Phase Field Modeling of Void Formation and Swelling 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 accounts for mutual recombination of point defects, interactions with extended defects, sinks, effects of applied stress, cascade-induced and thermally induced fluctuations. Using two dimensional solutions, we demonstrate our model capabilities for void nucleation, growth and swelling resulting from cascade-induced defects in single crystal metals.

Introduction

- High energy particles (neutrons, ions, electrons or gamma rays) collide with lattice atoms 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

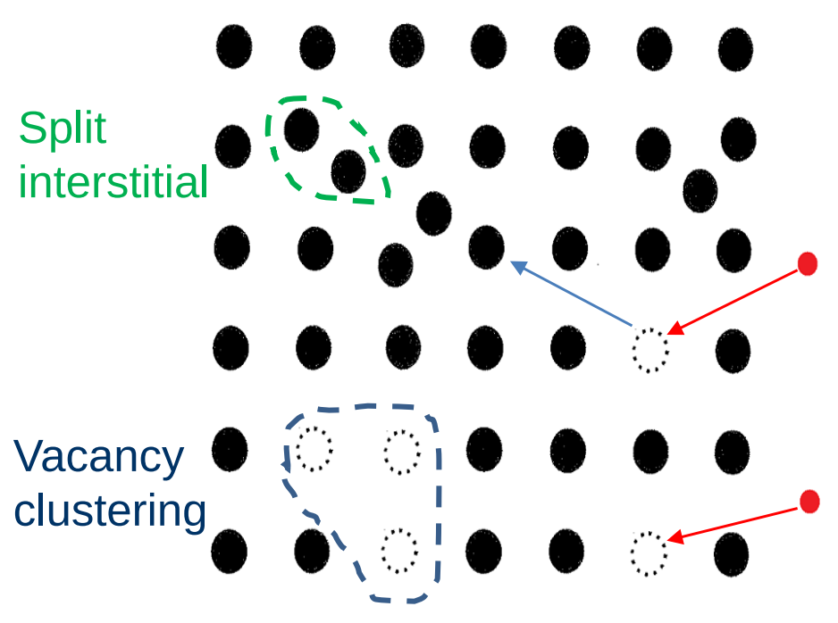


Figure 1: Schematic of radiation damage

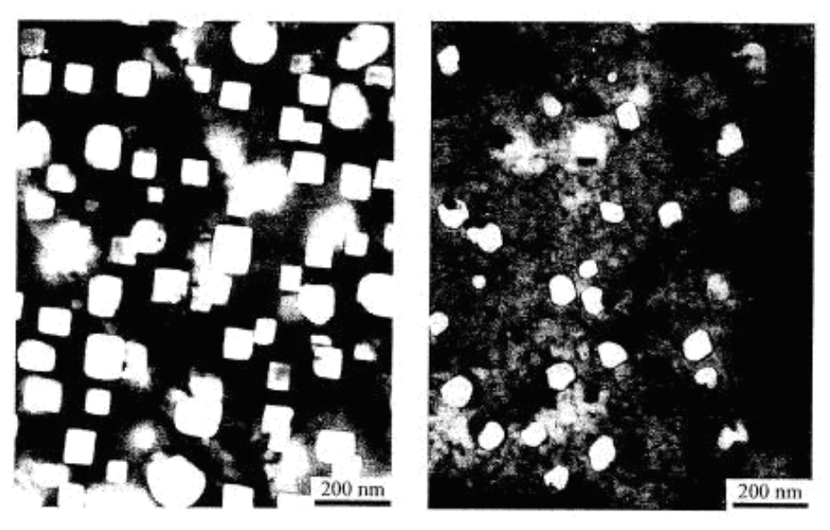


Figure 2: Micrographs of irradiation-induced voids in magnesium [Adda, 1972]

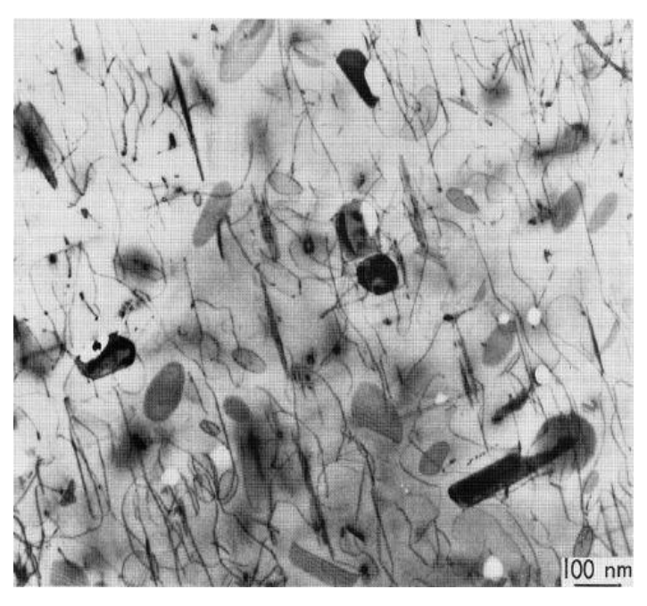


Figure 3: Dislocation loops and voids in a transmission electron micrograph (TEM) of a 300 series stainless steel irradiated at 500°C to a dose of 10 dpa [Mansur, 1994]

