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DETERMINATION OF THE BIAS FACTOR BY THE MOMENTS SOLUTION TO THE FOKKER–PLANCK EQUATION *

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An important parameter in the rate theory of swelling is the dislocation loop bias factor, Z_i^l, which is a measure of the rate of interstitial atom absorption relative to vacancy absorption at interstitial loops. The Fokker–Planck (F–P) equation is used to describe interstitial loop evolution, with a kinetic nucleation current boundary condition at di-interstitial atomic clusters. The majority of loop nucleation is shown to be finished after one milli-dpa, which allows the shape of the loop distribution function to be governed mainly by the drift (F) and dispersion (D) functions in the F–P equation. Since collision cascades contribute significantly to D, their effects must be suppressed by using low-energy ions or high-energy electrons to produce spatially homogeneous atomic displacements. Under these conditions, both F and D are shown to be proportional to the square root of the number of atoms in a loop. The proportionality parameters depend on material and irradiation conditions, and are linearly proportional to Z_i^l. The ratio F/D, which resembles the Peclet number in fluid flow, can be used in a unique way to determine Z_i^l without the usual complications of uncertainties in material and irradiation conditions. This is shown to constitute an internal variable measurement of the bias factor.

1. Introduction

Interstitial loops form by atomic clustering of self-interstitials during irradiation. Once a small cluster of a few interstitial atoms form, a strain field is set around the atomic cluster. The strain field results in a preferential attraction of interstitials over vacancies. Swelling by vacancy agglomeration into cavities is a consequence of this biasing effect of interstitial loops. Of course, other factors control the magnitude of swelling rate, such as the presence of gas atoms, point defect recombination centers, and other microstructural features (e.g., dislocation network and precipitates). However, dislocation loop bias towards interstitials remains as an important ingredient determining the swelling rate.

Important as it is, no experimental method has ever been proposed to measure the loop bias factor in a direct way. Rate theory has been pursued during the past two decades to predict the swelling rates of various metals and alloys [1–3]. The large number of parameters used in the theory of swelling makes the unique determination of each one uncertain. It is sometimes possible, as is shown in this paper, to discover an experimental method that allows the unique determination of a specific parameter. The method is based on the analysis of the interstitial loop microstructure evolution using a moments solution to the F–P equation.

Systems of non-linear ordinary differential equations have been used (e.g., Ghoniem [4]) to represent the concentrations of hierarchies of atomic clusters of increasing size. Solutions of these systems of equations are not possible without mathematical or numerical approximations. For example, grouping methods, where a group of equations are assigned the same reaction rate, have been introduced by Kiritani [5] and Hayns [6]. In the study of multi-state kinetic transitions, the hierarchy of rate equations is replaced by an equivalent parabolic partial differential equation. The resulting continuum equation is of the F–P type, and it describes a process of particle diffusion in a general drift field. This approach was used to describe vacancy and interstitial atom clustering by Sprague, Russell and Choi [7]. Wolfer, Mansur and Sprague [8], and also by Hall [9].

Recent efforts by Gurol [10], Clement and Wood [11], Trinkaus [12], Kitajima and co-workers [13–16], and by Ghoniem [17] have considered an interpretation of atomic clustering within the framework of statistical mechanics. The F–P equation resulting from Taylor series expansion of rate equations [7–9] is a representation of stochastic size fluctuations produced by single atomic additions. When collision cascades are to be considered, size fluctuations in atomic clusters are dramatically influenced by direct cascade collisions as well as by point-defect arrival produced from random cascades.

An equation for the nucleation of interstitial loops is developed in section 2 which is used as one of the kinetic boundary conditions necessary for the solution of the F–P equation. A moments solution to the one-dimensional (1-D) interstitial-loop F–P equation for general drift and diffusion functions is given in section 3. This is followed by a specific example illustrating a suggested procedure for determining the loop bias factor.

