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L. K. Mansur
W. G. Wolfer

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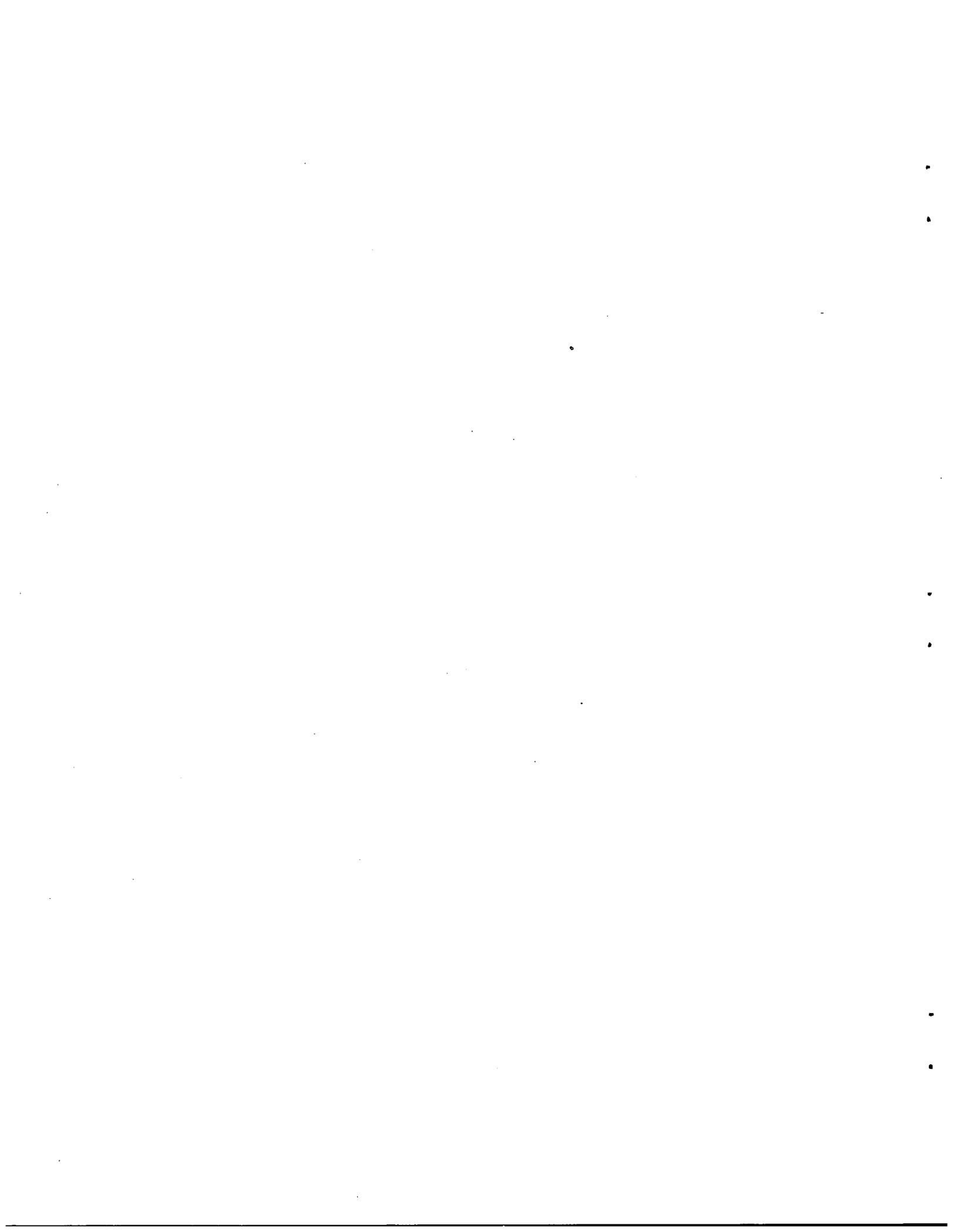
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A STUDY OF THE EFFECT OF VOID SURFACE COATINGS
ON RADIATION-INDUCED SWELLING

L. K. Mansur and W. G. Wolfer*

ABSTRACT

A theoretical investigation has been conducted into the effects on point defect diffusion of the presence of a shell of segregated material about a spherical void. The elastic image interaction of the point defect with the void surrounded by a shell of material of elastic constants which differ from those of the matrix is first computed. The interaction, which depends on the properties of the shell and the dilatation of the defect, results in an energy barrier which must be overcome when the defect diffuses to the void surface. The capture efficiency of the void for the defect is computed for a range of parameters employing this interaction. This capture efficiency is then used directly to obtain predicted effects of segregation on void nucleation and growth under irradiation. It is found that the absorption of the interstitial is reaction controlled at the void. The principal implications are that void nucleation is strongly enhanced and that the kinetics of void growth are altered significantly. Detailed derivations supporting these results are given.

INTRODUCTION

Current experimental research has shown that impurity and alloying element content largely control the swelling behavior of metals.^{1,2†} This fact has great significance for the development of low swelling alloys to be used in structural components of future fission and fusion reactors. To make such development systematic it is necessary to understand the bases of impurity action. In light of our current understanding of swelling,³ two ways in which impurities may act are by altering the free migration of point defects in the material through trapping reactions and by changing the relative capture efficiencies of point

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†For *List of References*, see p. 24.

defect sinks, such as voids, dislocations and precipitates. The former effect has often been invoked in speculative explanations of differences in swelling behavior, but has received more detailed attention recently.⁴⁻⁶ However, we believe it is important to investigate physical mechanisms for the latter possibility since swelling depends so delicately on small differences in the point defect capture efficiencies of the sinks in the swelling material.³

Solute segregation to void surfaces in stainless steels⁷ and aluminum⁸ has been observed. Furthermore, segregation to void surfaces is expected to occur ubiquitously based upon observations of segregation on virtually all free surfaces. This is thought to be due to the usual lowering of the thermodynamic free energy of the system as well as to impurity binding with radiation-induced point defect fluxes of vacancies and interstitials. It is important to answer the question as to what effects such segregation may have on the efficiency with which a void absorbs point defects and ultimately on swelling. By altering the diffusion coefficients of point defects near the void, a segregated layer may affect swelling. This has been explored by Brailsford.⁹ A further mechanism by which a segregated layer may affect swelling behavior is through alteration of the point defect-void image interaction. The purpose of this paper is to describe a theoretical study of this effect.

