

Theory of microstructure evolution under fusion neutron irradiation [★]

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New concepts are reviewed which replace the conventional separation of microstructure evolution analysis into nucleation and growth. Classical nucleation theory is inadequate under fusion conditions (high helium-to-dpa ratios) and the usual “mean field” approximation of microstructural growth cannot account for cascade effects. A comprehensive theory of microstructure evolution under fusion conditions is formulated based on non-equilibrium statistical mechanics. Dynamic re-solution of helium gas in cavities is shown to result in continuous nucleation of helium-filled cavities. Microstructure evolution (e.g., dislocation loops and cavities) is modeled by kinetic rate equations for small size features and Fokker–Planck (F–P) equations for sizes larger than few atomic dimensions. Semi-analytical and numerical methods are developed for the analysis of microstructure evolution and the results are compared to experiments. The problem of spatial self-organization of microstructures under irradiation is described in terms of a newly developed Ginzburg–Landau-type equation and the results are also compared to experiments.

1. Introduction

Understanding the relationships between microstructures and material properties is central to the development of new alloys. This is particularly significant for improving the radiation resistance of structural alloys for fusion reactors. Under the intense neutron radiation environment, large amounts of point defects and gases are generated, driving the solid far from thermodynamic equilibrium. Restoration of the equilibrium state is achieved by a slow evolutionary process of the solid's microstructure. In this respect, point-defect generation by atomic displacements can be viewed as the main driving force for microstructure evolution and, hence, for all property changes in a fusion irradiation environment.

The problem of cavity swelling under irradiation has occupied a central position in the theory of microstructure evolution because of the impact of volumetric swelling on the lifetimes of structural components under irradiation. Traditionally, cavity formation in irradiated metals has been theoretically analyzed in two distinct phases: nucleation and growth. It has been implicitly assumed that the nucleation of cavities is a fairly rapid process which is followed by the growth phase. Cavity nucleation theories, as formulated by Katz and Wiedersich [1,2] and equivalently by Russell [3,4], have been motivated by the classical nucleation theory of droplet formation, developed earlier by Becker and Doring [5] and by Zeldovich [6]. Cavity growth, however, has been treated in the “mean field” approximation of identical spherical sinks which grow in the diffusion fields of point defects. The rate theory of

“average” cavity growth has been contributed to by many investigators (e.g., refs. [7–13]), and many features of experimentally observed cavity growth behavior were explained or predicted. However, theoretical inconsistencies within this framework have persisted until recently and they are summarized below.

(1) The continuous production of gas atoms and point defects is in contradiction with the termination of nucleation by a sudden decrease in the vacancy supersaturation. (This is a requisite in classical nucleation theory.)

(2) Classical nucleation theory predicts nucleation rates which are extremely sensitive to parametric variations, such as surface energy, supersaturation ratio, and number of gas atoms in a cavity. It also does not give quantitative information on cavity densities.

(3) Rate theory of cavity growth is unable to explain variations in the size distribution of cavities, their spatial inhomogeneities, and the effects of collision cascades. Coalescence, which can lead to a broadening of the size distribution, is not included in the present analysis. The migration of dislocation loops or cavities is normally a very slow process, relevant only at high temperatures.

Another recent approach to the theory of microstructure evolution has been developed which is based on extending the concepts of nonequilibrium statistical mechanics to describe microstructure evolution, particularly dislocation loops [14–19] and gas-filled cavities [20–25]. This approach overcomes problems associated with the artificial separation of nucleation and growth of microstructural components and the extreme parametric sensitivity of nucleation theory. Additionally, the approach allows for a detailed description of the size distribution of evolving microstructural features (e.g., dislocation loops and cavities), and for the influence of collision cascades.

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Certain aspects of the spatial self-organization of microstructures (i.e., void and bubble lattices, and dislocation loop walls and 3-D cubic arrangements) can be explained as dynamical instabilities resulting from the interaction of the spatio-temporal diffusion fields of mobile point defects and the immobile microstructures. Recent progress in this area can be found in refs. [26–32].