2. Theory and moments solution

The concentrations of single vacancies, C_v, and single self-interstitial atoms, C_i, are given by [4,17]

$$\frac{dC_v}{dt} = \epsilon P - \alpha C_v C_i - \lambda_v C_v, \tag{1}$$

$$\frac{dC_i}{dt} = \epsilon P + K_v(2) C_v C_{2i} + 2\delta C_{2i} - 2K_i(1) C_i^2 - \alpha C_v C_i - K_i(2) C_i C_{2i} - \lambda_i C_i, \tag{2}$$

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where P is the point-defect generation rate, ϵ the cascade survival efficiency, α the recombination rate, δ the di-interstitial dissociation rate, and $K_a^b(x)$ is the reaction rate constant between a mobile specie (a) and an immobile specie (b) containing x atoms. The parameters λ_i and λ_v represent effective loss rates to homogeneous microstructural sinks. Equations for λ_i and λ_v and details of all equations in the paper are given by Ghoniem [17]. In eqs. (1) and (2), vacancy clustering is assumed to be negligible during the early phase of loop formation, and a trapping model is used to describe the effective migration of self-interstitials.

The formation of di-interstitial clusters is governed by the following rate equation for their concentration, C_{2i}:

$$\frac{dC_{2i}}{dt} = J_1 - J_2. \tag{3}$$

The di-interstitial cluster is assumed to be mobile with a homogeneous absorption rate λ_{2i}. J_1 is the net rate of transformation of single interstitial atoms to di-interstitial clusters and J_2 is the net rate of transformation to tri- or tetra-clusters. These are given by

$$J_1 = K_i(1)C_i^2 - \delta C_{2i} - \lambda_{2i}C_{2i}, \tag{4}$$

$$J_2 = K_i(2)C_iC_{2i} + 2K_{2i}(2)C_{2i}^2. \tag{5}$$

When the di-interstitial binding energy is large ($E_{2i}^b \simeq 1$ eV), the backward reaction rates of clusters with $x \geq 2$ are negligibly small [18]. The di-interstitial cluster can then be assumed to be the critical nucleus and transformations to tri- or tetra-clusters are therefore nearly irreversible. With this simplification, eq. (3) can be written as

$$\frac{\partial C^*}{\partial t} = \frac{\partial}{\partial x}J^*. \tag{6}$$

Eq. (6) is coincident with the F–P equation for any size, x, given by

$$\frac{\partial C}{\partial t} + \nabla \cdot \boldsymbol{J} = 0, \tag{7}$$

where the operator $\nabla = \partial/\partial x$ in this case, and the current $\boldsymbol{J}$ is only the 1-D component:

$$\boldsymbol{J} = J_x = FC - \frac{\partial}{\partial x}(DC), \tag{8}$$

where F is a drift function and D a dispersion function. F and D can be estimated to include size fluctuations caused by single atomic transitions, and fluctuations due to cascades (e.g., Kitajima [13,15]).

The zeroeth moment of eq. (7) is obtained by direct integration over the distribution function, $C(x)$, between x^* and the maximum loop size. Therefore

$$\frac{dN}{dt} = J^*, \tag{9}$$

where

$$N(t) = \int_{x^*}^{\infty} C(x)\,dx, \tag{10}$$

N is the total loop density. The boundary and initial conditions for the unique solution of eq. (7) are given by

$$\left.\begin{aligned} &C(\infty, t) = 0 \\ &C(x^*, t) = C_{2i}(t) = C^*(t) \\ &C(x, 0) = 0 \end{aligned}\right]. \tag{11}$$

The average size, $\langle x \rangle$, is obtained by taking the time derivatives of both sides of the equation

$$\langle x \rangle N = \int_{x^*}^{\infty} xC(x, t)\,dx, \tag{12}$$

and substituting eq. (7) in eq. (12), together with the conditions expressed by eq. (11). Expanding the drift functions, $F(x)$, as a Taylor series and using the definition of the rth moment as

$$M_r = \langle (x - \langle x \rangle)^r \rangle, \tag{13}$$

where the symbol $\langle\ \rangle$ is used for averaging over the distribution function, we obtain

$$\frac{d\langle x \rangle}{dt} = F\langle x \rangle + \sum_{k=2}^{\infty} \frac{M_k}{k!}\frac{d^k}{dx^k}(F\langle x \rangle)\,|_{\langle x \rangle} + \frac{D^*C^*}{N} - (\langle x \rangle - x^*)\frac{d}{dt}(\ln N). \tag{14}$$

