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# THE EFFECTS OF IMPURITY TRAPPING ON IRRADIATION-INDUCED SWELLING AND CREEP

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ABSTRACT

A general theory of the effects of point defect trapping on radiation-induced swelling and creep deformation rates is developed. The effects on the fraction of defects recombining, and on void nucleation, void growth and creep due to the separate processes of dislocation climb-glide and dislocation climb (the so-called SIPA mechanism) are studied. Trapping of vacancies or interstitials increases total recombination and decreases the rates of deformation processes. For fixed trapping parameters, the reduction is largest for void nucleation, less for void growth and creep due to dislocation climb-glide, and least for creep due to dislocation climb. With this formation, the effects of trapping at multiple vacancy and interstitial traps and of spatial and temporal variation in trap concentrations may be determined. Alternative pictures for viewing point defect trapping in terms of effective recombination and diffusion coefficients are derived. It is shown that previous derivations of these coefficients are incorrect. A rigorous explanation is given of the well-known numerical result that interstitial trapping is significant only if the binding energy exceeds the difference between the vacancy and interstitial migration energies, while vacancy trapping is significant even at small binding energies. Corrections which become necessary at solute concentrations above about 0.1% are described. Numerical results for a wide range of material and irradiation parameters are presented.

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## 1. INTRODUCTION

The earliest experimental studies to discover the characteristics of irradiation-induced void swelling revealed that impurity and alloy additions in a metal strongly affect its propensity to swell. It was soon hypothesized that trapping of point defects by solutes could increase the fraction of vacancies and interstitials undergoing mutual recombination and thus reduce the fraction flowing to voids [1]. The development of this concept has been the subject of several recent papers [2–6]. Somewhat later the conceptually separate effects of changes in void capture efficiencies for point defects and the consequent changes in swelling due to solute segregation at these sinks were studied [7,8]. Both modes of impurity action are now known to be important.

Towards the long-range goal of incorporating both modes of impurity action into a unified picture, the purpose of the present paper is to develop a comprehensive theory to describe the effects of solute trapping upon radiation-induced deformation processes. Derivations are given together with results based upon computations using parameter ranges of interest.

Section 2 describes the formulation of the problem. Included are alternative pictures for viewing the effects of point defect trapping using effective diffusion and recombination coefficients. Corrections to the theory which become necessary at higher solute concentrations are described. Results for wide ranges of temperature, dose rate, dose, solute content and solute binding energy are presented in Section 3. In Section 4 the paper concludes with a summary and discussion of implications for experimental work with suggestions for further development.

## 2. THEORY

The equations describing conservation of free point defects [9] must be generalized to include point defect trapping. This is accomplished by the straightforward logic of subtracting and adding terms corresponding to trapping and recombination at traps, and release of point defects from traps. Additional equations describing conservation of the new entities, trapped defects, are also required.

## 2.1 General Rate Equations

The general system of equations governing point defect conservation is given below.

Free Vacancies

$$\frac{\partial C_v}{\partial t} = \nabla\cdot\left(D_v \nabla C_v + \frac{D_v C_v}{kT}\nabla U_v\right) + G_v + \sum_\ell \tau_{v\ell}^{-1} C'_{v\ell} - RC_i C_v - C_v \sum_\ell R_{i\ell} C'_{i\ell}$$

$$- C_v \sum_\ell \kappa_{v\ell}\left(C_\ell^t - C'_{v\ell} - C'_{i\ell}\right) - K_v C_v \quad . \qquad (1)$$

Free Interstitials

$$\frac{\partial C_i}{\partial t} = \nabla\cdot\left(D_i \nabla C_i + \frac{D_i C_i}{kT}\nabla U_i\right) + G_i + \sum_\ell \tau_{i\ell}^{-1} C'_{i\ell} - RC_i C_v - C_i \sum R_{v\ell} C'_{v\ell}$$

$$- C_i \sum_\ell \kappa_{i\ell}\left(C_\ell^t - C'_{v\ell} - C'_{i\ell}\right) - K_i C_i \quad . \qquad (2)$$

In the above equations, the summations extend over the $\ell = 1,2, \ldots, p$ types of traps. Similarly, the following equations apply to each of the $\ell = 1,2, \ldots, p$ types of traps.

Traps (concentration in general a function of position and time)

$$\frac{\partial C_\ell^t}{\partial t} = f_\ell(\underset{\sim}{r},t) \qquad (3)$$

Trapped Vacancies

$$\frac{\partial C'_{v\ell}}{\partial t} = C_v \kappa_{v\ell}\left(C_\ell^t - C'_{v\ell} - C'_{i\ell}\right) - \tau_{v\ell}^{-1} C'_{v\ell} - C_i R_{v\ell} C'_{v\ell} \qquad (4)$$

Trapped Interstitials

$$\frac{\partial C'_{i\ell}}{\partial t} = C_i \kappa_{i\ell}\left(C^t_\ell - C'_{v\ell} - C'_{i\ell}\right) - \tau^{-1}_{i\ell}\, C'_{i\ell} - C_v R_{i\ell} C'_{i\ell} \quad . \qquad (5)$$

The left-hand sides of all these equations represent the time rates of change of the corresponding concentration. The first terms in Eqs. (1) and (2) describe diffusion and drift of free point defects due to the presence of discrete sinks such as free surfaces, the G's describe production of point defects by radiation and thermal emission from sinks, and the terms in $\tau^{-1}$ describe release of point defects from traps. The remaining four terms in Eqs. (1) and (2) describe point defect losses due to recombination with opposite free defects, opposite trapped defects, trapping at impurities and absorption at sinks. Equations (4) and (5) describe conservation of trapped defects generated by trapping reactions at impurities and lost through release due to detrapping and by recombination with opposite free defects.

Subscripts i and v denote interstitials and vacancies. $C_{i,v}$ denotes the concentration of free interstitials, vacancies. These equations allow for trapping of both interstitials and vacancies at multiple traps. Thus, for example, $C'_{i\ell}$ denotes the population of trapped interstitials at trap type $\ell$, the concentration of which is denoted by $C^t_\ell$. An obvious analog notation is used for vacancies. $R = 4\pi r_0\left(D_i + D_v\right)$ denotes the recombination coefficient of free vacancies with free interstitials, where $r_0$ is the radius of recombination volume and D denotes diffusion coefficient. Symbols of the type $R_{i\ell} = 4\pi r_{i\ell} D_v$ or $R_{v\ell} = 4\pi r_{v\ell} D_i$ denote the recombination coefficient of free vacancies with trapped interstitials at trap type $\ell$, and free interstitials with trapped vacancies at trap type $\ell$, respectively; $r_{i\ell}$ and $r_{v\ell}$ denote the corresponding radii of the recombination volumes. The mean time a defect is trapped at trap type $\ell$ is denoted by $\tau_{\alpha\ell} = \left(b^2/D_\alpha^0\right) \exp[\left(E_{\alpha\ell}^b + E_\alpha^m\right)/kT]$ where $\alpha$ denotes i or v, $D_\alpha^0$ is the pre-exponential of the diffusion coefficient $[D_\alpha = D_\alpha^0 \exp\left(-E_\alpha^m/kT\right)]$, b is the order of an atomic distance, $E_{\alpha\ell}^b$ is the binding energy of the point defect at the $\ell$-th type trap, $E_\alpha^m$ is the point defect migration energy, k is Boltzmann's constant, and T is absolute temperature. We obtain the pre-exponential, $b^2/D_\alpha^0$, by realizing that when $E_\alpha^b \rightarrow 0$ then $\tau$ approaches the time for an ordinary diffusion step, b/(D/b). $\kappa_{v\ell} = 4\pi r'_{v\ell} D_v$ and $\kappa_{i\ell} = 4\pi r'_{i\ell} D_i$ denote capture coefficients similar to recombination coefficients. These describe capture of a free vacancy and of a free interstitial, respectively, at trap type $\ell$. $r'_{v\ell}$ and $r'_{i\ell}$ are the capture radii of the trap type $\ell$ for the point defects. In the terms of the type containing $\kappa_{v\ell}$ the concentration difference $\left(C_\ell^t - C'_{v\ell} - C'_{i\ell}\right)$ appears. This accounts for the fact that the fraction $C'_{v\ell}/C_\ell^t + C'_{i\ell}/C_\ell^t$ traps of type $\ell$ are already occupied by vacancies and interstitials and hence not available as traps for free defects. This feature also accounts for the fact that a given trap may have a binding energy for both vacancies and interstitials but would not trap both simultaneously, since when one is trapped the site is then a recombination center. $K_\alpha$ is the rate per defect for absorption at all internal sinks such as voids, dislocations, dislocation loops, etc. $\left(\text{i.e., } K_\alpha = \sum_j K_\alpha^j,\right.$ where j represents each individual type sink$\left.\right)$.