In this paper, we present a review of a statistical theory of cavity and loop evolution under fusion neutron-irradiation conditions. We also show that spatial instabilities in the point-defect diffusion fields can lead to an explanation of experimentally observed features of dislocations-loop self-organization. In section 2, we give the main equations which define, in a unified way, the nucleation and growth aspects of microstructures in the framework of statistical mechanics. This is followed by the theory of self-organization in section 3. The results of example computations are given in section 4, and conclusions are stated in section 5.

2. Statistical theory of microstructure evolution

Helium migration under irradiation is determined by a competition between the rate of transport through the lattice until it is trapped, and the detrapping rate from helium–vacancy (He–V) clusters. Near steady state, however, the most effective traps are single vacancies, and the dominant detrapping mechanism has been shown to be by thermal dissociation at high temperatures, by self-interstitial replacement at intermediate temperatures, and by displacement reactions at lower temperatures [33].

The starting point of the theory of microstructure evolution is the Smoluchowsky–Chapman–Kolmogorov (SCK) equation for a Markovian process [34], i.e.,

$$\frac{\partial f}{\partial t} = \int [\omega(\bar{x}', \bar{x}, t')f(\bar{x}', t) - \omega(\bar{x}, \bar{x}', t')f(\bar{x}, t)] d\bar{x}', \tag{1}$$

where $f(\bar{x}, t)$ is the probability distribution for the stochastic variable \bar{x} at time t . The transition probability per unit time from state \bar{x} to state \bar{x}' at time t' is $\omega(\bar{x}, \bar{x}', t')$. Eq. (1) can be reduced to a deterministic rate equation for the concentration of a specific defect cluster (e.g., di-interstitials, di-vacancies, tri-vacancies and two helium atoms, etc.) if the transition probabilities, ω , are replaced by reaction rates. This mean-field approximation does not take into account the statistical nature of defect production, cascade effects, and the arrival and absorption of single and multiple defects at defect clusters.

Since defect clustering is driven by the concentration of three types of monomers (vacancies, interstitials, and helium), we would have a coupled hierarchy of discrete equations for the probability distribution, f , using rate

or master equations of the form given by eq. (1). One unique feature of microstructure evolution under irradiation, however, is that the size of atomic transitions ($\bar{x} - \bar{x}'$) is generally smaller than the defect-cluster size \bar{x} . The mobility of large defect clusters (≥ 3) is negligible and, hence, one can derive a F–P equation as an approximation to the master equation (eq. (1)).

The transition probability from a defect-cluster size \bar{x}' to size \bar{x} , $\omega(\bar{x}', \bar{x}, t')$, can be redefined in the following way:

$$\omega(\bar{x}', \bar{x}, t') = W(\bar{x}', \bar{x} - \bar{x}', t') = W(\bar{x} - \bar{r}, \bar{r}, t, \tau). \tag{2}$$

In eq. (2), we distinguish between slow (\bar{x}, t) and fast ($\bar{r} = \bar{x} - \bar{x}', \tau$) variables. By expanding the function $W(\bar{x} - \bar{r}, \bar{r}, t, \tau)f(\bar{x} - \bar{r}, t)$ in a Taylor series for the slow variable (\bar{x}), truncating to second order, and integrating over an appropriate correlation time, T , we obtain the F–P equation:

$$\frac{\partial f(\bar{x}, t)}{\partial t} = \frac{1}{2} \sum_{i,j} \frac{\partial^2}{\partial x_i \partial x_j} (\langle r_i r_j \rangle f) - \sum_i \frac{\partial}{\partial x_i} (\langle r_i \rangle f), \tag{3}$$