INTERACTION ENERGY

This work employs the results of the recent derivation of Wolfer and Mansur¹⁰ for the elastic image interaction energy between a center of dilatation and a spherical cavity surrounded by a shell of material whose elastic constants differ from those of the matrix. The expressions for the interaction energy when the defect is in the matrix and in the shell are presented below.

$$E_m = -\frac{\Gamma_m}{2r^3} \sum_{n=0}^{\infty} \frac{n(n-1)(4n^2-1)}{[(\gamma-1) X_n(-v_m) + (4n^2-1)(1-v_m)]} \\ \times \left\{ \frac{4\gamma}{\Delta_n} (4n^2-1)^2 (1-v_s)(1-v_m) [\gamma(1-v_s)(2n+1)(2n+3) + (1-\gamma) \right. \\ \left. \times (1-n^{-2n-3}) X_n(v_s)] + \gamma - 1 \right\} \xi^{2n+2} \quad (1)$$

$$\begin{aligned}
E_s = & -\frac{\Gamma_s}{r_m^3} \sum_{n=0}^{\infty} \Delta_n^{-1} \left\{ n(n-1)(4n^2-1) \right. \\
& \times [(1-\nu_m)(4n^2-1) - (1-\gamma) X_n(-\nu_m)] [2\gamma(1-\nu_s)(2n+1)(2n+3) \\
& + 2(1-\gamma)(1-\eta^{-2n-3}) X_n(\nu_s)] \xi^{2n+2} + (1-\gamma)(n+1)(n+2)(2n+1)(2n+3) \\
& \times \{2\gamma(1-\nu_s)(4n^2-1) X_n(-\nu_m) + 2(\eta^{2n-1}-1) X_n(-\nu_s) \\
& \times [(1-\nu_m)(4n^2-1) - (1-\gamma) X_n(-\nu_m)]\} \xi^{-2n} \\
& - 2(1-\gamma)n(n-1)(n+1)(n+2)(4n^2-1)(2n+3)(1-\eta^{-2}) \\
& \left. \times [(1-\nu_m)(4n^2-1) - (1-\gamma) X_n(-\nu_m)] \xi \right\}. \quad (2)
\end{aligned}$$

Subscripts s and m denote shell and matrix, $\Gamma = v^2 \mu(1+v)^2/[18\pi(1-v)]$, where

v = point defect dilatation volume,

μ = shear modulus,

ν = Poisson's ratio,

$\gamma = \mu_m/\mu_s$,

$\xi = r_m/r_d$,

$\eta = r_m/r_s$,

r_m = interface radius,

r_s = void radius,

r_d = distance of defect from center of void,

$X_n(v) = n^2 + n(1-2\nu) + 1 + \nu$, and

$\Delta_n = I_n J_n - K_n L_n$,

where

$I_n = 2\gamma(1-\nu_s)(2n+1)(2n+3) + 2(1-\gamma)(1-\eta^{-2n-3}) X_n(\nu_s)$,

$J_n = 2\gamma(1-\nu_s)(4n^2-1) X_n(-\nu_m) + 2(\eta^{2n-1}-1) X_n(-\nu_s) Y_n(-\nu_m, \gamma)$,

$K_n = (1-\gamma)(1-\eta^{-2}) n(n+2)(2n-1)$,

$L_n = (1-\eta^{-2})(n^2-1)(2n+3) Y_n(-\nu_m, \gamma)$, and

$Y_n(v, \gamma) = (1-|\nu|)(4n^2-1) - (1-\gamma) X_n(v)$.

Two special cases of these equations are also of interest. When the thickness of the shell goes to zero, Eq. (1) yields the image interaction

between a center of dilatation and a void. Setting $r_m = r_s = r$, $\Gamma_m = \Gamma_s = \Gamma$, $v_m = v_s = v$, $\xi = r/r_d$ we obtain

$$E_o = -\frac{\Gamma}{2r^3} \sum_{n=0}^{\infty} \frac{n(n-1)(4n^2-1)}{X_n(-v)} \xi^{2n+2}. \quad (3)$$

This can be shown by taking Eq. (1) with the above limiting values of the parameters. By inspection, Eq. (1) then becomes

$$E_m = -\frac{\Gamma_m}{2r^3} \sum_{n=0}^{\infty} \frac{4n(n-1)(4n^2-1)^2(1-v)^2(2n+1)(2n+3)\xi^{2n+2}}{\Delta_n}. \quad (4)$$

Writing out Δ_n gives

$$\begin{aligned} \Delta_n &= 2\{\gamma(1-v_s)(2n+1)(2n+3) + (1-\gamma)(1-\eta^{-2n-3} X_n(v_s))\} \\ &\times \{2\gamma(4n^2-1)(1-v_s) X(-v_m) + 2(4n^2-1)(1-v_m)(\eta^{2n-1}-1) X_n(-v_s) \\ &\quad - 2(1-\gamma)(\eta^{2n-1}-1) X_n(-v_s) X_n(-v_m)\} \\ &- \{(1-\eta^{-2})(n-1)(n+1)(2n+3)[(4n^2-1)(1-v_m) - (1-\gamma) X_n(-v_m)]\} \\ &\quad \times \{(1-\gamma)(1-\eta^{-2}) n(n+2)(2n-1)\}. \quad (5) \end{aligned}$$

With the above limiting values of parameters this can be seen to reduce to

$$\Delta_n = 4(1-\gamma)(2n+1)(2n+3)(4n^2-1)(1-v) X_n(-v).$$

Substituting this last expression into the equation preceding Eq. (5) yields Eq. (3).