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

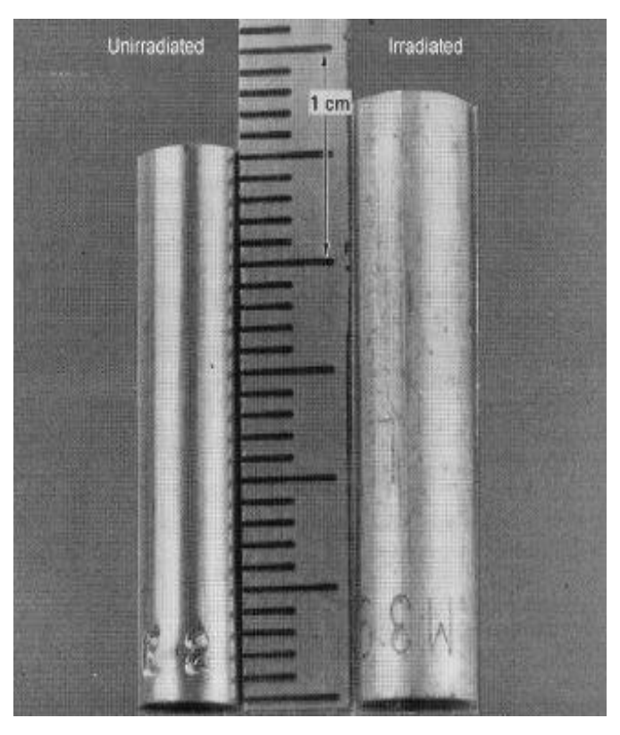
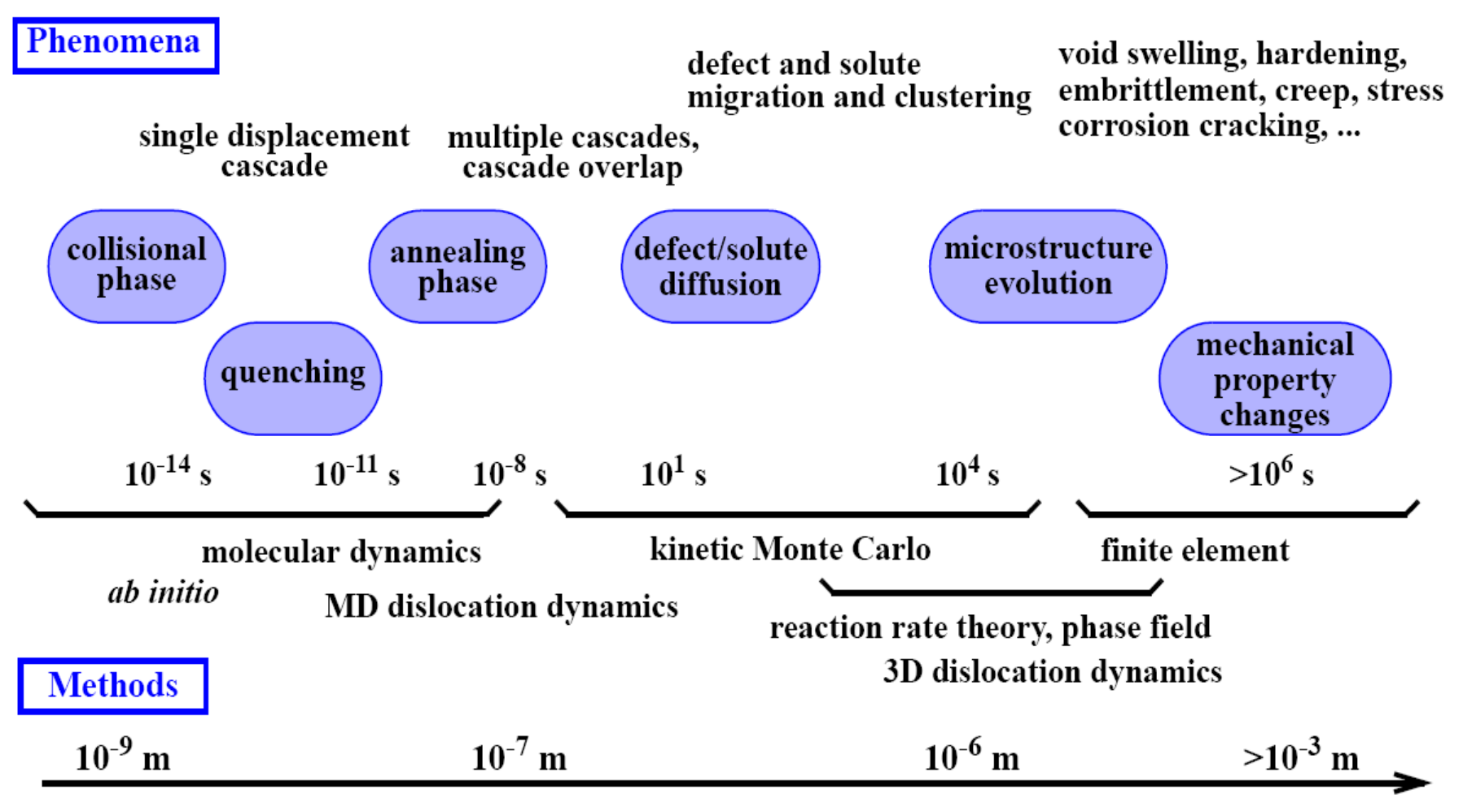


Figure 4: Photograph of 20% cold-worked 316 stainless steel rods before (left) and after (right) irradiation at 533°C to a fluence of 1.5×1023 neutrons m-2 in the EBR-11 reactor [Mansur, 1994]

Multiscale Paradigm of Phenomena



Phenomena

single displacement cascade

multiple cascades, cascade overlap

defect and solute migration and clustering

void swelling, hardening, embrittlement, creep, stress corrosion cracking, ...

collisional phase

quenching

annealing phase

defect/solute diffusion

microstructure evolution

mechanical property changes

10^{-14} s, 10^{-11} s, 10^{-8} s, 10^{1} s, 10^{4} s, $>10^{6}$ s

molecular dynamics

ab initio

MD dislocation dynamics

kinetic Monte Carlo

reaction rate theory, phase field

3D dislocation dynamics

finite element

Methods

10^{-9} m, 10^{-7} m, 10^{-6} m, $>10^{-3}$ m

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

Phase Field Method for Void Microstructure Evolution

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
- Voids grow by absorption of vacancies or emission of interstitials and vice-versa.
- In order to distinguish, solid from void regions we use a long-range order parameter (η).
- Cascade induced defects are modelled using a core-shell type description of cascade zone so as to obtain a vacancy rich core and interstitial rich shell
- Main features include: point defect production and annihilation, diffusion, nucleation and growth of voids and volumetric swelling of material.