We solve Eqs. (1) through (5) for the free vacancy and interstitial concentrations, $C_v$ and $C_i$, by methods to be discussed later. Once having these, the void nucleation and growth rates and the irradiation creep rate may be determined by using the relationships summarized below. First, however, we describe alternative pictures for viewing the effects of trapping in the following three sections.

For simplicity we ignore spatial and time derivatives in Eqs. (1) through (5), corresponding to a quasi-steady state far from discrete sinks, and consider only one vacancy and one interstitial trap. Equations (1), (2), (4), and (5) become

$$G_v + \tau_v^{-1} C'_v - RC_iC_v - C_vR_iC'_i - C_v\kappa_v \left(C^t - C'_v - C'_i\right) - K_vC_v = 0 \ , \tag{6}$$

$$G_i + \tau_i^{-1} C'_i - RC_iC_v - C_iR_vC'_v - C_i\kappa_i \left(C^t - C'_v - C'_i\right) - K_iC_i = 0 \ , \tag{7}$$

$$C_v\kappa_v\left(C^t - C'_v - C'_i\right) - \tau_v^{-1}C'_v - C_iR_vC'_v = 0 \ , \tag{8}$$

$$C_i\kappa_i\left(C^t - C'_v - C'_i\right) - \tau_i^{-1}C'_i - C_vR_iC'_i = 0 \ . \tag{9}$$

## 2.2 Alternative Form

If we add Eq. (9) to (7) and Eq. (8) to (6), we obtain

$$G_i - C_i\left(RC_v + R_vC'_v\right) - K_iC_i - C_vR_iC'_i = 0 \ , \tag{10}$$

$$G_v - C_v\left(RC_i + R_iC'_i\right) - K_vC_v - C_iR_vC'_v = 0 \ . \tag{11}$$

These equations describe conservation of the total (free plus trapped) populations of interstitials and vacancies, respectively. Smidt and Sprague [2], taking $C'_i = 0$ since they did not consider trapped interstitials, used the equivalent of Eqs. (10), (11), and (8) to study the effects of vacancy trapping on void nucleation.

## 2.3 Effective Recombination Coefficient

Substituting into Eqs. (10) and (11) the expressions for $C_i'$ in terms of $C_i$ and $C_v'$ in terms of $C_v$ given by Eqs. (8) and (9), we obtain

$$G_i - R^{ef} C_i C_v - K_i C_i = 0 \quad , \tag{12}$$

$$G_v - R^{ef} C_i C_v - K_v C_v = 0 \quad , \tag{13}$$

where

$$R^{ef} = R + \left(C^t - C_v' - C_i'\right) \left[\frac{R_v \kappa_v \tau_v}{1 + \tau_v C_i R_v} + \frac{R_i \kappa_i \tau_i}{1 + \tau_i C_v R_i}\right] \quad . \tag{14}$$

Equations (12) and (13) are identical to the conservation equations with no point defect trapping [9], except that $R^{ef}$ now appears rather than R.* This effective coefficient, however, is a function of the free and trapped point defect populations and thus we have at best only introduced a simplification over Eqs. (6) through (9) in our mental picture but not in the mathematical solution. This type of simplification was pointed out by Koehler [4] and Okamoto et al. [5] in their treatments of trapping at immobile traps. However, their effective recombination coefficient expressions corresponding to our Eq. (14) are incorrect. This stems from their neglect of the last terms in our Eqs. (8) and (9), though they include them in their free defect equations corresponding to our Eqs. (6) and (7) [cf. Eq. (29) in ref. 4 and Eq. (8) in ref. 5].

In Fig. 1 the ratio of the effective recombination coefficient given by Eq. (14) to that given in ref. 5 is plotted for interstitial

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*Of course the free defect concentrations, $C_i$ and $C_v$, are also different. Thus the ratio $R^{ef}/R$ alone does not measure the increase in the number of defects recombining in the matrix.

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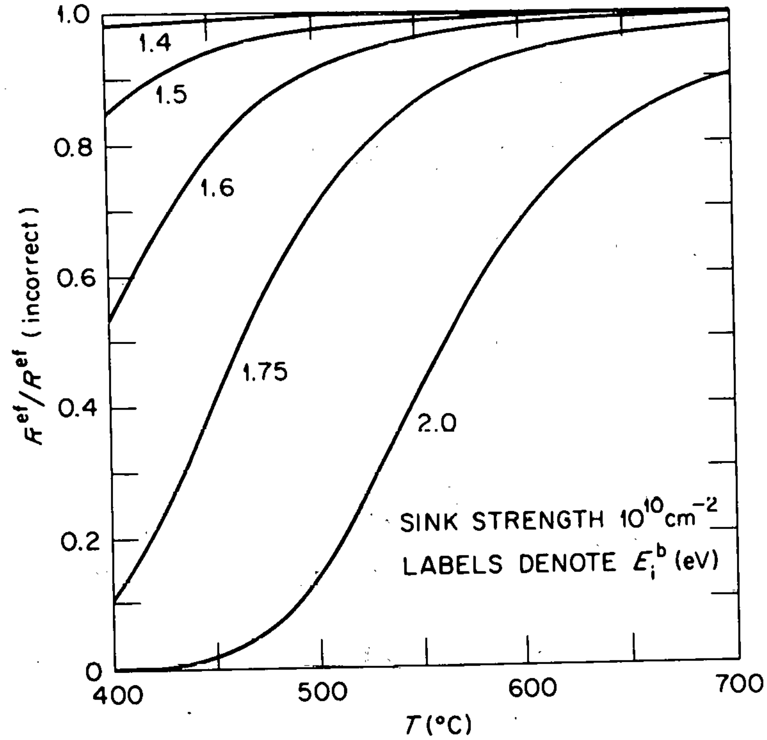


Fig. 1. The ratio of the correct effective recombination coefficient with interstitial trapping to that obtained by neglecting the recombination of trapped interstitials with free vacancies versus temperature.

trapping. It can be seen that when trapping is important refs. 4 and 5 significantly overestimate the effective recombination coefficient.

## 2.4 Effective Diffusion Coefficient

In a similar spirit we may visualize an effective diffusion coefficient. Here the diffusivity of the entire population of defects, trapped plus free, is characterized by an effective diffusion coefficient, $D^{ef}$, although only free defects are moving and these with the usual diffusivity, $D_i$ or $D_v$. Then, by definition,

$$D_i C_i = D_i^{ef} \left(C_i + C_i^t\right) \quad , \tag{15a}$$

and

$$D_v C_v = D_v^{ef} \left(C_v + C_v'\right) \quad . \tag{15b}$$

Using these equations together with Eqs. (8) and (9) gives

$$D_i^{ef} = D_i/\left(1 + \kappa_i \tau_i (C^t - C_v' - C_i')/(1 + \tau_i C_v R_i)\right) \quad ; \tag{16a}$$

$$D_v^{ef} = D_v/\left(1 + \kappa_v \tau_v (C^t - C_v' - C_i')/(1 + \tau_v C_i R_v)\right) \quad . \tag{16b}$$

The effective diffusion coefficient picture was invoked by Schilling and Schroeder [3]. However, their effective diffusion coefficients are incorrect and correspond to replacing the terms in parentheses at the end of Eqs. (16a) and (16b), with unity. As in refs. 4 and 5, this again amounts to neglecting recombination between free defects and the opposite trapped defects, the third terms in Eqs. (8) and (9), the trapped defect conservation equations.

In Fig. 2 the ratio of the effective diffusion coefficient given in ref. 3 to that given by Eq. (16a) is plotted. Clearly when trapping is important the previous results significantly underestimate the effective diffusion coefficients.