In the opposite limit, when the thickness of the shell equals the radius, we obtain the image interaction between a center of dilatation and an inhomogeneous inclusion. Setting $r_s = 0$, $r_m = r_l$, and $\xi = r_l/r_d$ we thus obtain

$$E_l = \frac{\Gamma}{2r_l^3} (1-\gamma) \sum_{n=0}^{\infty} \frac{n(n-1)(4n^2-1)}{[(\gamma-1) X_n(-v_m) + (4n^2-1)(1-v_m)]} \xi^{2n+2} \quad (6)$$

This result is obtained from Eq. (1) by simply noting that Δ_n , given by Eq. (5), becomes infinite as $r_s \rightarrow 0$, recalling the definition $\eta = r_m/r_s$. Hence, only $\gamma - 1$ remains in the curly brackets of Eq. (1). This gives Eq. (6) directly.

Equations (3) and (6), obtained as special cases of the coated void result [Eq. (1)], are identical with the results of Moon and Pao¹¹ who derived the interaction energy of a center of dilatation with a single region inclusion.

Figure 1 shows the interaction energies of a center of dilatation with a coated void for the case where the shear modulus of the shell is twice as large as that of the matrix (solid curve), and for the case where the shear modulus of the shell equals that of the matrix [*i.e.*, no shell (broken curve)]. Table 1 gives the other parameters necessary for these computations, together with their values selected for these sample computations. When the shell is stiffer than the matrix, $\mu_s > \mu$ and the point defect is in the matrix, it is repelled from the interface at short distances. When the defect is in the shell, it is attracted toward the interface at short distances. When the matrix is stiffer than the shell, $\mu_m > \mu_s$, (not shown) these regions of repulsion and attraction (positive and negative interaction energy, respectively) simply occur on the opposite sides of the interface. Near the free surface, the interaction energy is always negative. In this region Eq. (2) takes on the form of Eq. (3) as shown in Fig. 1. Very close to the free surface, the surface curvature becomes unimportant and Eq. (2) takes on the even simpler form characteristic of a flat surface

$$E_f = \frac{\Gamma/2}{(r_d - r)^3} \quad (7)$$

where, by the definitions of r_d and r , $r_d - r$ is simply the distance of the point defect from the free surface. Equation (7) agrees with the result obtained by Eshelby¹² and Bacon¹³ for the image interaction of a center of dilatation with a flat surface.

Equation (7) is derived as follows: Write $\xi \equiv r/r_d$ in the form $\xi = (1 + \alpha/r)^{-1}$, where $\alpha \equiv r_d - r$ is the distance of the defect from the surface. Equation (3) then may be written

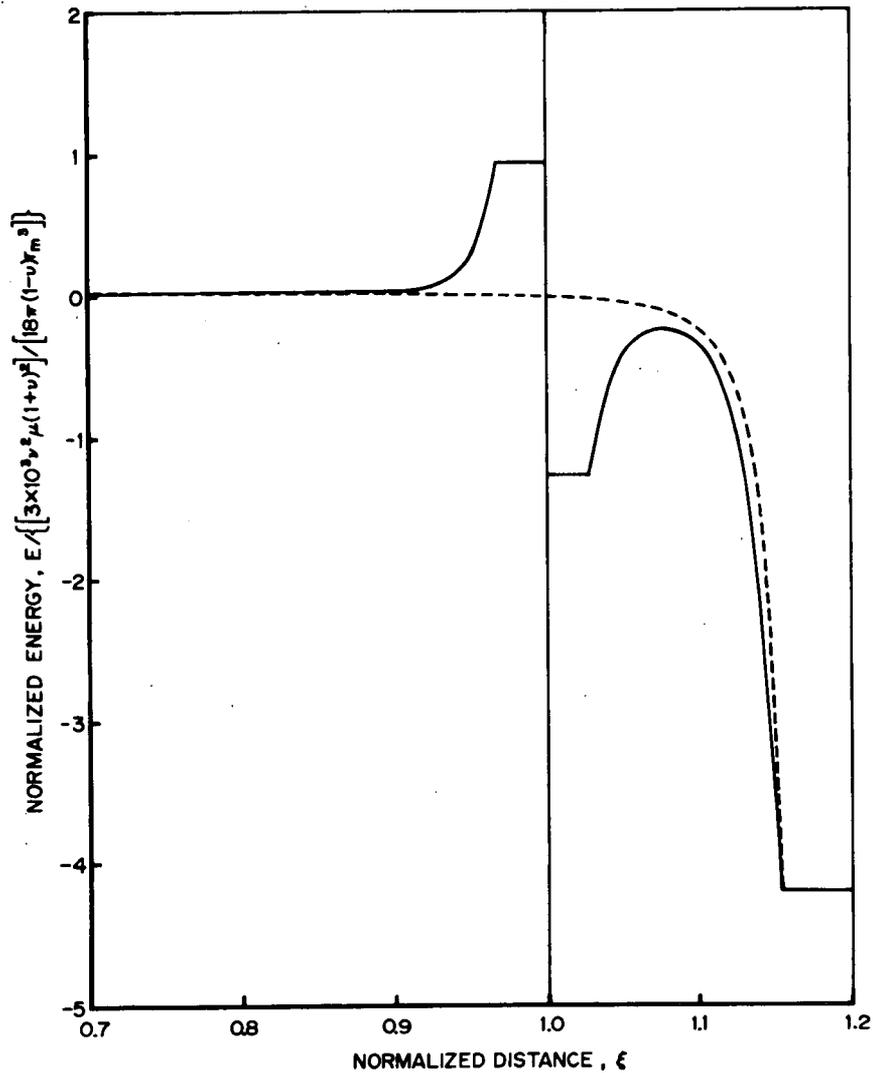


Fig. 1. Interaction Energy Versus Inverse of Distance to Void Center. Coating occupies region from $\xi = 1$ to 1.2. Energy saturation distance parameter $\xi = \Gamma/r_m = 0.033$. For $r_m = 3$ nm this corresponds to a physical distance $\approx b/2 \approx 0.1$ nm. When coating is softer than matrix $\mu_m/\mu_s > 1$ (not shown), positions of maximum and minimum are reversed; for $\mu_m/\mu_s = 1$, there is no region where interaction is repulsive.

$$E_f = -\frac{\Gamma/2}{r^3} (1 + \alpha/r)^{-2} \sum_{n=1}^{\infty} \frac{n(n-1)(4n^2-1)}{n^2 + n(1-2\nu) + 1-\nu} [(1 + \alpha/r)^{-2}]^{-n}.$$

When the defect is near the surface, that is when $\alpha \ll r$, we expand the expressions containing α and neglect terms of higher order than α/r .