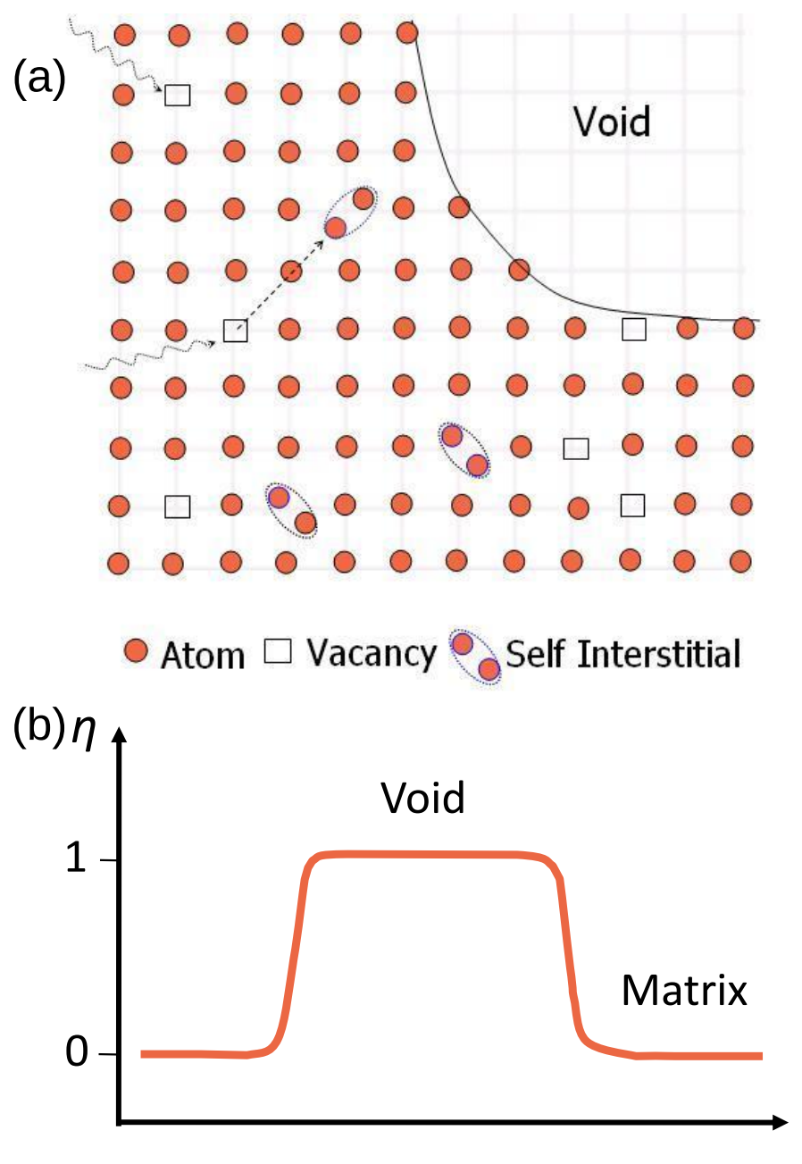


Figure 6: Schematic of (a) void phase and defects in a quasi-lattice, (b) void phase field representation

Free Energy of the System

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 regular solid regions with vacancies and interstitials, along with void regions can be obtained in terms of of vacancy concentration $c_v(\mathbf{r},t)$, interstitial concentration $c_i(\mathbf{r},t)$ and phase field $\eta(\mathbf{r},t)$ as below :

$$\psi(c_v,c_i,\eta)=N\int_\Omega\Big[\underbrace{\underbrace{h(\eta)\psi^m(c_v,c_i)}_{\text{Defect Free Energy}}+\underbrace{w(c_v,c_i,\eta)}_{\text{Void phase Energy}}}_{\text{Bulk free energy}}\ \underbrace{+\kappa_v|\nabla c_v|^2+\kappa_i|\nabla c_i|^2+\kappa_\eta|\nabla\eta|^2}_{\text{Gradient energy terms (due to field inhomogeneity)}}\underbrace{+\psi^{el}(c_v,c_i,\eta)}_{\text{Elastic effects due to defects and applied stress}}\Big]d\Omega$$

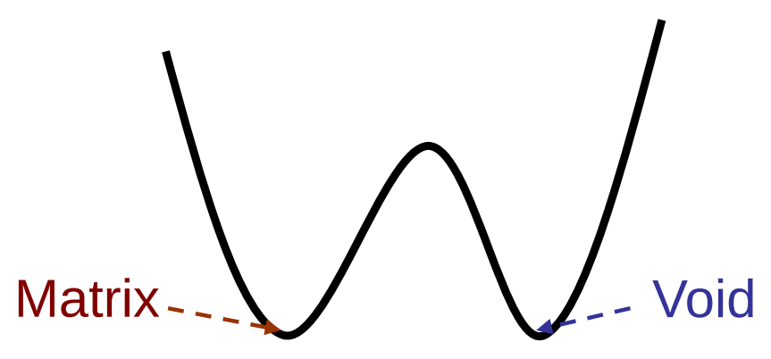


Figure 7: Schematic of the wells due to bulk free energy

The local free energy density of the system is derived in terms of the enthalpic and entropic contributions of point defects.