## 2.5 Asymmetry in Effects of Interstitial and Vacancy Trapping

In results to be presented later it is found that vacancy trapping with binding energies of the order of 0.1 eV is effective in reducing swelling, but that interstitial trapping is effective only when the binding energy is greater than the difference between vacancy and interstitial migration energies, $\sim$1.2 eV for nickel using the parameters we have adopted. This result may be anticipated by the following argument based on effective diffusion coefficients. Considering only interstitial

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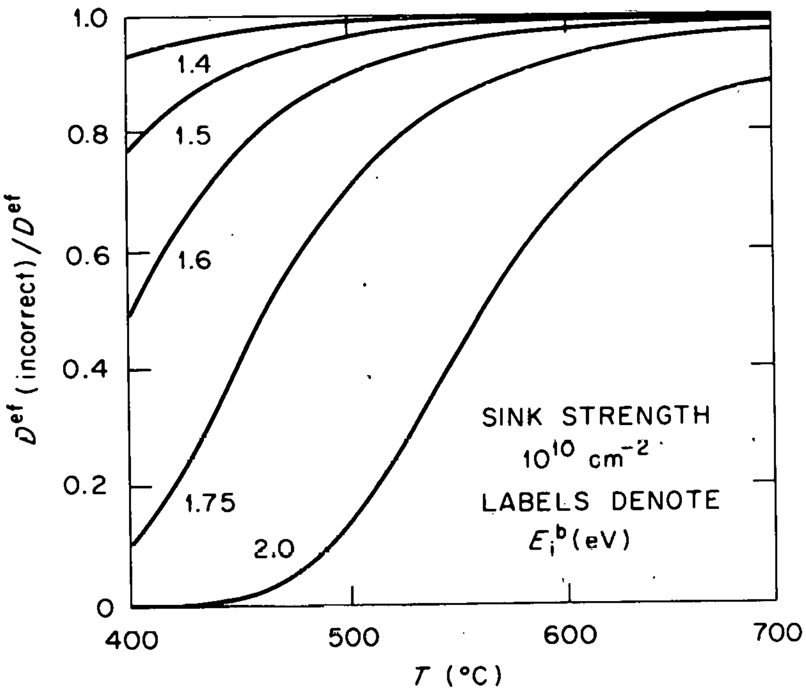


Fig. 2. The ratio of the effective diffusion coefficient of interstitials with interstitial trapping obtained by neglecting recombination of trapped interstitials with free vacancies to the correct effective diffusion coefficient versus temperature.

trapping $(r_v = r_v' = C_v' = 0)$ and adding Eq. (9) to (7), Eqs. (6) through (9) become simply

$$G_i - RC_iC_v - C_vR_iC_i' - K_iC_i = 0 \quad ; \tag{17}$$

$$G_v - RC_iC_v - C_vR_iC' - K_vC_v = 0 \quad . \tag{18}$$

By definition $R = 4\pi r_0(D_i + D_v)$ and $R_I = 4\pi r_I D_v$. For simplicity take $r_0 \sim r_I$. If $r_0 \neq r_i$ the conclusion still applies though the exact form of the equations changes slightly. Equations (17) and (18) can then be rewritten

$$G_i - 4\pi r_0\, D_iC_iC_v - 4\pi r_0\, D_vC_v\,(C_i + C_i') - K_iC_i = 0 \;; \tag{19}$$

$$G_v - 4\pi r_0\, D_iC_iC_v - 4\pi r_0\, D_vC_v\,(C_i + C_i') - K_vC_v = 0 \;, \tag{20}$$

or using definition (15a)

$$G_i - R'\left(C_i + C_i'\right) C_v - K_i C_i = 0 \quad ; \qquad (21)$$

$$G_v - R'\left(C_i + C_i'\right) C_v - K_v C_v = 0 \quad ; \qquad (22)$$

where

$$R' = 4\pi r_0 \left(D_i^{ef} + D_v\right) \ . \qquad (23)$$

$G_i$ and $G_v$ are the generation rates of free defects, but this is negligibly different from the generation rate of total defects for solute concentrations $\lesssim$0.1% (see our later discussion of higher order effects). Similarly, the sink loss rate terms of the form $K_i C_i$ describe losses of total (free plus trapped) defects to the sinks since we have assumed that the trapped defects are immobile. Thus, Eqs. (21) and (22) are the correct expressions for conservation of the total interstitial and vacancy populations. These are similar to the conservation equations with no traps [9], except that R′ replaces R or equivalently $D_i^{ef}$ replaces $D_i$ in those equations. Thus, any result based on the formulation with no trapping will be correct in the presence of interstitial trapping if the diffusion coefficient for interstitials, $D_i$, is now replaced with $D_i^{ef}$. However, from Eq. (16a) we see that $D_i^{ef}$ is approximately proportional to $D_i \exp\left(-E_i^b/kT\right) = D_i^0 \exp[-\left(E_i^m + E_i^b\right)/kT]$. Thus, unless $E_i^m + E_i^b$ approaches $E_v^m$, we may replace $D_v$ with zero in Eq. (23). This results in no effect of interstitial trapping and justifies our previous mention of the effect of interstitial trapping on temperature shift with dose rate in terms of effective diffusion coefficient [9]. This is because when we can neglect $D_v$ in comparison to $D_i^{ef}$, then $D_i^{ef}$ cancels out of the expressions for void formation and temperature shift [9].

## 2.6 Swelling and Creep

Based on the extension of classical nucleation theory to an irradiation environment developed by Katz and Wiedersich [10] and by Russell [11], we may derive the following expressions [8]

$$\Delta G(n) = -kT \sum_{\ell=1}^{n-1} \ln\left\{\frac{\ell^{1/3} Z_v^V(\ell) D_v C_v}{(\ell+1)^{1/3}\left[Z_i^V(\ell+1) D_i C_i + Z_v^V(\ell+1) D_v C_v^e(\ell)\right]}\right\} ; \quad (24)$$

$$M = \frac{2(6\pi^2\Omega)^{1/3} D_v C_v^2}{\sum_{n=1}^{\infty} \frac{\exp[\Delta G(n)/kT]}{n^{1/3} Z_v^V(n)}} . \quad (25)$$

In these equations $\Delta G(n)$ is the free energy of formation of a vacancy cluster containing n vacancies and M is the nucleation rate (i.e., the number per unit time of vacancy clusters that grow past the critical size). $Z_{i,v}^V$ denotes the capture efficiency for interstitials or vacancies of a void containing n vacancies.

The growth of voids or interstitial dislocation loops is described by the net flux of vacancies or interstitials. The expressions may be written as follows [9]

$$\frac{dr_V}{dt} = \frac{\Omega}{r_V}\left\{Z_v^V(r_V) D_v\left[C_v - C_v^e(r_V)\right] - Z_i^V(r_V) D_i C_i\right\} ; \quad (26)$$

$$\frac{dr_L}{dt} = a^2\left\{Z_i^L(r_L) D_i C_i - Z_v^L(r_L) D_v\left[C_v - C_v^e(r_L)\right]\right\} , \quad (27)$$

where $r_V$ and $r_L$ are the void and loop radii, and $\Omega$ denotes atomic volume.

$$C_v^e(r_V) = C_v^e \exp\left[-\left(p - \frac{2\gamma}{r_V}\right) \Omega/kT\right] \quad (28)$$

is the thermal equilibrium vacancy concentration near a void of radius $r_V$ with a surface free energy $\gamma$ and containing a gas at pressure p.

$$C_v^e(r_L) = C_v^e \exp\left[-\left(\gamma_f + E_L\right) a^2/kT\right] \tag{29}$$

is the thermal equilibrium vacancy concentration near an interstitial loop of radius $r_L$, where $\gamma_f$ is the stacking fault energy, $E_L$ the elastic energy, and a denotes a lattice dimension.

$$C_v^e = \Omega^{-1} \exp\left(S_v^f/k\right) \exp\left(-E_v^f/kT\right) \tag{30}$$

is the bulk thermal vacancy concentration where $S_v^f$ and $E_v^f$ are the entropy and energy of vacancy formation.