Thus,

$$E_f = -\frac{\Gamma/2}{r^3} (1 + \alpha/r)^{-2} \sum_{n=0}^{\infty} \frac{n(n-1)(4n^2-1)}{n^2 + n(1-2\nu) + 1-\nu} (1 - 2\alpha/r)^n.$$

Table 1. Parameter Values Used in Calculations

<u>Interaction Energy</u>						
γ	ν_m	ν_s	$\frac{\nu_s^s \text{ or } m}{\nu_v}$	$\frac{\nu_i^s \text{ or } m}{\nu_i}$	$\mu_m \text{ (n/m}^2\text{)}$	$\mu_s \text{ (n/m}^2\text{)}$
0.5, 2.0	0.3	0.3	0.2 Ω	1.4 Ω	5×10^{10} , 1×10^{11}	1×10^{11} , 5×10^{10}
<u>Capture Efficiency</u>						
$r_s \text{ (nm)}$	$r_m - r_s \text{ (nm)}$	c		$T \text{ (}^\circ\text{C)}$		
0.15 to 5	0.25, 0.50	$r_a, b/2$		500		
<u>Nucleation</u>						
$\sum_j K_i^j / D_i \text{ (cm}^{-2}\text{)}$	$\sum_j K_v^j / D_v \text{ (cm}^{-2}\text{)}$	R/D_i	$D_v \text{ (cm}^2 \cdot \text{sec}^{-1}\text{)}$	$c_v^e \text{ (cm}^{-3}\text{)}$		
7.5×10^9	5×10^9	25b	$0.015 \exp(-E_v^m/kT)$	$\Omega^{-1} \exp(1.5 - E_v^f/kT)$		
$E_v^m \text{ (eV)}$	$E_v^f \text{ (eV)}$	$r_a \text{ (nm)}$	$b \text{ (nm)}$	$\Omega \text{ (m}^{-3}\text{)}$	$G \text{ (dpa/sec)}$	\bar{c}_v / c_v^e
1.4	1.6	0.125	0.21	1.1×10^{-29}	1.1×10^{-7}	1

Because the last factor in the summation terms is near unity, only the higher order terms in the summation contribute to E_f since there the terms in the summation increase in size roughly as n^2 . The asymptotic form is then justified

$$E_f \sim -\frac{2\Gamma}{r^3} \sum_{n=0}^{\infty} n^2 (1 - 2\alpha/r)^n$$

However, since as already noted, the terms for low n make negligible contribution, the summation in this equation is equivalent to the summation $\sum_{n=0}^{\infty} (n+1)(n+2)\theta^n$, with θ given by $\theta = 1 - 2\alpha/r$. This summation can be expressed in closed form as $2/(1-\theta)^3$ (ref. 14). Substituting this closed form into the above equation gives Eq. (7) directly.

It can be seen from Fig. 1 that in order for the point defect to diffuse to the free surface of the coated void it must overcome the repulsive energy near the shell-matrix interface. This is true whether the shell is harder or softer than the matrix. This barrier hinders the

diffusion of point defects and thus decreases the capture efficiency of the coated void for point defects. Capture efficiencies are derived and illustrated in the following section.

Before proceeding, however, a further point should be clarified. The above calculations employ elasticity theory, with centers of dilatation modeled as mathematical points. The interface and free surface are modeled as abrupt boundaries. These idealizations cause the interaction energies to diverge at these boundaries. This nonphysical behavior is avoided by realizing that the point defect has a finite radius. In these computations we allow the interaction energy to saturate at its value one defect radius away when it is nearer to an interface than one defect radius. This results in the plateaus and flat-bottomed wells shown in Fig. 1. Their precise height or depth depends upon the distance c at which the interaction energy is allowed to saturate. Both the atomic radius r_a , and one-half the {111} interplanar spacing, $b/2 = a/2\sqrt{3}$, where a is the lattice parameter, are considered reasonable distances.

CAPTURE EFFICIENCY

In this section, the manner in which the interaction energy affects point defect absorption at a void is determined. The effect is measured by the point defect capture efficiency of the sink. The capture efficiency is defined as the ratio of the actual point defect current to the sink to the current to the sink if there were no interaction energy and $C = C^e(r)$ at the sink is specified as a boundary condition. We define C as the point defect concentration variable and $C^e(r)$ as the thermal equilibrium point defect concentration at the sink of radius r .

To derive the capture efficiency the void is placed in the medium containing one type of point defect whose concentration at large distances has a fixed value, say C^∞ . In the vicinity of the void there is no net leakage of point defects from any infinitesimal volume element. There is thus a continuity condition which can be expressed in spherical coordinates as

$$\frac{1}{\rho^2} \frac{\partial}{\partial \rho} (\rho^2 J_\rho) = 0, \quad (8)$$

where ρ is the distance from the center of the void and J_ρ is the radial flux of point defects. Thus, the form for J_ρ is given by

$$J_\rho = -A/\rho^2, \quad (9)$$

where A is a constant. This must equal the flux determined by Fick's law generalized to include a drift interaction energy $E(\rho)$,

$$J_\rho = -\exp[-E(\rho)/kT] \frac{\partial}{\partial \rho} \{DC \exp[E(\rho)/kT]\}, \quad (10)$$

where D is the diffusion coefficient. The capture efficiency of a void of radius r_s defined in words above is given by

$$Z = \frac{I}{4\pi r_s DC^\infty} = \frac{4\pi r_s^2 J_\rho(r_s)}{4\pi r_s DC^\infty} \quad (11)$$

where I is the actual current to the sink and $4\pi r_s D[C^\infty - C^e(r_s)]$ is the current to the sink without a drift interaction, assuming a boundary condition of the form $C(r_s) = C^e(r_s)$. In Eq. (11) $C^e(r_s)$ has been set equal to zero, corresponding to the case where thermal point defect emission is negligible, since retaining it adds little to the derivation. From Eqs. (9) and (11) we obtain

$$Z = \frac{-A}{r_s DC^\infty} \quad (12)$$

In order to determine A we multiply Eqs. (9) and (10) by $\exp[E(\rho)/kT]$, integrate from r_s to infinity, and obtain

$$A = \frac{DC \exp[E(\rho)kT] \Big|_{r_s}^{\infty}}{\int_{r_s}^{\infty} \frac{\exp[E(\rho)/kT]}{\rho^2} d\rho} \quad (13)$$