Two stable wells:

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

$\eta=1\ ,\ c_v=1,\ c_i=0$ (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:

Vacancy concentration field : modified *Cahn-Hilliard* type evolution equation

$$\frac{\partial c_v(\mathbf{r},t)}{\partial t}=\nabla\cdot M_v\nabla\frac{1}{N}\frac{\delta\psi}{\delta c_v}+\xi_v(\mathbf{r},t)+\underset{\text{source}}{P_v(\mathbf{r},t)}-\underbrace{R_{iv}(\mathbf{r},t)-R_v^{GB}(\mathbf{r},t)}_{\text{sinks}}$$

Interstitial concentration field : modified *Cahn-Hilliard* type evolution equation

$$\frac{\partial c_i(\mathbf{r},t)}{\partial t}=\nabla\cdot M_i\nabla\frac{1}{N}\frac{\delta\psi}{\delta c_i}+\xi_i(\mathbf{r},t)+\underset{\text{source}}{P_i(\mathbf{r},t)}-\underbrace{R_{iv}(\mathbf{r},t)-R_i^{GB}(\mathbf{r},t)}_{\text{sinks}}$$

Void phase field : *Allen-Cahn* (or time dependent Ginzburg-Landau) type equation

$$\frac{\partial\eta(\mathbf{x},t)}{\partial t}=-L\frac{\delta\psi}{\delta\eta}+\zeta(\mathbf{r},t)$$

Here, $\xi_v(\mathbf{r},t),\xi_i(\mathbf{r},t),\zeta(\mathbf{r},t)$ - stochastic terms accounting for thermal fluctuations.

$P_\alpha(\mathbf{x},t)$ - amount of α defects entering the system due to radiation damage.

Numerical Solution Scheme

- Substituting the variational derivatives, and mobility, $M_\alpha=D_\alpha c_\alpha(1-c_\alpha)/k_BT$ we get the temporal evolution equations (in the absence of elastic effects).
- We transform the evolution equations using a length scale, $l=\sqrt{\kappa_\eta/k_BT}$ and timescale $\tau=l^2/D_v$
- The evolution equations gives us three coupled partial differential equations which are solved using finite differences on a 2D domain. We use explicit finite differences in space and forward Euler time marching scheme.

Vacancy field

$$c_v\Big|_{j,k}^{n+1}=c_v\Big|_{j,k}^{n}+\frac{\Delta t}{(\Delta x)^2}\left(f_v\Big|_{j+1,k}^{n}+f_v\Big|_{j-1,k}^{n}+f_v\Big|_{j,k+1}^{n}+f_v\Big|_{j,k-1}^{n}-4f_v\Big|_{j,k}^{n}\right)+P_v(\tilde{x},\tilde{t})-\left(R_{vi}c_vc_i\right)_{j,k}^{n}$$

Interstitial field

$$c_i\Big|_{j,k}^{n+1}=c_i\Big|_{j,k}^{n}+\frac{\Delta t}{(\Delta x)^2}\left(f_i\Big|_{j+1,k}^{n}+f_i\Big|_{j-1,k}^{n}+f_i\Big|_{j,k+1}^{n}+f_i\Big|_{j,k-1}^{n}-4f_i\Big|_{j,k}^{n}\right)+P_i(\tilde{x},\tilde{t})-\left(R_{vi}c_vc_i\right)_{j,k}^{n}$$

Void phase field

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

Where $f_v\Big|_{j,k}^{n}, f_i\Big|_{j,k}^{n}$ and $f_\eta\Big|_{j,k}^{n}$ are functional derivatives evaluated at time step *n* and point *(x_j,y_k)*