There are a number of proposed mechanisms for irradiation creep. Two mechanisms giving order-of-magnitude agreement with experimental data have received more detailed attention recently. Since these two mechanisms may act simultaneously and show different dependences on parameters of interest, we shall investigate the effects of impurity trapping on both. Stress-induced preferred absorption (SIPA) [12–14] results from the induced inhomogeneity interaction between a point defect and a dislocation. This interaction varies with the orientation of the dislocation Burgers vector with respect to the applied stress direction* and results in a greater flux of point defects to those dislocations having Burgers vectors near parallel to the stress direction. This can be expressed in terms of the free defect concentrations $C_i$ and $C_v$ as [15]

$$\dot{\varepsilon} = \frac{2\Omega L}{9}\left(\Delta Z_i^d D_i C_i - \Delta Z_v^d D_v C_v\right) \tag{31}$$

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*For clarity, we consider only simple tensile stresses in this paper.

where $\dot{\varepsilon}$ is the strain rate, L is the dislocation density, and $\Delta Z_i^d$ or $\Delta Z_v^d$ denotes the difference between the capture efficiencies for point defects of dislocations with Burgers vectors parallel and perpendicular to the applied stress direction and can be approximated by

$$\Delta Z_{i,v}^d = \frac{3(1-\nu)\left\{2\pi/\ell n\left[2R_d\Big/\left|\frac{(1+\nu)\mu b\ \Delta V_{i,v}}{3\pi(1+\nu)kT}\right|\right]\right\}^2 \varepsilon\ \ V_{i,v}a_{i,v}}{2\pi(1+\nu)\ \Delta V_{i,v}}, \quad (32)$$

where $\nu$ is Poisson's ratio, $R_d = (\pi L)^{-1/2}$, $\mu$ is the shear modulus, b the Burgers vector, $\Delta V$ the relaxation volume, $\varepsilon$ the applied strain equals $\sigma/E$ where $\sigma$ is the applied stress and E is Young's modulus. V is the effective inclusion volume of the defect and

$$a_{i,v} = \frac{15(1+\nu)\ \Delta\mu_{i,v}}{15(1-\nu)\ \mu + 2(4-5\nu)\ \Delta\mu_{i,v}}$$

where $\Delta\mu$ is the difference in shear moduli of the matrix and the effective modulus of the point defect.

The climb-glide mechanism for irradiation creep [15,16] may be superposed on the SIPA contribution. This mechanism would operate only if there were other sinks such as voids in addition to the climbing dislocations to allow an asymmetrical partitioning of point defects between types of sinks. The excess interstitials absorbed at dislocations would then enable them to climb past an obstacle which would otherwise have allowed only a limited bowing out in the slip plane. The creep rate by this mechanism is described by

$$\dot{\varepsilon} = (\pi L)^{1/2}\ \varepsilon a^2\left[Z_i^d D_i C_i - Z_v^d D_v\left(C_v - C_v^e\right)\right] \quad (33)$$

where the $Z^d$ are average dislocation capture efficiencies for point defects. Note that the climb-glide creep rate is proportional to the void growth rate since the net excess of vacancies going to voids [Eq. (26)] must just equal the net excess of interstitials going to dislocations [Eq. (33)] if these are the only sinks.

To complete the picture we must include diffusional creep which will be important at high temperatures and results from the different probabilities of emission of vacancies from dislocations oriented parallel and perpendicular to the applied stress axis.

$$\dot{\varepsilon} = \frac{2\Omega L\ D_v C_v^e Z_v^d}{9}\left[\exp(\sigma\Omega/kT) - 1\right] \quad . \tag{34}$$

## 2.7 High Solute Concentrations

As the solute concentration exceeds the order of 0.1%, Eqs. (1) through (5) or (6) through (9) become only approximate, with the degree of approximation becoming worse as the solute concentration is increased. To illustrate, we write the form to which Eq. (8) reduces in the absence of radiation

$$\frac{C_v^{e'}}{C_v^e} = \tau_v \kappa_v \left(C^t - C_v^{e'}\right) \tag{35}$$

where $C_v^{e'}$ is the thermal vacancy population trapped on vacancy traps and $C_v^e$ is the free thermal vacancy population. However, it is possible to independently determine the correct ratio of trapped to free vacancies from equilibrium statistics [17].

For fcc metals,

$$C_v^{e'} = 12\ C^t \exp\left(S_v^f/k\right) \exp\left(-S_v^b/k\right) \exp\left[-\left(E_v^f - E_v^b\right)/kT\right] \quad (36)$$

and

$$C_v^e = \left[\Omega^{-1} - C_v^{e'} - 12\ C^t\left(1 - C^{e'}/C^t\right)\right] \exp\left(S_v^f/k\right) \exp\left(-E_v^f/kT\right) \quad . \quad (37)$$

Here $S_v^f$ and $E_v^f$ are the entropy and energy of free vacancy formation, $E_v^b$ is the vacancy binding energy to the trap, and $S_v^b$ is the change in entropy due to binding. It is obvious from Eq. (36) that $C_v^{e'} \ll C^t$ if $E_v^b \lesssim 0.5$ eV, i.e., even with a rather high vacancy binding energy, since $E_v^f$ is $\gtrsim 1$ eV in nickel and other metals of interest. Under this condition Eq. (35) may be written

$$\frac{C_v^{e'}}{C_v^e} = \tau_v \kappa_v C^t \quad (38)$$

and we take the ratio of Eqs. (36) to (37) assuming $S_v^b/k \ll 1$.

$$\frac{C_v^{e'}}{C_v^e} = \frac{12\ \Omega\ C^t \exp\left(E_v^b/kT\right)}{\left(1 - 12\ C^t\Omega\right)} \quad . \quad (39)$$

Comparing the correct expression (39) to the approximate expression (38) and recalling that $\tau_v = \frac{b^2}{D_v^0} \exp\left[\left(E_v^m + E_v^b\right)/kT\right]$ and $\kappa_v = 4\pi r_v D_v^0 \exp\left(-E_v^m/kT\right)$ we may establish that (38) is equivalent to (39) $\left(12\ \Omega \simeq 4\pi r_v b^2\right)$ except that the factor $\left(1 - 12\ C^t\ \Omega\right)$ is missing from Eq. (38). Thus, if $C^t\ \Omega \sim 0.1\%$, then Eq. (38), or as a consequence, the sets of Eqs. (1) through (5) or (6) through (9), are inaccurate by approximately 1%. By including this additional factor for site exclusion, we could extend the range of validity of the trapping model to cover several percent solute if no additional corrections were necessary.

Recently, however, Brailsford and Bullough [6] have found that effectively the trapping radius $r_{v\ell}$ or $r_{i\ell}$ must be corrected upward for higher solute concentration by a factor $(1 + k'r)$. This multiple-sink or correlation correction would result from the interference of the diffusion fields of the traps, if such traps were diffusion controlled. We may approximate $k'$ by $\sqrt{4\pi r_i C^t}$ for vacancy trapping on one type of trap. Taking $r_i \sim 10^{-8}$/cm and $C^t \sim 10^{20}/\mathrm{cm}^3$, the correction factor is approximately 1.03. This correction is larger than the site exclusion correction discussed above although of the same order of magnitude.

## 2.8 Methods of Solution

Numerical methods have been employed to solve for the concentrations of point defects using Eqs. (6) through (9) or their corresponding generalized forms, Eqs. (1) through (5). Then, from Eqs. (24) through (34) the void swelling and irradiation creep behavior are determined.

For studying the effects of vacancy and interstitial trapping separately, the set of Eqs. (6) through (9) reduces to three simplified equations. These can be combined to yield a cubic equation for $C_i$ or $C_v$. For vacancy trapping for example $\left(r_i = r_i' = C_i' = 0\right)$ we obtain

$$K_iR_iR\tau_v\, C_i^3 + \left[RR_i\tau_v\left(G_v - G_i\right) + K_vK_iR_i\tau_v + K_iR_i\tau_v\kappa_v\left(C^t - C_v'\right) + K_iR\right]C_i^2$$
$$+ \left[R_i\kappa_v\tau_v\left(C^t - C_v'\right) + R\right]\left(G_v - G_i\right) + K_iK_v - G_iR_i\tau_vK_v\, C_i - G_i = 0 \quad . \tag{40}$$

Where $C_v'/C^t << 1$, the difference $\left(C^t - C_v'\right)$ in Eq. (40) can be approximated by $C^t$. Under these conditions Eq. (40) can be solved iteratively for $C_i$ using Newton's method. Once having $C_i$, then $C_v$ and $C_v'$ can be obtained from Eqs. (8) and (7). Figure 3 shows $C_v'/C^t << 1$ for typical reactor and ion bombardment conditions.