The numerator at the upper limit is just DC^∞ and at the lower limit it is zero since $E(\rho)$ goes to zero at infinity and to large negative values at the void surface, r_s . Substituting Eq. (13) into (12) gives

$$Z(\rho) = \left[r_s \int_{r_s}^{\infty} \frac{\exp[E(\rho)kT]}{\rho^2} d\rho \right]^{-1} \quad (14)$$

Changing variables to $\xi \equiv r_m/r_d = r_m/\rho$, $d\xi = -r_m d\rho/\rho^2$, we obtain from Eq. (14)

$$Z(\rho) = \dot{n} \left[\int_0^{\rho} \exp[E(\xi)/kT] d\xi \right]^{-1} . \quad (15)$$

Since $E(\xi)$ has been given in *Interaction Energy* (p. 2, this report), the capture efficiency is now determined.

Before illustrating computed values of the capture efficiency, we discuss Eq. (15). A form similar in limiting cases to Eq. (15), but not containing the necessary information for evaluating Z , can be derived by simple consideration of boundary conditions. Consider a void with transfer velocity w for the point defect across the void-matrix interface. The solution to the spherical continuity Eq. (8) for an uncoated void of radius r in steady state, allowing for no drift interaction is always of the form

$$C(\rho) = C^\infty(1 - Z r/\rho) . \quad (16)$$

We use the logarithmic boundary condition

$$D \left(\frac{dC}{d\rho} \right)_{\rho=r} = wC(r) , \quad (17)$$

which specifies that the flux to the void as obtained from Fick's law must equal the flux determined by the ambient concentration at the void surface. From Eqs. (16) and (17) we find

$$Z = [1 + D/wr]^{-1} . \quad (18)$$

When $w = D/b$, where b is a lattice spacing (which also equals the point defect transfer velocity in the matrix far from the void), then the capture efficiency Z approximately equals unity for normal size voids, $r \gg b$. This is spoken of as a diffusion-controlled case since the diffusion through the matrix controls the rate of point defect absorption. If, on the other hand, the transfer velocity is low, $w \ll D/b$, the void is a poor absorber and we find from Eq. (18) $Z \approx wr/D$. This is termed a surface-reaction-controlled case since the point defect current is

determined by the transfer rate at the void surface. In this case Eq. (11) yields

$$I_s = 4\pi wr^2 C^\infty, \quad (19)$$

while in the diffusion-controlled case, $w = D/b$, $Z \approx 1$, Eq. (14) yields

$$I_D = 4\pi Dr C^\infty. \quad (20)$$

The point defect current depends upon different powers of the void radius in these two cases. It is on this basis that it has been determined that void growth is surface-reaction limited in stainless steels.^{15,16}

To see that Eq. (15) takes a similar form to Eq. (18) and therefore produces similar limiting forms to Eqs. (19) and (20), we break the integral in Eq. (16) into two parts,

$$Z(\xi) = \eta \left[\int_0^{\eta-\epsilon} \exp[E(\xi)/kT] d\xi + \int_{\eta-\epsilon}^{\eta} \exp[E(\xi)/kT] d\xi \right]^{-1}. \quad (21)$$

From Fig. 1 it is seen that the interaction energy is negligible outside a small region beyond the void. The entire region where it is negligible is expressed in normalized distance units as from 0 to $\eta - \epsilon$. Thus, the value of the first integral is $\eta - \epsilon$.

Since $E(\xi)$ appears in an exponential and the integration range is small, we may replace the second integral by $\epsilon \exp[E(\xi^*)/kT]$ where $E(\xi^*)$ measures the height of the barrier depicted in Fig. 1 and ξ its width. Thus, Eq. (21) can be approximated by

$$Z = \eta \{ (\eta - \epsilon) + \epsilon \exp[E(\xi^*)/kT] \}^{-1} \quad (22)$$

Substituting the definitions $\eta \equiv r_m/r_s$ and $\epsilon \equiv (r_m - r^*)/r_s$, where r^* is the distance from the void center to the positive energy peak, into Eq. (22) we obtain

$$Z = \left(\frac{r^*}{r_m} + D/wr \right)^{-1} \quad (23)$$

where

$$w = \frac{D \exp[-E(\xi^*)/kT]}{\epsilon r} \quad (24)$$

Equation (23) is essentially identical to Eq. (18) provided $r_m \approx r^*$ $\approx r_s \approx r$ (i.e., for a thin coating). Thus, if $E(\xi^*)$ in Eq. (24) is small or negative (no barrier) then $w \approx D/\epsilon r$ giving from Eq. (23) $Z \approx 1$ and thus reproducing Eq. (19). If $E(\xi^*)$ is large then from Eq. (24) $w \ll D/\epsilon r$ giving $Z \approx wr/D$ from Eq. (23) and thus reproducing Eq. (20). The important point here is that by using the interaction energy $E(\xi)$ derived in the previous section we can evaluate, in the language of Eq. (18), the transfer velocity, w , using a physical relationship, Eq. (24). However, the derivation of Eq. (18) solely from a boundary condition offers no information on how to determine the essential parameter w . Thus, although its limiting forms are approximately correct, Z determined in that way would remain an unknown parameter.