Performing similar manipulations, and starting from eq. (13), we obtain the following equation for the rth moment:

$$\begin{aligned} \frac{dM_r}{dt} = r(r-1)\Bigg\{ &\delta_{r2}D(\langle x \rangle) \\ &+ \sum_{k-2}^{\infty} \frac{M_k}{k!}\frac{d^k}{dx^k}\left[(x - \langle x \rangle)^{r-2}D\right]\Big|_{\langle x \rangle}\Bigg\} \\ &+ r\Bigg\{ \delta_{r1}F(\langle x \rangle) + \sum_{k=2}^{\infty} \frac{M_k}{k!}\frac{d^k}{dx^k} \\ &\times \left[(x - \langle x \rangle)^{r-1}F\right]\Big|_{\langle x \rangle} \\ &- M_{r-1}\left[F(\langle x \rangle) + \sum_{k=2}^{\infty} \frac{M_k}{k!}\frac{d^k}{dx^k}F\,|_{\langle x \rangle}\right]\Bigg\} \\ &+ r\frac{D^*C^*}{N}\Bigg\{\left[(x^* - \langle x \rangle)^{r-1} - M_{r-1}\right] \\ &+ \frac{d}{dt}(\ln N) \\ &\times \left[(x^* - \langle x \rangle)^r - M_r - rM_{r-1}(x^* - \langle x \rangle)\right]\Bigg\}, \end{aligned}$$

$$r = 2, 3, 4, \ldots . \tag{15}$$

δ_{rm} is the usual Dirac-Delta function. The terms enclosed by the first pair of brackets are ascribed to the effects of the dependence of D on x; those enclosed by the second pair represent distortions in the distribution functions caused by $F(x)$; the third pair give the effects of continuous nucleation on the distribution function.

The distribution function, $C(x, t)$, can be formally reconstructed, albeit not uniquely, from the infinite set of moments.

3. Determination of loop bias factor

The F–P equation for loops is linear, hence the distribution function can be represented by a small set of moment equations. For the purposes of this paper, the moments above the second are assumed to be zero. If experimental conditions are such that cascade-induced size fluctuations are negligible, the functions F and D can be shown [17] to be of the form

$$F(x) = \frac{4}{3}\left(\frac{3}{\pi}\right)^{1/4}\left[\alpha Z_i^l C_i + \gamma - \beta C_v\right] x^{1/2} = g_1 x^{1/2}, \tag{16}$$

$$D = D_s(x) = \frac{2}{3}\left(\frac{3}{\pi}\right)^{1/4}\left[\alpha Z_i^l C_i + \gamma + \beta C_v\right] x^{1/2} = g_2 x^{1/2}, \tag{17}$$

where γ is the vacancy emission rate from the loop, α and β are impingement frequencies for insterstitials and vacancies, respectively, and Z_i^l is the interstitial loop bias factor.

Detailed numerical computations, presented in section 4, show that nucleation effects on the moments are small and that quasi-static conditions are quickly established. Inserting $F(x)$ and $D(x)$ given by eqs. (16) and (17) into the truncated series [eqs. (14) and (15)] gives

$$\frac{d\langle x\rangle}{dM_2} \simeq \frac{g_1[1-(\lambda/2)]}{2g_2\{1-\lambda[1-(2g_1/g_2)]\}}, \tag{18}$$

where $\lambda = M_2/(2\langle x\rangle)^2 \ll 1$ and $g_1/g_2 \ll 1$. At temperatures where vacancy emission is negligible, an approximate quasi-static relationship can be obtained from eq. (18), i.e.,

$$\frac{d\langle x\rangle}{dM_2} \simeq \frac{Z_i^l - Z}{Z_i^l + Z} = S. \tag{19}$$