Results and Discussion

Void growth in a supersaturated system

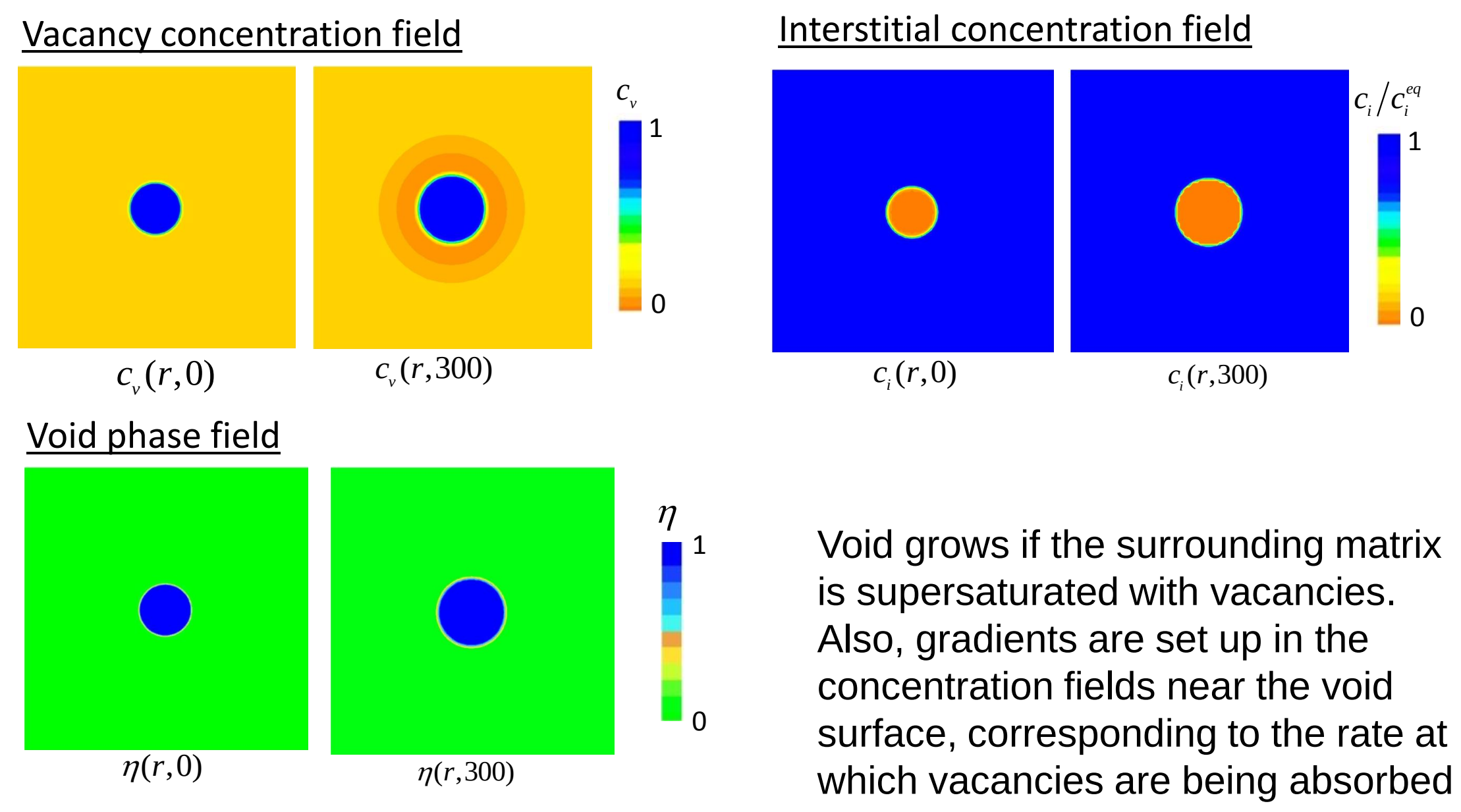


Figure 8: Evolution of fields in a system which is initially supersaturated with vacancies, Sv = 20, Si = 1

Void grows if the surrounding matrix is supersaturated with vacancies. Also, gradients are set up in the concentration fields near the void surface, corresponding to the rate at which vacancies are being absorbed into the void.

Void growth and shrinkage dynamics

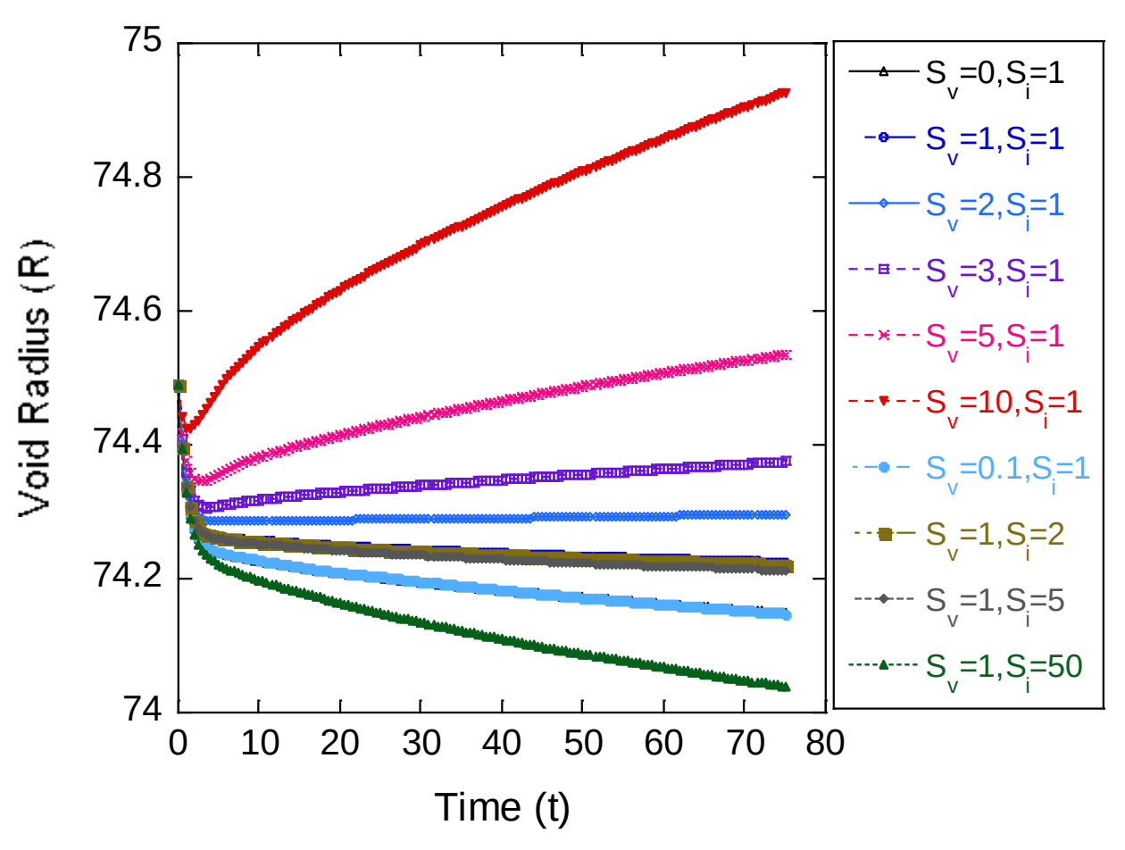


Figure 9: Void radius as a function of time for different supersaturation ratios

Void phase grows by absorbing vacancies from the supersaturated matrix and shrinks by absorbing interstitials or emitting vacancies. Void of a given radius can sustain only if a certain background supersaturation exists (a manifestation of *Gibbs-Thomson effect* in our model).