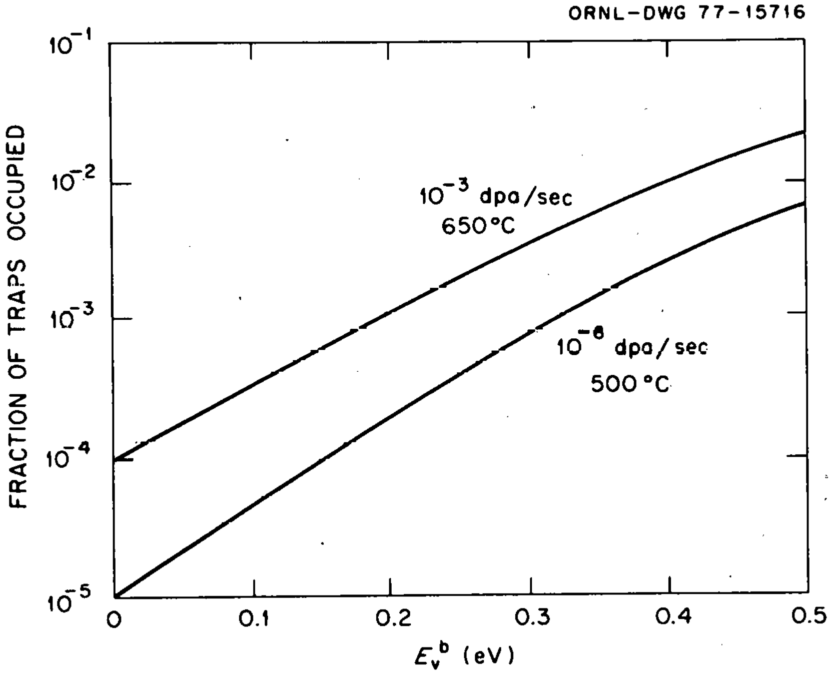


Fig. 3. The fraction of traps occupied for vacancy trapping at typical charged particle and fast reactor dose rates and temperatures versus binding energy. Under these conditions, the fraction of traps occupied is small even at high vacancy binding energies.

More powerful numerical methods have been used to solve the more general set of conservation equations [(1) through (5)] by extending the numerical method developed earlier [18]. These techniques are of particular advantage when the surfaces of a thin foil or ion bombardment specimen must be taken into account, when the effects of multiple vacancy and/or interstitial traps are to be considered or when the fraction of occupied traps is not negligible. The general method is described elsewhere [19].

The cumulative void swelling and creep strain may be determined by numerical integration accomplished by computing the C's (as above), computing the corresponding deformation rates, allowing the microstructure to change at these rates for a short time increment, and then recomputing the C's in response to the new microstructure and so on.

## 3. RESULTS

The effects of point defect trapping utilizing two measures of interest will be presented. These are the effects upon the fraction of point defects undergoing recombination in the bulk, and ultimately upon void nucleation and growth and irradiation creep. For the results discussed in this section, the parameters used are nominally those for nickel. In the cases considered, the largest vacancy binding energy is 0.5 eV and the largest interstitial binding energy is 1.7 eV. The results for these energies and for no trapping form an envelope which is expected to give reasonable upper and lower limits for the possible effects of point defect trapping. Table I gives the parameter values used in all of the calculations unless stated otherwise on the figures or in the figure captions. The values of the remaining parameters necessary for these calculations are given on the figures.

Figures 4, 5, and 6 show the fraction of defects recombining in the matrix, i.e., not being removed at sinks, as functions of temperature, dose rate, and solute concentration. In Fig. 4, the results for two different sink strengths, $10^{10}$ $cm^{-2}$ and $5 \times 10^{11}$ $cm^{-2}$, are shown corresponding roughly to pure nickel and cold-worked stainless steel at moderate doses. In addition to illustrating the effects of point defect trapping these curves also show that even at low reactor dose rates, point defect recombination would be important for pure nickel. Figure 5 shows the effects of trapping on point defect recombination with dose rate

TABLE I

Parameter Values Used in Obtaining Results Given in the *Results* Section. Additional parameters required and any changes in the parameters of this table are indicated on the figures themselves.

| Parameter | Value |
|---|---|
| Sink strength, $cm^{-2}$ | $5 \times 10^{10}$ |
| Vacancy diffusivity, $cm^2/s$ | $1.4 \times 10^{-2}$ exp(−1.38 eV/kT) |
| Interstitial diffusivity, $cm^2/s$ | $8 \times 10^{-3}$ exp(−0.15 eV/kT) |
| Dislocation-interstitial preference, $Z_i^d/Z_v^d$ | 1.1/1 |
| Generation rate, dpa/s | $10^{-3}$ |
| Impurity concentration, atom/atom | $10^{-3}$ |
| Equilibrium vacancy concentration, $cm^{-3}$ | $0.91 \times 10^{23}$ exp(1.5) exp(−1.4 eV/kT) |
| Surface free energy, $ergs/cm^2$* | 700 |
| Injected/generated interstitials | $4 \times 10^{-4}$ |

*The void nucleation rate is sensitive to the value of the surface free energy. For values significantly higher than 700 $ergs/cm^2$ the nucleation rate obtained is too low to reproduce the $10^{14}$ to $10^{15}$ $voids/cm^3$ usually observed in ion bombardment experiments. It is believed that such low surface energies are physically more realistic as discussed in ref. 8.

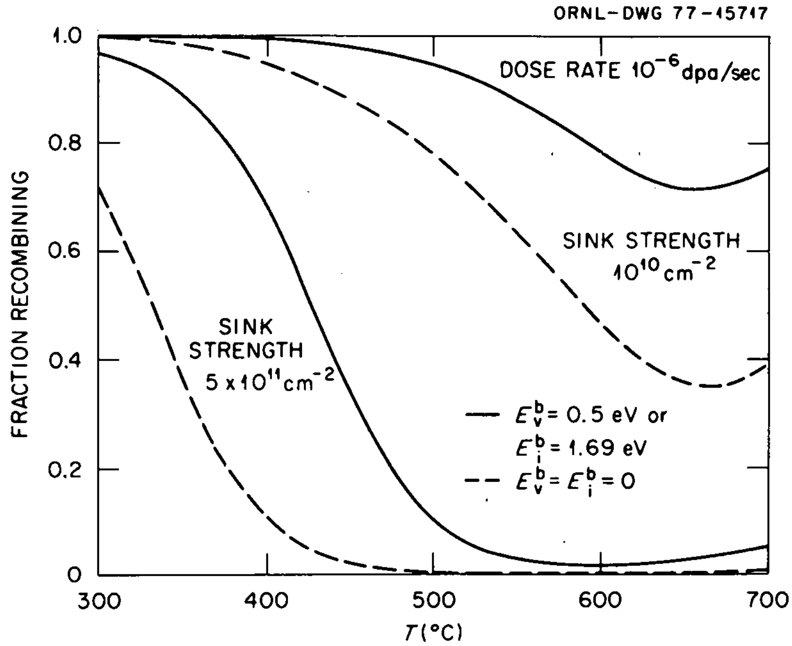


Fig. 4. The fraction of defects recombining versus temperature (i.e., not absorbed at sinks) with no point defect trapping and with either vacancy trapping with $E_v^b$ = 0.5 eV or interstitial trapping with $E_i^b$ = 1.7 eV. Two different sink strengths are considered, $5 \times 10^{11}$ cm$^{-2}$ corresponding to heavily cold-worked material and $10^{10}$ cm$^{-2}$ corresponding to annealed material after moderate radiation dose.

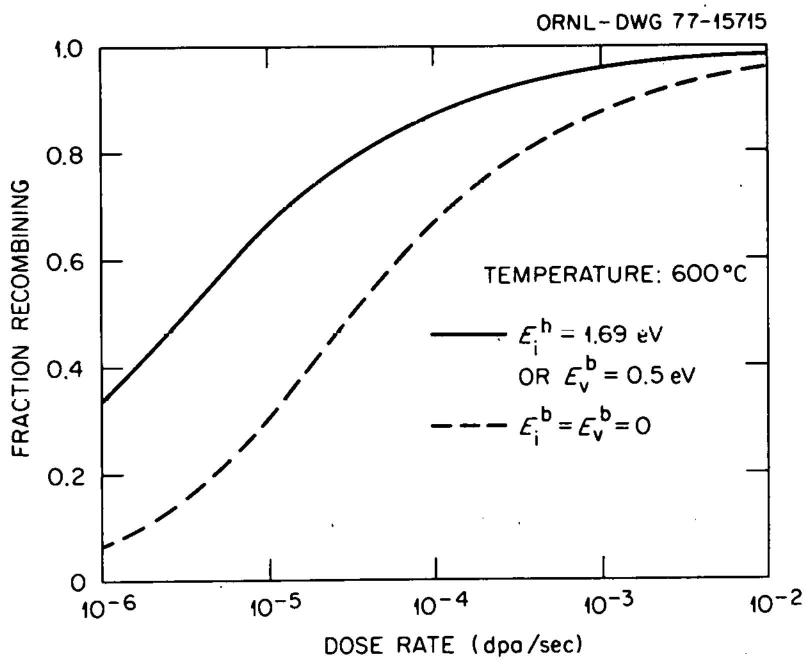


Fig. 5. The fraction of defects recombining versus dose rate with no trapping and with either vacancy trapping at $E_v^b$ = 0.5 eV or interstitial trapping at $E_i^b$ = 1.7 eV. For a given temperature, at higher dose rates the difference in the fraction recombining with and without trapping becomes smaller since nearly all recombine even without trapping at high dose rate.