Eq. (19) indicates that eventually the rate of increase of the average loop size will be proportional to the rate of increase of the second moment. The proportionality constant, S, is thus an internal variable and is independent of all material and irradiation conditions. It should, in principle, be insensitive to defect parameters and can therefore be used to determine Z_i^l. The bias factor for straight dislocations, Z, is close to unity. Hence, once S is experimentally measured, Z_i^l is given by

$$Z_i^l \simeq \frac{1+S}{1-S}. \tag{20}$$

The bias factor Z_i^l has been assumed to be size-independent for simplicity. However, its functional dependence on x can be introduced without loss of generality.

To apply eq. (20), the loop size distribution must be experimentally determined at various irradiation doses. Since the set of equations [eqs. (14) and (15)] is truncated beyond the second moment, it can be shown [17] that the size distribution is approximately given by a propagating Gaussian function of the form

$$C(x, t) \simeq \frac{N}{(2\pi M_2)^{1/2}} \exp\left[-\frac{(x-\langle x\rangle)^2}{2M_2}\right]. \tag{21}$$

The loop number density, N, is shown by many experiments to be nearly independent of the irradiation dose after an initial short transient period. The experimental procedure is to fit the data to eq. (21), thus determining $\langle x\rangle$ and M_2 as functions of dpa. Taking an average values of S can thus uniquely determine Z_i^l without having to know any of the defect parameters!

4. Numerical application of the theory

It seems unlikely that the exact experimental conditions prescribed in this paper have been attempted. Therefore, the theory is applied to available experiments where cascade fluctuations exist. Comparisons between theory and existing experiments are made through parametric variations of defect properties. Hall and Potter [19] carried out a series of experiments where 3 MeV $^{58}Ni^+$ ions were used to bombard Ni–Si samples at 465° C. Their calculated peak displacement rate is 3×10^{-4} dpa/s. Eqs. (1) through (3), (9), and (14) through (15) were numerically integrated, assuming that all moments higher than the second are approximately zero. A standard set of defect parameters was used throughout the calculations [19]; only the effective migration energy of interstitials (E_i^M), the di-interstitial binding energy (E_{2i}^b), the interstitial loop bias factor (Z_i^l), and the ratio of cascade to single transition dispersion functions (D_c/D_s) were treated as free parameters.

An increase in Z_i^l results in a corresponding increase in the average loop diameter (fig. 1); however, the total

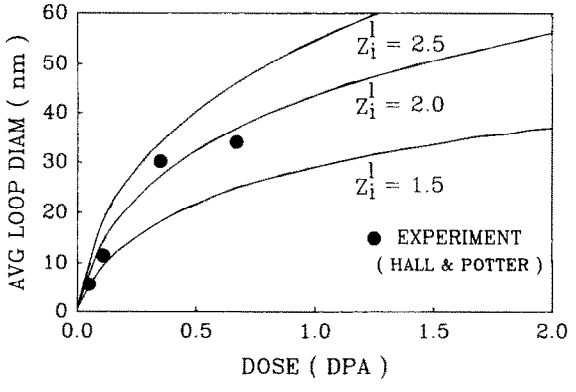

Fig. 1. Effect of Z_i^l on the average diameter of interstitial loops (theory: ———; experiment (Hall and Potter): ●).

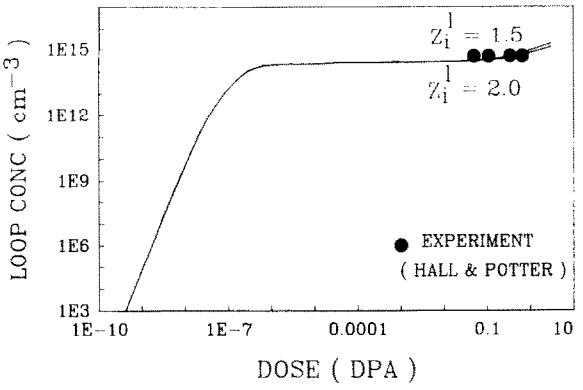

Fig. 2. Effect of Z_i^1 on the interstitial loops (theory: ———; experiment (Hall and Potter): ●).

