, respectively. We observe, that voids grow by vacancy absorption or shrink by vacancy emission, corresponding the super-or under-saturation in the system. Void-void interactions and void growth in the presence of radiation is investigated. Further, nucleation and growth of voids is observed in high supersaturated system in the presence of radiation induced defect source. It has been found that the model reproduces the distinct three stages of void formation - incubation, nucleation and growth. Results from our simulations were fit to classical KJMA nucleation and Ostwald ripening process*.

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* For more details and references : S. Rokkam, A. El-Azab, P. Millet, D. Wolf. *Phase field modeling of void nucleation and growth in irradiated metals* , Modeling and Simulation in Material Science and Engineering, 2009 (under review).