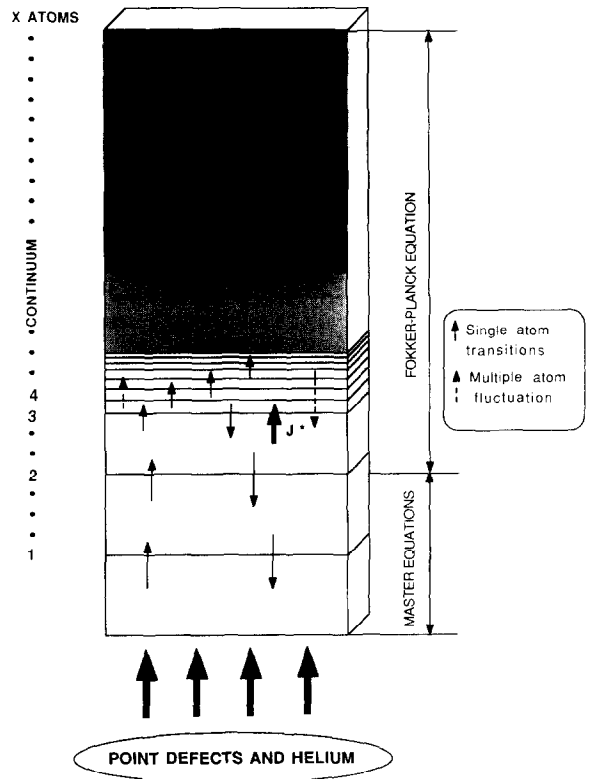


Fig. 1. Schematic representation of the clustering model. The number of interstitials, vacancies, or helium atoms in a cluster are represented by x .

where

$$\langle r_i \rangle(\bar{x}, t) = \frac{1}{T} \int_0^T \int_1^\infty r_i w(\bar{x}, r, t, \tau) d\bar{r} d\tau, \quad (4)$$

and

$$\langle r_i r_j \rangle(\bar{x}, t) = \frac{1}{T} \int_0^T \int_1^\infty r_i r_j w(\bar{x}, \bar{r}, t, \tau) d\bar{r} d\tau. \quad (5)$$

The indexes i or j represent the dimension in cluster size space (1-D for interstitial loops and 2-D for a He-V cluster). Eqs. (4) and (5) give the first and second moments of the transition probabilities. The correlation time, T , is chosen to represent the appropriate physics of the relevant transition (e.g., inverse of arrival frequency for single-atom transitions or cascade-production frequency for cascade-induced transitions). Eq. (4) represents the elements of a drift vector, while eq. (5) is used to derive the elements of a dispersion tensor. A schematic representation of this model is shown in fig. 1.

The following is a summary of the general procedure for the implementation of the theory. A set of rate equations for the concentrations of single defects and small defect aggregates is formulated to represent time-dependent nucleation. Larger-size defect clusters are treated by the F-P approximation given by eq. (3), with the transition moments obtained via eqs. (4) and (5). The rate equations are coupled to the F-P equation through a current boundary condition, and the solution is obtained semi-analytically as in ref. [19], or numerically as in ref. [25].

2.1. Interstitial loop evolution

The diffusion coefficient of self-interstitials is generally several orders of magnitude higher than that of vacancies. As a result, all clustering transition probabilities driven by self-interstitials have much faster time variations than those driven by vacancies. Moreover, an interstitial loop cluster containing 2 to 3 atoms is stable against thermal dissociation. Consequently, several investigators [14–18] have shown that the time scale for interstitial loop evolution is much shorter than that for cavity evolution. This observation simplifies the analysis and the loop evolution can be approximately separated from that of cavities.