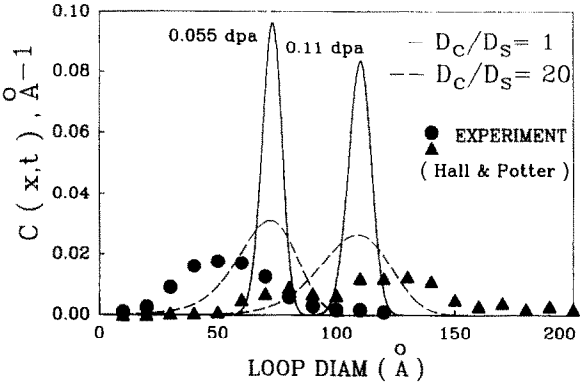

Fig. 4. Effect of cascade-induced fluctuations on the loop distribution function (Hall and Potter).

loop density is insensitive to variations in the bias factor (fig. 2). This interesting feature can be used in determining groups of defect parameters by comparison to kinetic data on loop densities. When E_{2i}^b is increased, the loop density increases dramatically (fig. 3) while the average loop size decreases because of the near-conservation of total number of atoms in loops, Variations in E_i^m produce opposite effects on the loop density and average size.

Comparison with the experimental data of Hall and Potter [19] shows that single-atom transitions may be an underestimation of the magnitude of stochastic fluctuations, and that collision cascade effects must be included. For statistically uncorrelated events, we take $D = D_c + D_s$, where D_c is for cascade fluctuations, and D_s for single-atom transitions. By a parametric increase of D_c/D_s, a closer comparison with experiments is obtained, as shown in fig. 4. The effects of collision cascades on the re-solution of loops and on point-defect concentrations can be calculated in a manner similar to that done by Kitajima [13–16] and Chou and Ghoniem [20,21].

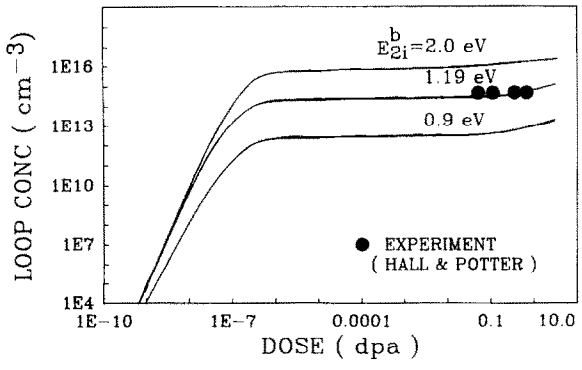

Fig. 3. Effect of E_{2i}^b on the interstitial loop density (theory: ———; experiment (Hall and Potter: ●).

5. Conclusions

The moments solution to the F–P equation, with transient nucleation conditions, can be effectively used to measure the interstitial loop bias factor Z_i^1. To take advantage of this solution, a number of experimental conditions must be satisfied. These conditions are: (1) Irradiation doses beyond a short transient period of ~ 0.001 dpa and before loop unfaulting at several dpa's; (2) Sample temperatures low enough such that vacancy emission from loops does not significantly contribute to their growth (i.e., below 450–500 °C for Ni at a dose rate of $\gtrsim 10^{-4}$ dps/s); (3) The use of low energy ions or high energy electrons to minimize the effects of collision cascades on the fluctuations in loop sizes during their growth.

Comparison between the current theory and experiments show that a combination of the parameters Z_i^1, E_{2i}^b, E_i^M, and D_c/D_s can produce all observed features of interstitial loop evolution in ion-irradiated nickel. The best set of these parameters is found to be $Z_i^1 = 2$, $E_{2i}^b = 1.19$ eV, $E_i^M = 0.55$ eV, and $D_c/D_s = 60$.

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