Figure 2 shows the results of numerical computations of Z as a function of void size for the coated and uncoated void using Eqs. (1), (2) and (15) together with the parameter values given in Table 1. Several features merit discussion. The first thing to note is that the void is not a neutral sink, $Z_i^V = Z_V^V = 1$ (the superscript denotes void), as has been generally assumed in the previous literature. The bare void (broken curves) strongly prefers interstitials, $Z_i^V > Z_V^V$, due to the larger dilatation volume and therefore larger attraction of the interstitial. However, when the void is coated with a material which is stiffer than the matrix (solid curves), this preference is reversed ($Z_V^V > Z_i^V$) due to a very large reduction in the interstitial capture efficiency, again due to its larger dilatation volume and therefore larger repulsion, especially at small void sizes. The upturn in the interstitial curve at small sizes occurs because the repulsion is smaller there. This can be understood as follows. The calculations are performed by varying r_m but saturating the energy at a fixed, physically based distance c from the interface, thus giving a small ξ at small r_m . From Eq. (1) small ξ yields small E_m . The reason that the vacancy capture efficiency is slightly higher for the coated void than for the bare void is, of course,

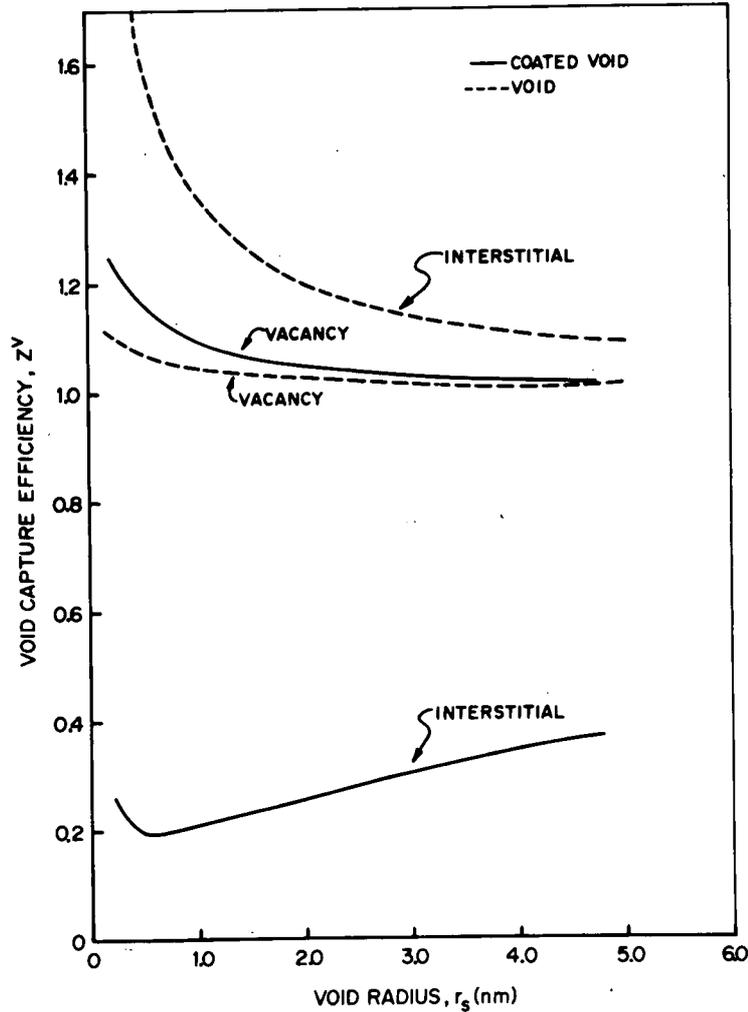


Fig. 2. Capture Efficiencies of Void and Coated Void for Vacancy and Interstitial as Functions of Void Radius r_s . The void prefers interstitials, especially at small void sizes. This preference is reversed for the coated void. Parameters used in obtaining this plot are $r_m - r_s = 0.25$ nm, $c = 0.1$ nm with the remainder given in Table 1.

the attraction at the interface which does not exist for the bare void (the repulsion for the vacancy at the opposite side of the interface is negligible).

SWELLING

Reaction rate equations³ can be used to determine the time and space averaged vacancy and interstitial concentrations, C_v and C_i ,

which are utilized in void nucleation and growth calculations. In a simplified form these may be written

$$G + G_T - RC_V C_i = \sum_j K_V^j C_V \quad (25)$$

$$G - RC_V C_i = \sum_j K_i^j C_i \quad (26)$$

Here G is the radiation-induced point defect generation rate, $G_T = \sum_j K_V^j C_V^j$ is the thermal vacancy generation rate where C^j is the thermal equilibrium vacancy concentration at the average size sink of type j , $R = 4\pi r^C (D_i + D_V)$ is the recombination rate coefficient where r^C is the radius of point defect recombination volume, D denotes diffusion coefficient and subscripts i and v denote interstitial and vacancy, respectively. The K 's are reaction rate constants for loss of point defects at internal sinks and may be approximated by

$$K^j = 4\pi \bar{r}_j \bar{Z}^j N_j D, \quad (27)$$

for "spherical" sinks* such as voids and

$$K^j = \bar{Z}^j L D, \quad (28)$$

for cylindrical sinks such as dislocations; \bar{r}_j is the average radius of sink type j , \bar{Z}^j the capture efficiency of sinks of type j and L the line density of dislocation sinks.

Void Nucleation

In order to determine the effect of surface coatings on vacancy clusters upon the energetics and rates of void nucleation, we employ the nucleation theory developed by Katz and Wiedersich¹⁷ and Russell.¹⁸ The change in the number of vacancy clusters containing n vacancies with respect to time, $\partial q(n)/\partial t$, is the difference between the number arriving

*Reference 3 gives the forms of the K 's for the general case when the sink size distribution is used rather than average size sinks.

per unit time from size $n - 1$, $M(n - 1)$, and the number leaving per unit time to size $n + 1$, $M(n)$, where

$$M(n) = \beta_v(n) q(n) - [\beta_i(n + 1) + \gamma_v(n + 1)] q(n + 1) , \quad (29)$$

and is given by

$$\begin{aligned} \frac{\partial q(n)}{\partial t} = M(n - 1) - M(n) = & [\beta_i(n + 1) + \gamma_v(n + 1)] q(n + 1) \\ & - [\beta_v(n) + \beta_i(n) + \gamma_v(n)] q(n) + \beta_v(n - 1) q(n - 1) , \end{aligned} \quad (30)$$

where $\beta_{v,i}(n)$ is the capture rate of vacancies, interstitials by a vacancy cluster containing n vacancies, and $\gamma_v(n)$ is the emission rate of vacancies from a vacancy cluster containing n vacancies. In Eq. (29) terms corresponding to the emission of interstitials from vacancy clusters of size n and $n - 1$ are omitted since the formation energy of the interstitial is much larger than that of the vacancy in materials of interest. Equation (30) has two steady-state solutions corresponding to $\partial q(n)/\partial t = 0$. If $M(n - 1) = M(n) = 0$ there is no net flux of vacancy clusters to larger sizes. This corresponds to the so-called constrained equilibrium distribution.¹⁹ We denote the number of vacancy clusters of size n for this constrained distribution as $q^0(n)$. Equation (29) gives

$$\beta_v(n) q^0(n) = [\beta_i(n + 1) + \gamma_v(n + 1)] q^0(n + 1) . \quad (31)$$