Eq. (3) can be written in the 1-D form for interstitial loops:

$$\frac{\partial f}{\partial t} = - \frac{\partial}{\partial x} (Ff) + \frac{\partial^2}{\partial x^2} (Df), \quad (6)$$

where x is the number of atoms in a loop, $F(x, t)$ is a scalar drift coefficient defined by eq. (4) and $D(x, t)$ is a scalar dispersion coefficient defined by eq. (5). Analytical moment equations have been developed from eq. (6) in the form of evolution equations for the average

size, $\langle x \rangle$, and higher order moments, M_r , $r = 2, 3, \dots, \infty$. These are given by:

$$\begin{aligned} \frac{d}{dt} \langle x \rangle &= \langle F \rangle + \xi_1, \\ \frac{d}{dt} M_r &= \psi_r + \Phi_r + \xi_r, \quad r = 2, 3, \dots, \infty. \end{aligned} \quad (7)$$

The symbol $\langle \rangle$ is used for averages over the loop distribution function. The dispersion functions, ψ_r , the distortion functions, Φ_r , and the nucleation functions, ξ_r , are defined in ref. [19]. They are also dependent upon the average size $\langle x \rangle$ and the moments, M_r , which couple the system of evolution of eq. (7).

This system of eq. (7) is usually solved numerically, except in special cases where simplifying assumptions can be invoked. Once the moments are determined, the loop size-distribution function, f , can be reconstructed as:

$$\begin{aligned} f(x, t) &= \frac{1}{\sqrt{2\pi M_2}} \exp\left[-\frac{y^2}{2}\right] \left\{ 1 + \frac{M_3}{6M_2^{3/2}} H_3(y) \right. \\ &\quad + \frac{1}{24} \left[\frac{M_4}{M_2^2} - 3 \right] H_4(y) \\ &\quad + \frac{1}{120} \left[\frac{M_5}{M_2^{5/2}} - 10 \frac{M_3}{M_2^{3/2}} \right] H_5(y) \\ &\quad \left. + \frac{1}{720} \left[\frac{M_6}{M_2^3} - 15 \frac{M_4}{M_2^2} + 30 \right] H_6(y) \right\}, \quad (8) \end{aligned}$$

where $y = [(x - \langle x \rangle) / \sqrt{M_2}]$ and H_j , $j = 1, 2, \dots, 6$ are the Hermite polynomials.

Application of the theory to interstitial loops, as outlined in this section, has shown that the probability distribution function given by eq. (8) is consistent with ion-irradiation experiments [16]. The comparison revealed that under cascade-irradiation conditions, cascade-induced fluctuations of loop sizes are the main contributors to the dispersion coefficient, D .

2.2. Cavity evolution

For the case where cascade-induced transitions are not dominant, the six elements in an F-P model of He-V cavity evolution are given by:

$$\begin{aligned} \langle r_v \rangle &= k^{vc} - (k^{ic+ve} + k^{hr}), \\ \langle r_h \rangle &= k^{hc} - (k^{he} + k^{hr}), \\ \langle r_v r_v \rangle &= \frac{1}{2!} (k^{ic+ve} + k^{hr} + k^{vc}), \\ \langle r_h r_h \rangle &= \frac{1}{2!} (k^{he} + k^{hr} + k^{hc}), \\ \langle r_v r_h \rangle &= \langle r_h r_v \rangle = k^{hr}, \end{aligned} \quad (9)$$

where the subscript/superscript notation is as follows: i = interstitial, v = vacancy, h = helium, c = capture, e = emission, r = replacements, and k = reaction rates. In