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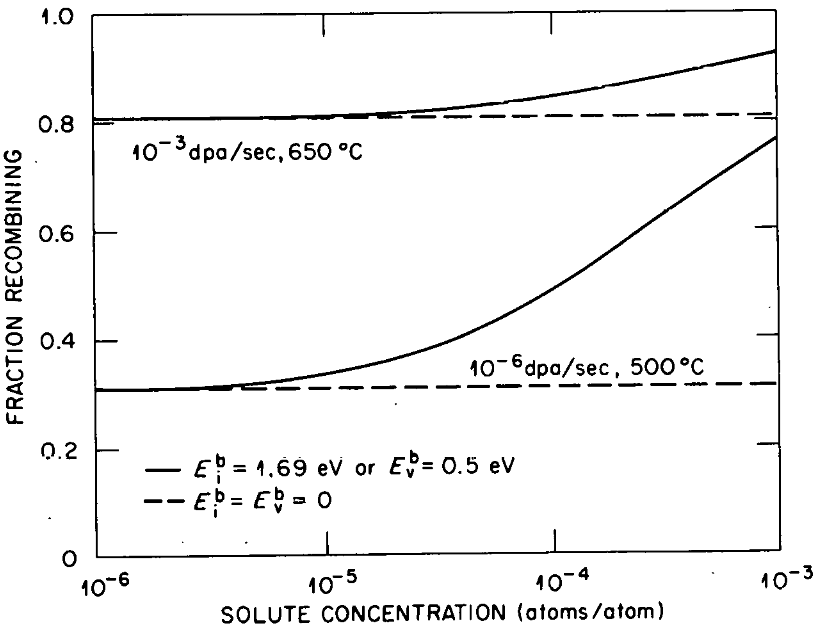


Fig. 6. Fraction recombining versus solute concentration with no trapping and with either vacancy trapping at $E_v^b$ = 0.5 eV or interstitial trapping at 1.7 eV. Results for both typical charged particle and fast reactor dose rates and temperatures are given. Trapping becomes effective at smaller solute concentrations and produces a larger increase in the fraction recombining at reactor conditions.

for a temperature of 600°C and sink strengths of $5 \times 10^{10}$ $cm^{-2}$. Above $\sim 3 \times 10^{-5}$ dpa/s, point defect recombination is the dominant mode of loss even without point defect trapping. The effect of point defect trapping is to increase this further. Figure 6 shows that the change in fraction recombining with increasing solute concentration is more pronounced at reactor conditions than at charged particle conditions.

Figures 7, 8, and 9 display the effects of point defect trapping on void nucleation, void growth and the dislocation climb (SIPA) mechanism of irradiation creep. The results are shown in Fig. 7 for the free energy of void nucleation, with the corresponding nucleation rates given in the figure caption. Figure 8 gives the results for void growth. The

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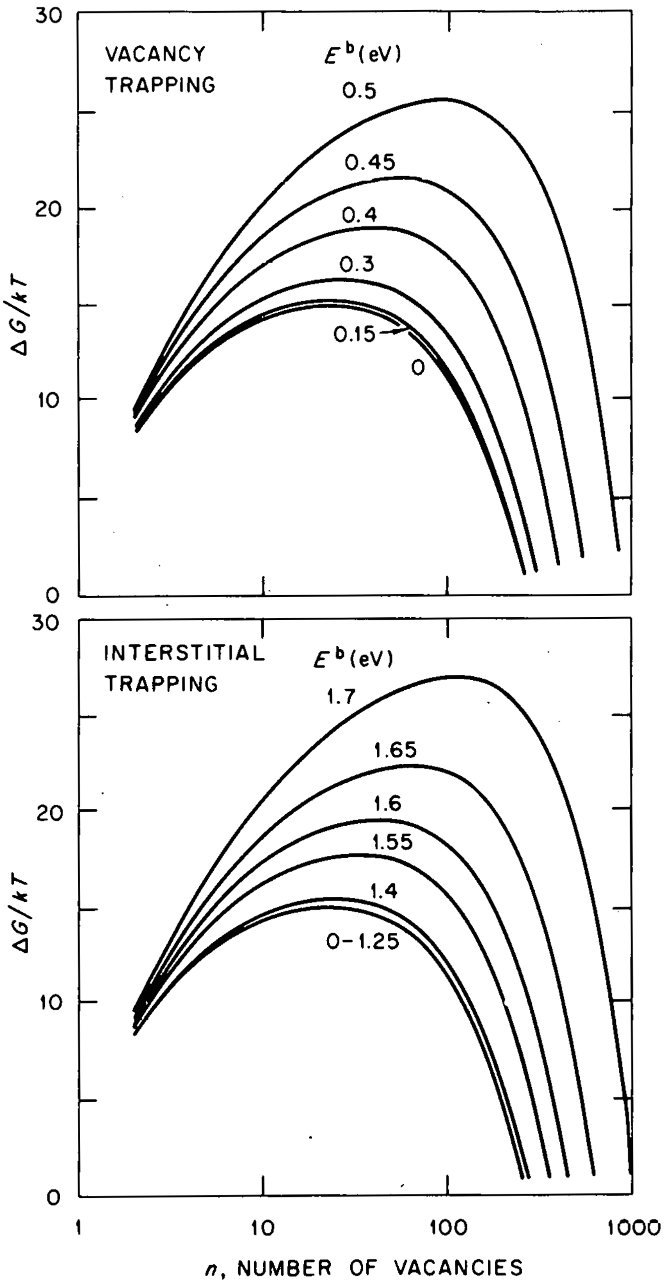


Fig. 7. The effect of impurity trapping on the free energy of void nucleation. Upper curves are for vacancy trapping and lower curves for interstitial trapping. The nucleation rates calculated from the curves are given below (in nuclei per second) as a function of binding energy in eV:

Vacancy trapping: $\left\{ \begin{array}{l} E_b = 0,\ 1.5 \times 10^{12},\ E_b = 0.3,\ 2.2 \times 10^{11}, \\ E_b = 0.4,\ 6 \times 10^{9},\ E_b = 0.5,\ 1.8 \times 10^{6}; \end{array} \right.$

Interstitial trapping: $\left\{ \begin{array}{l} E_b = 1.25,\ 1.4 \times 10^{12},\ E_b = 1.4,\ 8 \times 10^{11}, \\ E_b = 1.6,\ 2.9 \times 10^{9},\ E_b = 1.7,\ 3.5 \times 10^{5}. \end{array} \right.$

In a typical ion bombardment experiment, voids will not be observable below nucleation rates of $\sim 10^{10}\ s^{-1}$.