Employing this as a recursion relation we obtain

$$q^0(n) = \sum_{\ell=1}^{n-1} \frac{\beta_v(\ell) q^0(1)}{\beta_i(\ell+1) + \gamma_v(\ell+1)} . \quad (32)$$

This may be written as

$$q^0(n) = q^0(1) \exp[-\Delta G(n)/kT] , \quad (33)$$

with the definition

$$\Delta G(n) = -kT \sum_{\ell=1}^{n-1} \ell n \frac{\beta_v(\ell)}{\beta_i(\ell + 1) + \gamma_v(\ell + 1)} , \quad (34)$$

where $\Delta G(n)$ is the free energy of formation of a cluster containing n vacancies. In these equations, $\gamma_V(n)$ and $\beta_V(n)$ can be expressed in terms of point defect concentrations as follows

$$\gamma_V(n) = 4\pi \Omega^{1/3} n^{1/3} Z_V^V(n) D_V C_V^V(n) / (4\pi/3)^{1/3} \quad (35)$$

where

$$C_V^V(n) = C_V^e \exp \left[\left(\frac{2\sigma}{r(n)} - P \right) \Omega / kT \right] \quad (36)$$

is the thermal equilibrium vacancy concentration at a vacancy cluster containing n vacancies, Ω is the atomic volume, $Z_V^V(n)$ is the capture efficiency of a vacancy cluster (void) of size n for vacancies, C_V^e is the bulk thermal equilibrium concentration of vacancies, σ is the surface energy of the void, P is the pressure exerted by any gas contained within the void, and $r(n)$ is the radius of the void containing n vacancies.

$$\beta_{i,V}(n) = 4\pi \Omega^{1/3} n^{1/3} Z_{i,V}^V(n) D_{i,V} C_{i,V} / (4\pi/3)^{1/3}, \quad (37)$$

where C_i and C_V are the point defect concentrations given by Eqs. (41) and (42) below.

The second steady-state solution to Eq. (30) is that for which $M(n) = M(n-1) = \dots \equiv M$, where M is defined as the steady-state nucleation rate. Using Eqs. (29) and (31), we find

$$M = \beta_V(n) q^0(n) \left[\frac{q(n)}{q^0(n)} - \frac{q(n+1)}{q^0(n+1)} \right] \quad (38)$$

An expression for M in terms of $\Delta G(n)$ defined by Eq. (34) can be obtained by the repeated summation of Eq. (38),

$$\sum_{\ell=1}^{\infty} \left[\frac{q(\ell)}{q^0(\ell)} - \frac{q(\ell+1)}{q^0(\ell+1)} \right] = \frac{q(1)}{q^0(1)} - \frac{q(\infty)}{q^0(\infty)} = \sum_{\ell=1}^{\infty} \frac{M}{\beta_V(\ell) q^0(\ell)} \quad (39)$$

We know, however, that $q(\infty)/q^0(\infty) \rightarrow 0$ and $q(1)/q^0(1) \rightarrow 1$ in analogy with classical nucleation theory.¹⁹ Using Eq. (33) and noting that $q^0(1) = C_V$, the vacancy concentration, we obtain for the steady-state nucleation rate,

$$M = \frac{c_v}{\sum_{n=1}^{\infty} \frac{\exp[\Delta G(n)/kT]}{\beta_v(n)}} \quad (40)$$

Equations (34) and (40) are the key relations which determine the effects of changes in void capture efficiencies on void nucleation behavior. It is useful to derive the explicit dependence of $\Delta G(n)$ and M on the capture efficiencies, $Z_{i,v}^V(n)$. Equations (25) and (26) give for the point defect concentrations

$$c_v = \frac{(\sum_j K_{ij}^j \sum_j K_{vj}^j - G_{TR})}{2R \sum_j K_j^j} \left\{ \left[1 + \frac{4(G + G_T) \sum_j K_{ij}^j \sum_j K_{vj}^j}{(\sum_j K_{ij}^j \sum_j K_{vj}^j - G_{TR})^2} \right]^{1/2} - 1 \right\} \quad (41)$$

$$c_i = \frac{(\sum_j K_{ij}^j \sum_j K_{vj}^j + G_{TR})}{2R \sum_j K_{ij}^j} \left\{ \left[1 + \frac{4RG \sum_j K_{ij}^j \sum_j K_{vj}^j}{(\sum_j K_{ij}^j \sum_j K_{vj}^j + G_{TR})^2} \right]^{1/2} - 1 \right\} \quad (42)$$

Substituting Eqs. (35), (36), (37), (41), and (42) into Eq. (34) yields

$$\frac{\Delta G(n)}{kT} = \sum_{\ell=1}^{n-1} \ln \left\{ \frac{(\ell + 1)^{1/3} Z_{i,v}^V(\ell + 1)}{\ell^{1/3} Z_{i,v}^V(\ell)} \frac{D_{i,v} \sum_j K_{vj}^j}{D_v \sum_j K_{ij}^j} \zeta + c^V[r(\ell)] \right\} - (n-1) \ln(\zeta + \bar{c}_v), \quad (43)$$

where

$$\zeta = \frac{\sum_j K_{ij}^j}{2R} \left\{ \left[\left[1 + \frac{RG_T}{\sum_j K_{ij}^j \sum_j K_{vj}^j} \right]^2 + \frac{4RG}{\sum_j K_{ij}^j \sum_j K_{vj}^j} \right]^{1/2} - \left[1 + \frac{RG_T}{\sum_j K_{ij}^j \sum_j K_{vj}^j} \right] \right\} \quad (44)$$

and

$$\bar{c}_v = G_T / \sum_j K_{vj}^j \quad (45)$$

Equation (43) reveals the explicit dependence of the free energy of cluster formation upon the capture efficiencies $Z_i^V(n)$ and $Z_v^V(n)$ of voids (vacancy clusters) for interstitials and vacancies, respectively. The nucleation rate M can also be expressed in terms of these parameters. From Eqs. (37) and (40) we obtain

$$M = \frac{4\pi\Omega^{1/3} C_v^2 D_v / (4\pi/3)^{1/3}}{\sum_{n=1}^{\infty} \frac{\exp[\Delta G(n)/kT]}{n^{1/3} Z_v^V(n)}} \quad , \quad (46)$$

where C_v is given by Eq. (41).