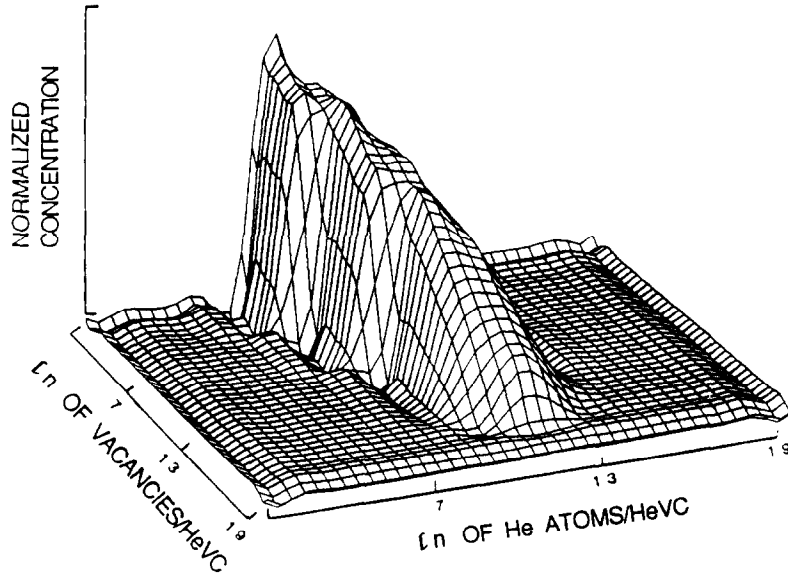


Fig. 2. Evolution of cavity size distribution for HFIR irradiation conditions at 450°C.

a 2-D size space, He-V clusters would have trajectories which depend on their initial conditions and the moments of the transition probabilities, given by eq. (9). Clusters would propagate by a Brownian-like motion in a drift field. The dispersion coefficients are given by the second moments while the drift coefficients are described by the first moments of eq. (9).

Fig. 2 shows the results of computations for the cavity size distribution in stainless steel for HFIR irradiation conditions at 450°C. It is to be noted that the majority of He-V clusters deviate from the state of thermodynamic equilibrium (gas pressure = surface tension stress), and tend to contain more vacancies than necessary for equilibrium. A comparison between com-

putations and HFIR experimental data [35] is shown in fig. 3. The moments of atomic transition probabilities are computed on the basis of single-atom transitions determined by eq. (9). The comparison with experimental data indicates that without cascade effects, the width of the computed cavity distribution function is smaller than that observed experimentally.

3. Spatial instabilities and self-organization

Many experimental observations have shown striking spatial regularities in microstructures (e.g., void and bubble lattices, dislocation loop walls, etc.). To illustrate the origins of this self-organization, we include the spatial diffusion operators for vacancies and interstitials, while larger clusters are considered to be immobile. A minimal model is proposed by Murphy [28] in which linear stability analysis is performed on the vacancy and interstitial concentrations and the vacancy dislocation-loop density, ρ . The wavelength predicted from the dispersion relationship by Walgraef and Ghoniem [32] is in good agreement with experiments on loop self-organization. The critical wavelength, λ_c , is given by the equation

$$\lambda_c = 2\pi \left[\frac{D_v (\bar{C}_{vL} - \bar{C}_{vN})}{(1+B)\epsilon K \rho_N} \right]^{1/4} \quad (10)$$

where D_v is the vacancy diffusion coefficient, ϵ is the cascade collapse efficiency, $(1+B)$ is the dislocation bias factor, K the displacement rate, ρ_N the network density, \bar{C}_{vL} the average thermodynamic vacancy concentration at vacancy loops, and \bar{C}_{vN} the average thermodynamic vacancy concentration at dislocations. The wavelength predicted by eq. (10) decreases with increas-

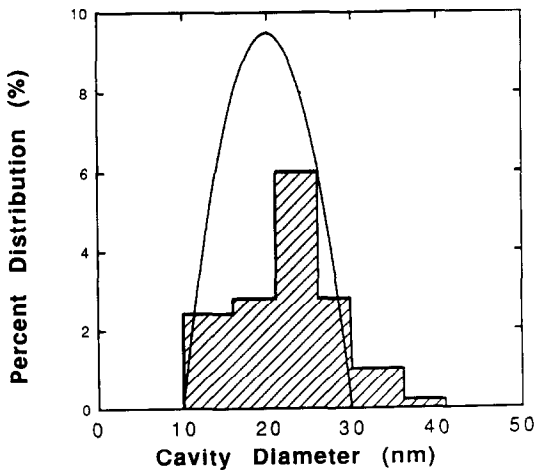


Fig. 3. Comparison of computed and experimentally measured cavity size distribution (histogram) at 14.3 dpa for HFIR irradiation conditions at 450°C (experimental data [35]).