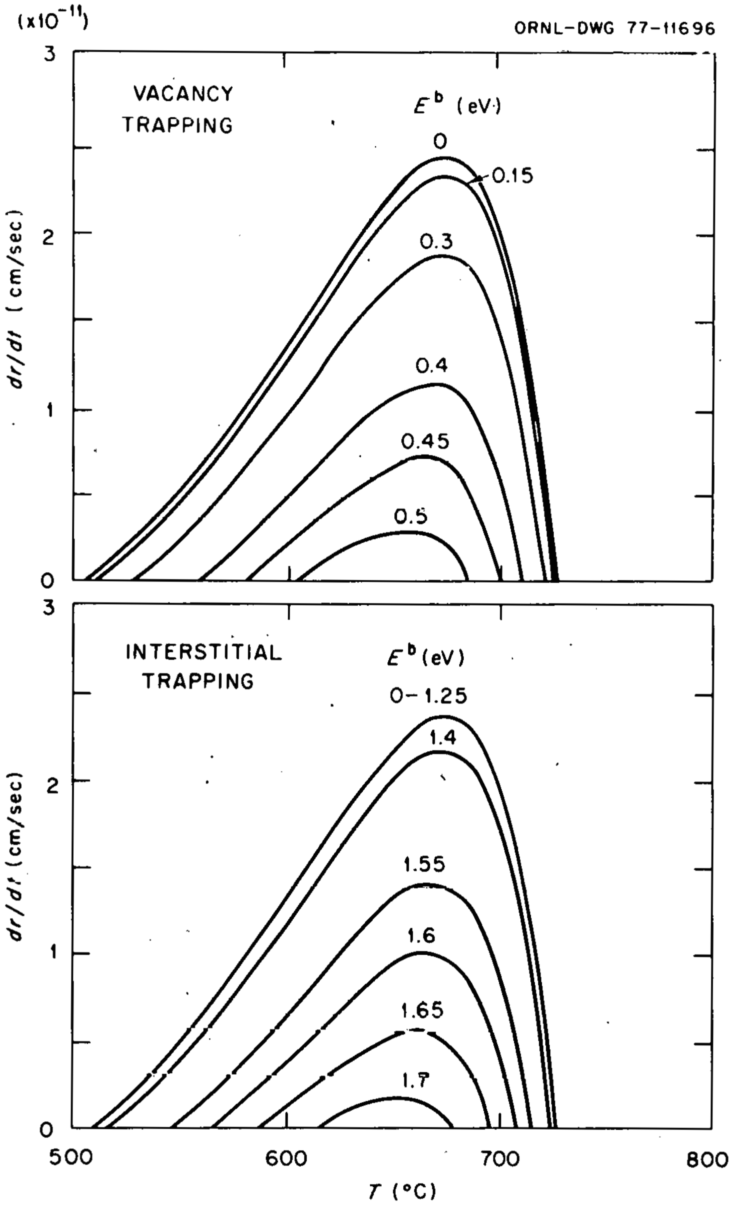


Fig. 8. The effect of impurity trapping on void growth rates. Upper curves are for vacancy trapping and lower curves for interstitial trapping. The parameters used here are given in Table 1 except that the dislocation interstitial preference is taken as 1.01/1. This reflects the fact that void growth takes place at higher doses than void nucleation where dislocation densities are generally in network rather than loops, and thus the dislocation interstitial preference is expected to be lower.

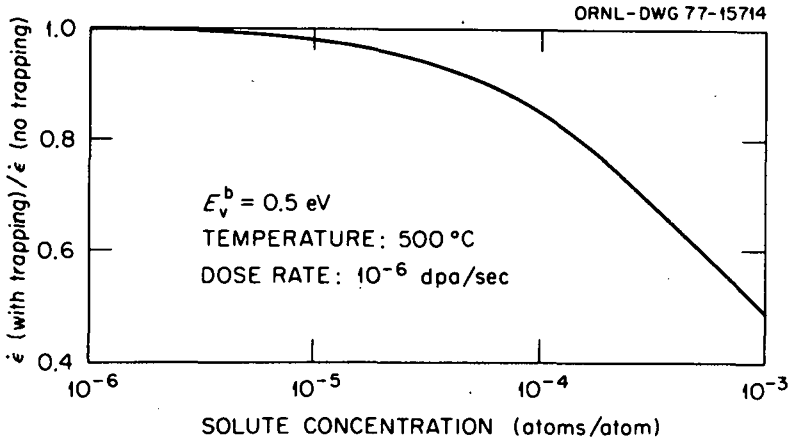


Fig. 9. Ratio of dislocation climb creep rate with vacancy trapping with E = 0.5 eV to that without trapping versus solute concentration for typical fast reactor conditions. Even at this high binding energy the dislocation climb creep rate is only reduced by a factor of 2 at the highest solute concentration. Table 2 gives the values of the other parameters used in this computation.

TABLE 2

Parameter Values Used in Obtaining Results Shown in Figure 9

| Parameter | Value |
|---|---|
| Stress, dyne/cm$^2$ | $10^9$ |
| Shear modulus, dyne/cm$^2$ | $7.75 \times 10^{11}$ |
| Poisson's ratio | 0.312 |
| $\Delta\mu_v$ , dyne/cm$^2$ | 0 |
| $\Delta\mu_i$, dyne/cm$^2$ | $-7.75 \times 10^{11}$ |
| Burger's vector, cm | $2.1 \times 10^{-8}$ |
| $\Delta V_v$, cm$^3$ | $-5.06 \times 10^{-24}$ |
| $\Delta V_i$, cm$^3$ | $1.4 \times 10^{-23}$ |
| $V_v$, cm$^3$ | $5.06 \times 10^{-24}$ |
| $V_i$, cm$^3$ | $1.4 \times 10^{-23}$ |

upper set of curves in each figure displays the effects of vacancy trapping with the lower set for interstitial trapping.

Both interstitial and vacancy trapping can be seen to produce strong reductions in swelling. To produce the same reduction in nucleation or growth rate, the binding energy required for the vacancy is much less than that for the interstitial. Indeed, to show a significant effect the interstitial binding energy must exceed the difference in migration energies between the vacancy and interstitial, 1.23 eV in these calculations. The basis of this effect is derived in Section 2.5.

Trapping reduces both the void nucleation and growth rates, with the reduction in nucleation rate by far exceeding the reduction in growth rate for a given binding energy. The nucleation rate is decreased with increases in binding energy due to increases in both the size and free energy of the critical nucleus.

Figure 9 shows that the reduction in the dislocation climb creep rate increases with increasing solute concentration for vacancy trapping under reactor irradiation conditions. This is entirely due to the reduction in free interstitial concentration since $\Delta Z_v^d$ in Eq. (31) vanishes with the parameter values used here. The physical meaning here is that there is no preferred absorption of vacancies regardless of the orientation of the dislocation Burgers vector with respect to the stress axis using the parameters of Table 2. Thus, the creep rate is directly proportional to the free interstitial population and hence less sensitive to trapping than is irradiation swelling which depends on both free interstitial and vacancy concentrations. With other parameter values there might also be a preferred absorption of vacancies, though to a smaller extent than for interstitials. The climb-glide creep rate, Eq. (32), is directly proportional to the void growth rate. Its proportionate reduction with trapping follows that shown in Fig. 8 and thus a separate figure for it need not be included.

The foregoing has illustrated the effects to be expected based upon trapping at a single type of trap as functions of temperature, dose rate, solute concentration and binding energy. With these concepts we now turn to more complex situations. Consider first the infinite medium containing traps for both vacancies and interstitials. Figures 10 and 11 show the predicted swelling as functions of dose and solute concentration, respectively, for nickel using the parameters given in Table I and on the figures. The results are shown for combined interstitial and vacancy trapping as well as for interstitial or vacancy trapping. For the binding energies illustrated, which correspond to moderately strong trapping, it is found that the combined trapping leads to less reduction in swelling than would be expected by taking the products of the fractional swellings

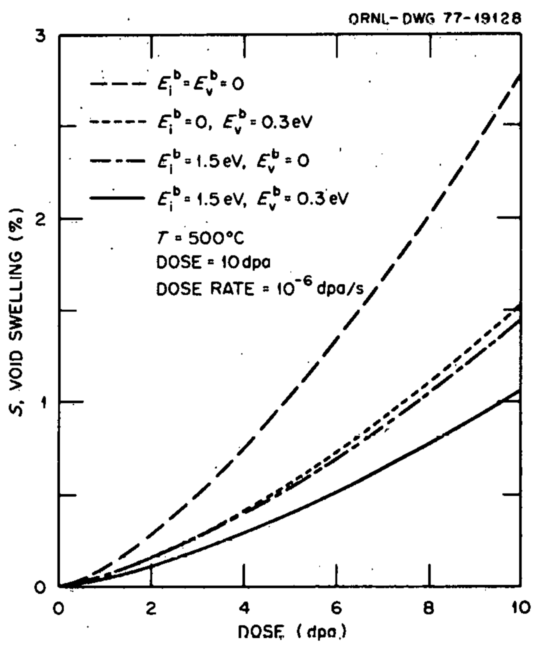


Fig. 10. The swelling versus dose for no trapping, vacancy trapping, interstitial trapping, and combined vacancy and interstitial trapping. $N_V$ denotes the number of voids (initial size 10b) and $N_L$ denotes the number of loops (initial size 10b). In Figs. 10, 11, and 12 we use the parameter values $N_V = 6 \times 10^{13}$ cm$^{-3}$, $N_L = 1 \times 10^{13}$ cm$^{-3}$, $L = 5 \times 10^9$ cm$^{-2}$, and