Results and Discussion … continued

Void nucleation and growth under irradiation

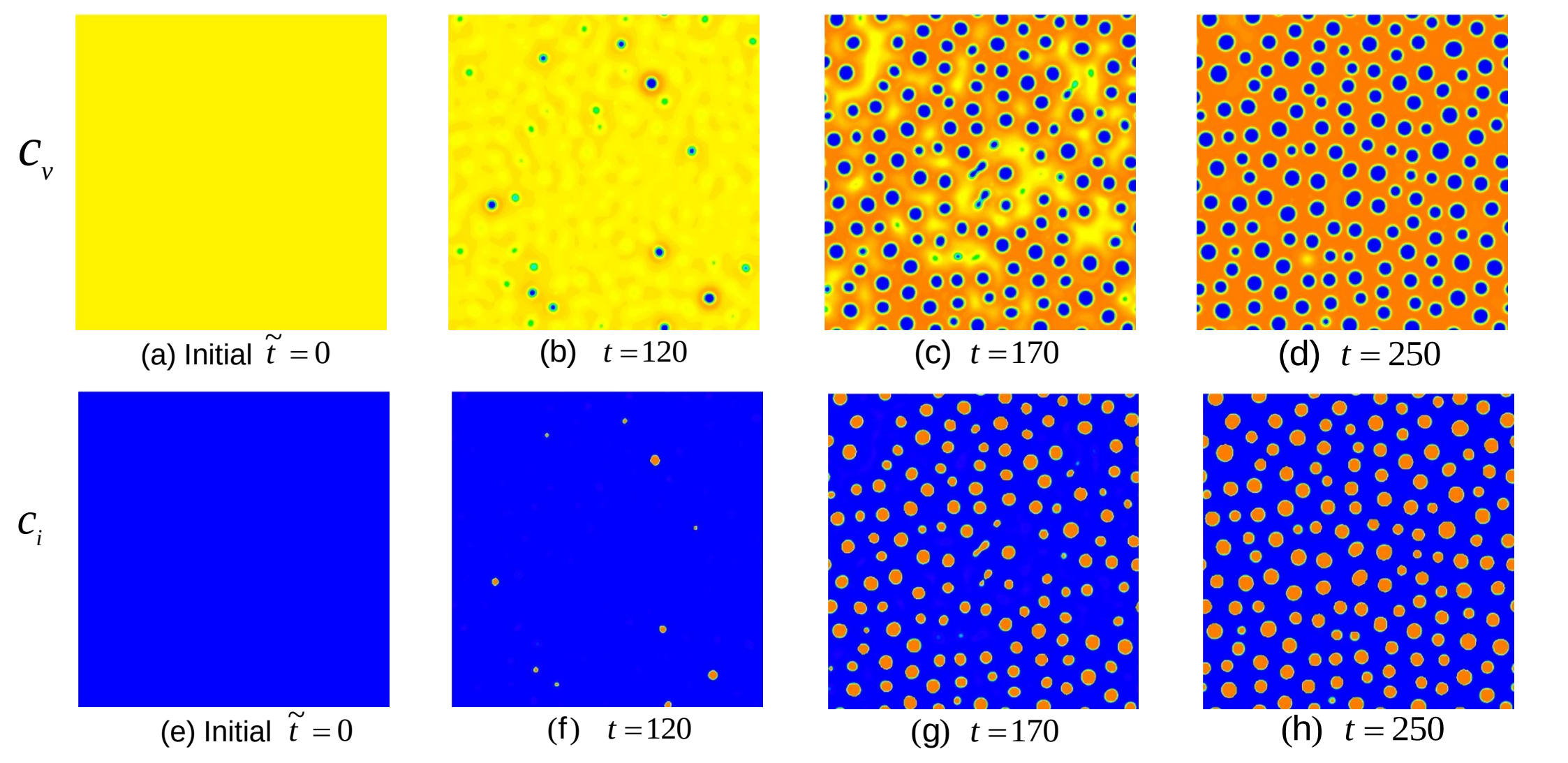


Figure 10: (a)-(d) Vacancy field evolution , (e)-(h) Interstitial field evolution, showing void formation in a supersaturated matrix due to point defects produced during irradiation.