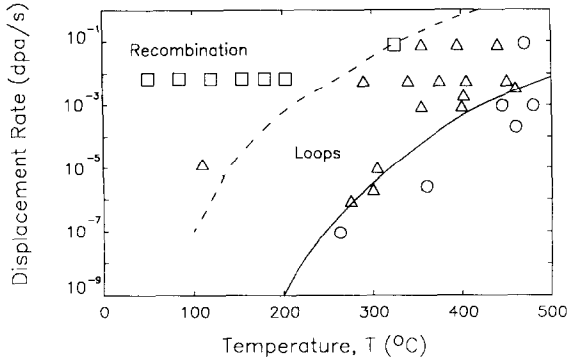


Fig. 4. Comparison between the Walgraef-Ghoniem theory and experimentally observed ordered microstructures in irradiated Ni [36,37]. The region of dominant recombination is calculated by Abromeit and Wollenberger [31] [random defect clusters and dislocations (□), periodic cluster arrays (Δ), random voids and dislocations (○)].

ing network dislocation density, cascade collapse efficiency, and damage rate. These predictions are consistent with experimental observations [32]. Physically, the strength of point-defect fluctuations is increased by an increase in ϵ or K , while a shorter distance to absorption at sinks tends to damp these fluctuations.

In the weakly nonlinear regime, point-defect concentrations can be expanded as a power series in the vacancy-loop density which leads to a Ginzburg-Landau equation for ρ of the form

$$\dot{\rho} = \left[\left(\frac{b - b_c}{b_c} \right) - \xi_0^2 (\bar{q}_c^2 + \nabla^2)^2 \right] \rho + v\rho^2 - u\rho^3, \quad (11)$$

where $b = B/\epsilon$, $\xi_0^2 = (\rho_N/\rho_0)$, $v = 2/(x_0)^{3/2}$, $u = 2/(x_0)^{5/2}$, and $x_0 = \rho_0/\rho_N$. The homogeneous solution for the loop density is ρ_0 , b_c is the critical value of the bifurcation parameter at the instability, and \bar{q}_c is the critical wave vector. The density in eq. (11) can be expressed in terms of a slowly varying amplitude and the critical wave vector, giving rise to a bifurcation diagram for the amplitude versus the bifurcation parameter. The details of the theory developed by Walgraef and Ghoniem are given in ref. [32]. A comparison between this theory and experimental observations is shown in fig. 4. Microstructural self-organization can be explained as a phase transition in analogy to the Landau-Ginzburg analysis.

4. Conclusions and outlook

The theory of microstructure evolution under irradiation conditions typical of fusion reactor environments (i.e., continuous helium generation and cascade damage) combines nucleation and growth of microstructural features in a unified way. Concepts of non-equilibrium statistical mechanics are used to combine these two

phases of microstructure evolution. Applications of the theory to fusion conditions show the following features.

(1) Nucleation of interstitial loops is very rapid and has little effect on the evolution of the size distribution function.

(2) Cascade-induced fluctuations dominate the dispersion of the loop and cavity distribution functions under fusion conditions.

(3) Nucleation of helium-filled cavities will likely be continuous because of the re-resolution of helium atoms trapped in He-V clusters. The size distribution function will be asymmetric with a long tail towards smaller sizes.

(4) Spatially organized microstructures are expected to exist under a wide range of irradiation conditions. They result from spatial instabilities in point-defect concentrations in the presence of cascades.

Detailed and careful experiments are needed to measure the effects of continuous cavity nucleation and of cascade fluctuations on the size distribution functions of microstructural features. Specific experiments can also be devised to directly measure the moments of the transition probabilities as defined by eqs. (4) and (5). Such experimental details, when correlated to theory, can enhance our understanding of the underlying physics of microstructure evolution under irradiation and, hence, our extrapolation of current data to anticipated fusion conditions.

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