$$Z_i^d/Z_v^d = 1.086.$$

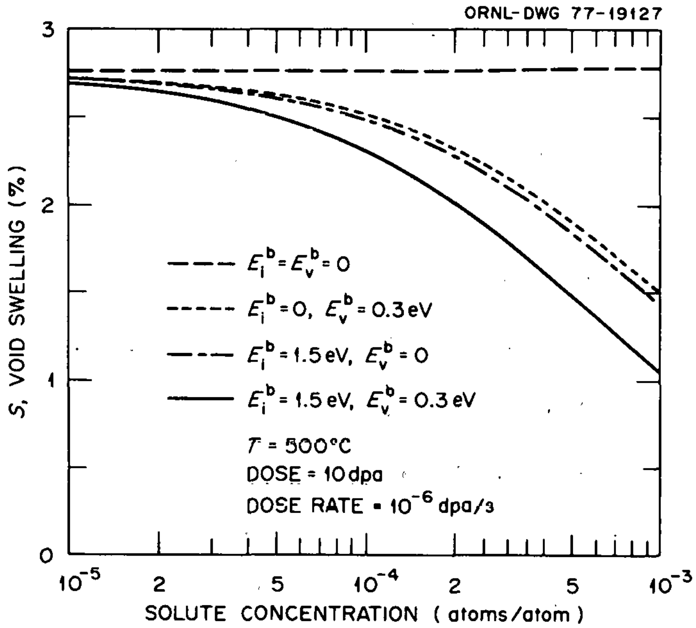


Fig. 11. The swelling at 10 dpa versus solute concentration, for no trapping, vacancy trapping, interstitial trapping, and combined vacancy and interstitial trapping.

for the cases of separate vacancy and interstitial trapping. However, for cases of weak trapping, e.g., $E_v^b = 0.1$ and $E_i^b = 1.3$ (not shown), it has been found that the fractional swelling for the combined case approximately equals the products of the fractional swellings for separate vacancy and interstitial trapping. By fractional swelling, we mean the ratio of swelling with trapping to swelling with no trapping. This result is expected intuitively since for weak trapping the entire reduction in swelling is quite small, and so each trap is operating on a system which is only slightly different from the system with no traps and will reduce swelling by roughly the same fraction as if no other traps were present.

Consider now the case of heavy-ion bombardment where the ion is chemically different from the specimen. If there is a binding energy between the implanted ion and vacancies or interstitials, this will represent a case of spatially and temporally dependent trap density. In addition, the point defect generation profile is spatially dependent.

Figure 12 shows the calculated swelling as functions of dose and position for this case. The defect generation and ion injection profiles are shown in the top part of the figure; the middle and bottom portions of the figure show the swelling at 10 and 100 dpa, respectively, for no trapping, trapping with a constant and uniform distribution of traps and for trapping by the injected ions. As expected, the swelling peak with injected traps is moved toward the surface where there are no injected traps, while the other swelling curves peak near the point defect production peak. Of course the parameters chosen are only for illustration. In particular, the injected ion profile is identical to that for 4 MeV $Ni^{++}$ in nickel. However, the result that the swelling peak may shift substantially from the point defect production peak should be taken into account when bombarding with dissimilar ions. We add parenthetically that

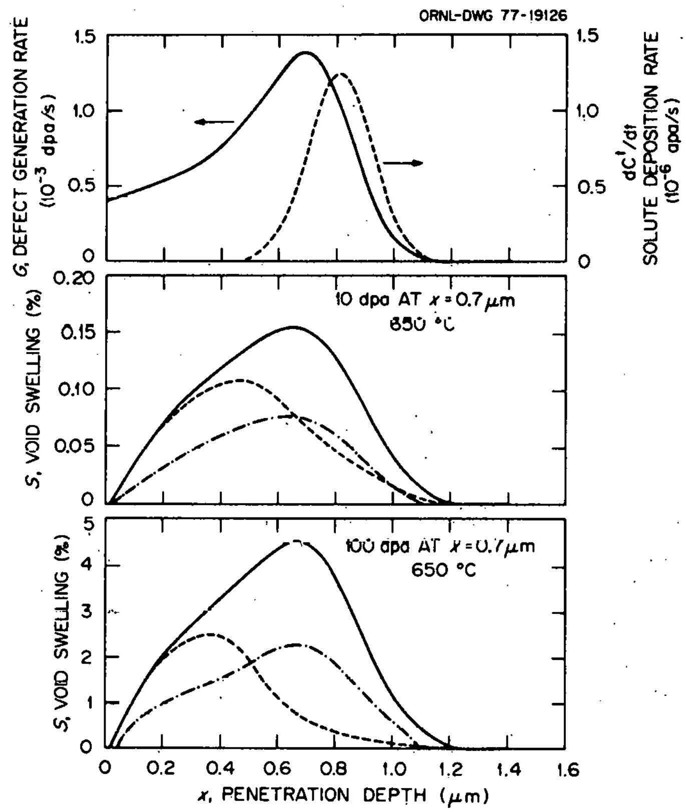


Fig. 12. Calculated swelling as a function of position for two different doses, for no trapping, trapping with constant and uniform trap distribution, and for trapping on injected ions. The solid curves in the middle and bottom portions of the figure give the swelling for no trapping, the dashed-dotted curves give the swelling for uniform solute concentration at $10^{-3}$ apa and the dashed curves for inhomogeneous solute deposition. The upper portion shows the energy and injected ion distributions.

a similar shifting of the peak in swelling may occur even when bombarding with specimen metal ions based on the injection of these ions as extra interstitials. The basis of this effect is derived in ref. 20 and the spatially dependent calculation given in ref. 19. For clarity, the solid curves in the middle and bottom portions of Fig. 12 do not include the effect of injected self-ions.

## 4. SUMMARY AND DISCUSSION

A general theory of point defect trapping and its effect on void formation and irradiation creep has been developed. This theory encompasses both vacancy and interstitial trapping at a multiplicity of traps and the effects of point defect trapping on void nucleation, void growth, and irradiation creep. The equations also explicity include spatial inhomogeneity and time dependence. The relationship to earlier work in this field is reviewed where necessary. The significance of higher order corrections are discussed. In particular the correction due to exclusion of sites near traps to free vacancies becomes important when the trap concentration is of the order of 1%. The traps are described by their concentration, trapping radius, capture radius for a free defect when the opposite type defect is trapped, and by their binding energy for point defects. By varying these parameters, either solute atoms, solute atom clusters, or precipitates may be included as traps [6,21].

Trapping can be viewed in terms of effective recombination or diffusion coefficients. The correct expressions for these effective coefficients are given. The general result that vacancy trapping is effective in reducing swelling and creep at relatively small binding energies but that interstitial trapping is only effective if the binding energy exceeds approximately the difference in the free vacancy and interstitial migration energies can be understood in terms of effective diffusion coefficients.

Point defect trapping increases the fraction of defects recombining in the matrix at the expense of those diffusing to sinks and thereby decreases the concentration of free point defects. Of the three processes considered — void nucleation, void growth, and irradiation creep — trapping is predicted to be most effective in decreasing the void nucleation rate; relatively low concentrations and binding energies of traps decreasing the nucleation rate by at least several orders of magnitude. The next largest effect of trapping is on void growth rate and to exactly the same degree on the climb-glide creep rate. The smallest effect is on the SIPA dislocation climb mechanism of irradiation creep. The effects predicted are illustrated primarily as functions of binding energy, trap concentration, temperature, and dose rate for parameter ranges of interest.

Implications for planning of future experiments and interpreting experimental results are significant. Certainly the well-known qualitative relation between increasing solute content and reduction in swelling is confirmed. For more exact testing of the theory, we need more detailed experiments with carefully controlled additions of given solutes at several levels and of solutes with variation in binding energy. From Figs. 6 and 11 we see, for example, that point defect trapping even with reasonably large binding energies is expected to have little effect if the solute concentrations are below about 10 ppm. Thus, if the effect of impurities at this level is found experimentally to be significant the implication is that point defect trapping is not the operative mechanism there. Use must be made of the results of more fundamental work to determine binding energies. With regard to creep, the theory predicts less sensitivity to solute content, a trend which is qualitatively borne out by experiment since many greatly different alloys do not have widely divergent creep rates [22]. The predicted limited sensitivity of creep results to trapping calls for even greater care in planning experiments of this type.

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