Void formation in polycrystalline metal

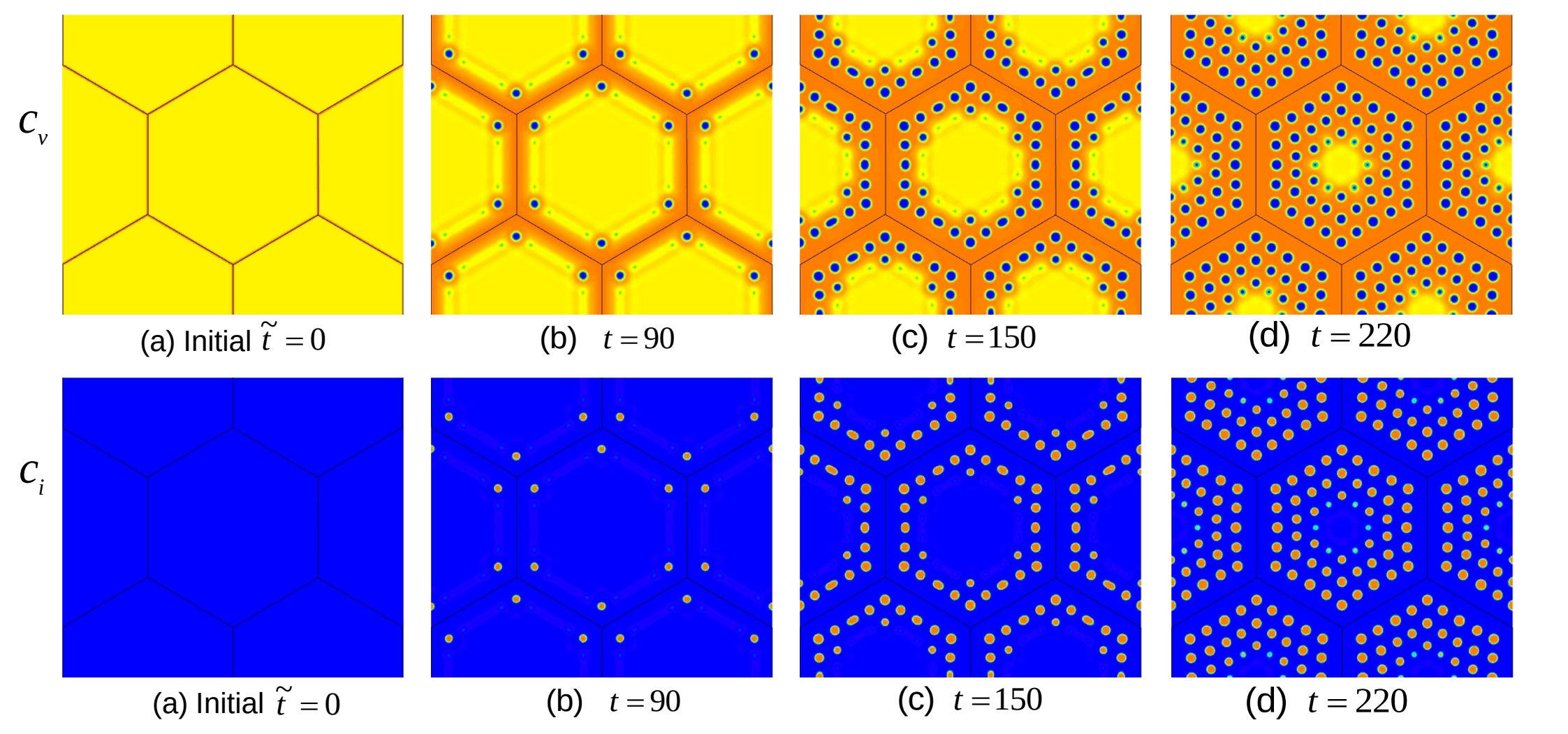


Figure 11: (a)-(d) Vacancy field evolution , (e)-(h) Interstitial field evolution showing void formation in a supersaturated polycrystalline matrix due to point defects produced during irradiation.

Void pattern formation

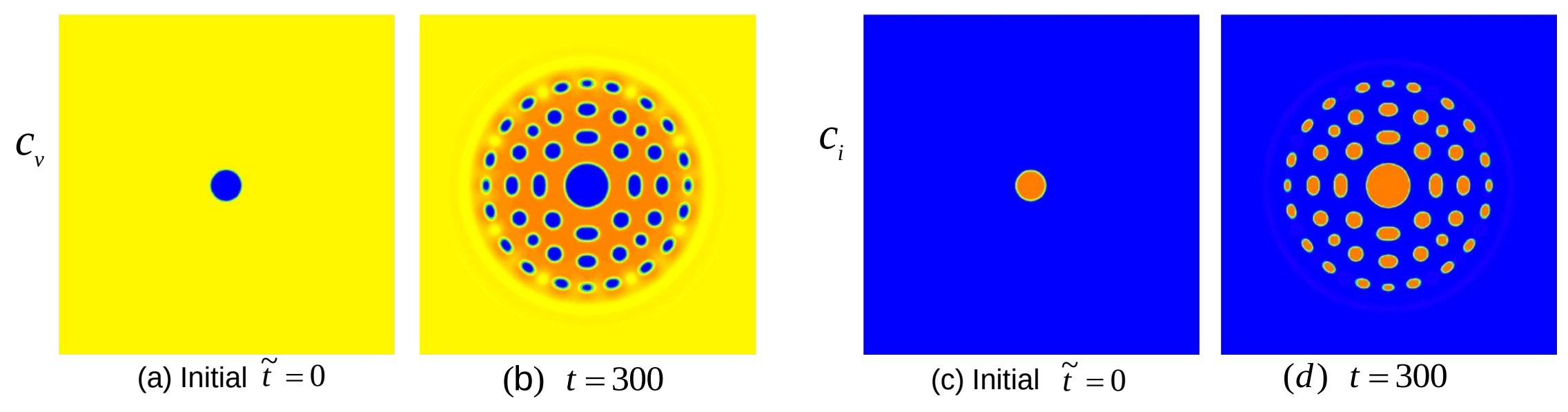


Figure 12: (a)-(b) Vacancy field evolution, (c)-(d) Interstitial field evolution showing void patterns in a supersaturated matrix (in the absence of irradiation). S_v=50, S_i = 1

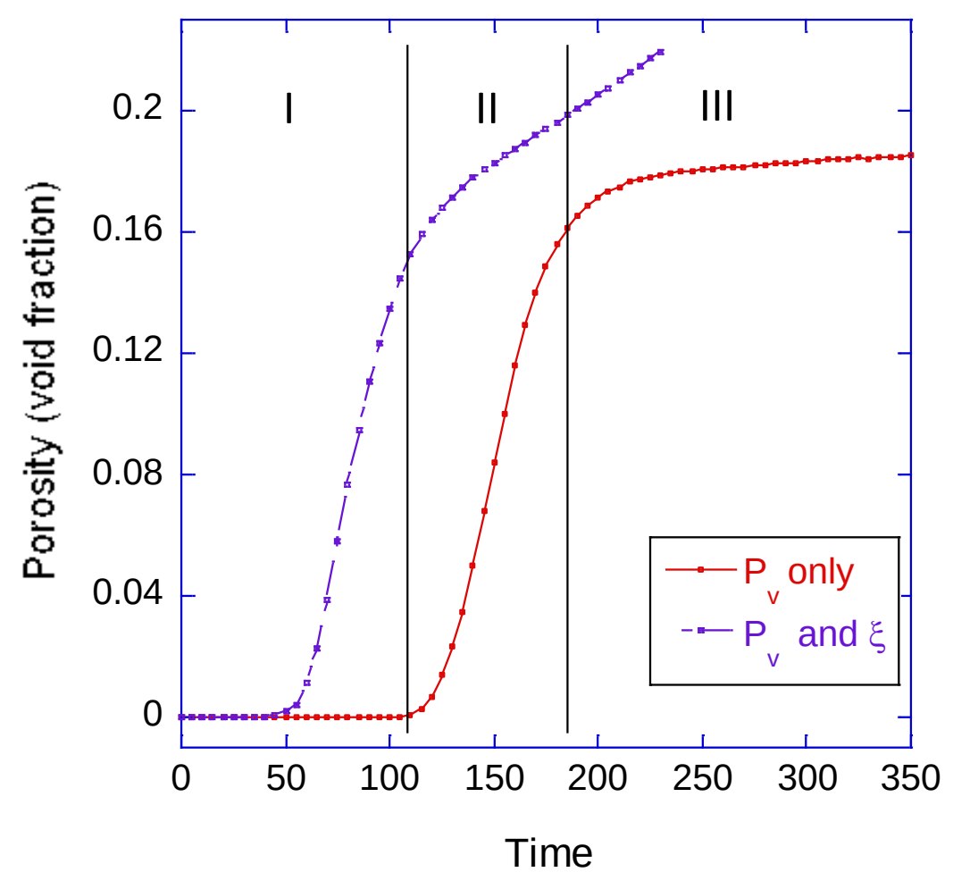


Figure 13: Different stages of void formation as a function of time

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

Stage I : Incubation period

(no voids are formed)

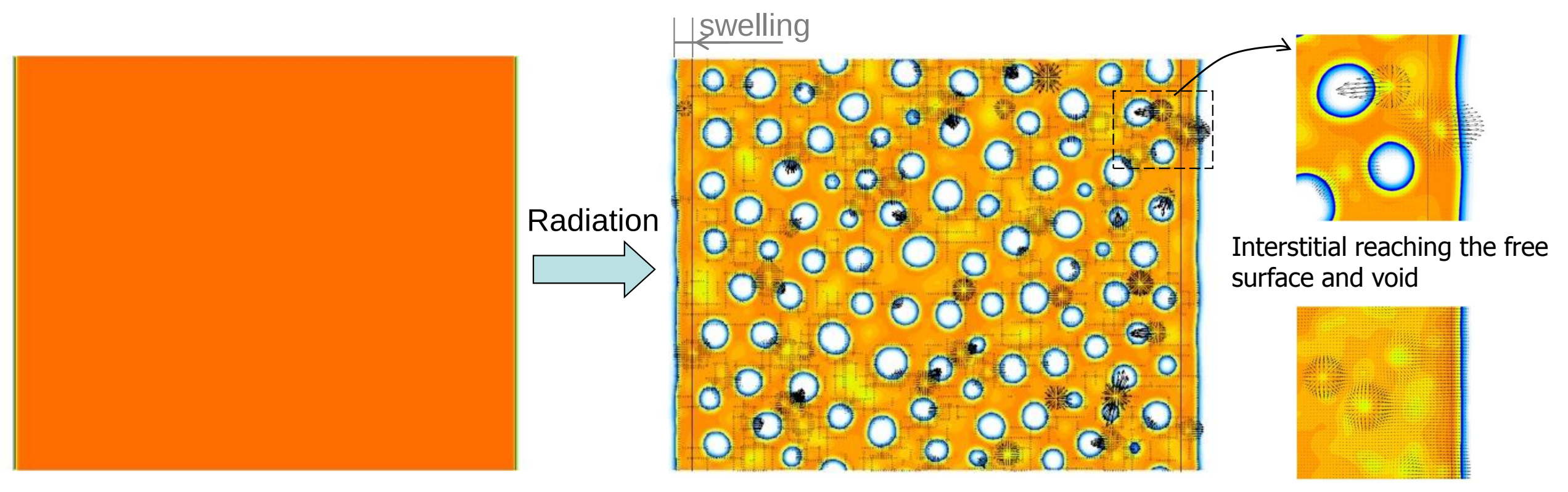
Stage II : Nucleation regime

(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).

Swelling of the material



Interstitial reaching the free surface and void

Figure 14: Finite Matrix domain (in 2D) subject to irradiation can undergo swelling in addition to the formation of voids. Shown here, vacancy concentration field, and interstitial flux lines. Circular exploding flux lines correspond to interstitials flux resulting from cascade damage process

Interstitials reaching the boundary push the matrix surface outward

Summary

A phase field formulation for modeling void formation and associated swelling in irradiated materials is presented. The point defects (vacancies, interstitials) and voids in the system are characterized by defect concentration fields 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 interstitial emission and shrink by interstitial absorption or 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 the presence of radiation induced defect sources and thermal fluctuations. 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. Finite domain simulations (with zero flux boundary condition) reveal swelling of the matrix domain. Further, we observe that the swelling of the material can occur prior to the nucleation of voids, by virtue of the free volume of interstitials reaching the material surface. It is noted that void nucleation in fact restricts the amount of swelling.

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