Using Eqs. (43) and (46) as the key equations and invoking the capture efficiencies obtained from Eqs. (15), (1), and (2) together with the parameter values shown in Table 1, free energies of vacancy cluster formation as well as void nucleation rates have been computed. The results for ΔG are shown in Fig. 3 with the corresponding nucleation rates given in the figure caption. For two values of the surface free energy (namely, 1100 and 800 ergs/cm²) the energy of formation of a vacancy cluster of size n versus n is plotted for both the coated and the uncoated void. The peaks in these curves correspond to the vacancy cluster critical size — that size above which the system will lower its energy as the cluster size increases further. For both choices of surface energy, the coated vacancy cluster has substantially smaller critical size and energy. Correspondingly, the nucleation rate is substantially increased over that for the bare vacancy cluster. However, even with a coating the vacancy cluster with a surface energy of 1100 ergs/cm², a typical value from the literature for a number of fcc metals, does not have a physically realistic nucleation rate under the conditions typical of nuclear reactor irradiation given in Table 1. Decreasing the surface energy to 800 ergs/cm² provides more than ample nucleation rate, with coated vacancy clusters nucleating more than two orders of magnitude faster than bare vacancy clusters. It is concluded that vacancy clusters must be coated in order to nucleate at physically reasonable rates. The coating is required both to decrease the strong preference of the bare vacancy cluster for interstitials (Fig. 2) and to decrease the surface energy enough to allow realistic nucleation rates.

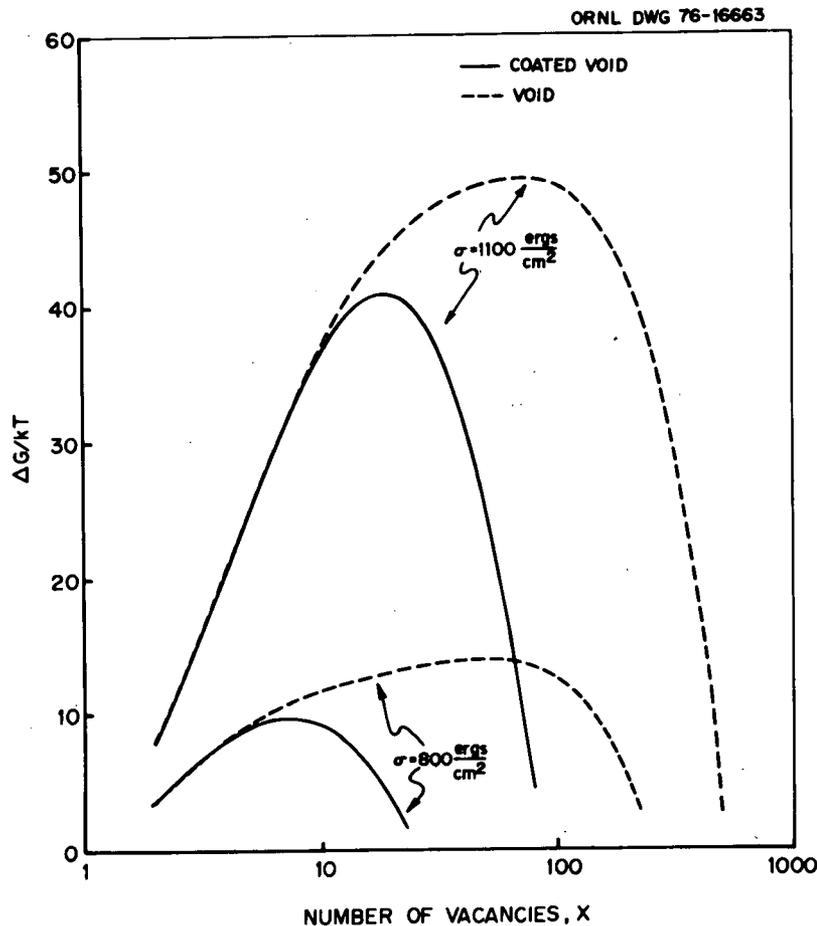


Fig. 3. Free Energy of Vacancy Cluster Formation Versus Cluster Size for Void and Coated Void at Two Values of the Surface Energy. The coating reduces formation energy through its effect on capture efficiencies shown in Fig. 2. Lowering surface energy causes further reduction. Corresponding nucleation rates proceeding from highest curve to lowest are ($\text{cm}^{-3} \cdot \text{sec}^{-1}$). 2.3×10^{-6} , 9.6×10^{-2} , 3.1×10^9 , 5.8×10^{11} .

Void Growth

The rate theory of void growth has been formulated in terms of diffusion-controlled²⁰⁻²² and surface-reaction-controlled^{15,16} absorption of vacancies and interstitials at voids. In accordance with our discussion (*Capture Efficiency*, p. 8, this report), it is found that the predicted swelling kinetics are quite different for these two cases. This is discussed extensively in ref. 3. Figure 2 indicates, however, that based only upon the void-point defect image interaction, the absorption rate of interstitials is surface-reaction controlled,

$$Z_i^V = w_i r/D_i \ll 1, \quad (47)$$

while the vacancy is diffusion controlled

$$Z_V^V \approx 1. \quad (48)$$

We therefore investigate the predicted kinetics of void growth under this mixed condition.

The radial growth rate of a void is expressed in terms of the net flux of vacancies to the void

$$\frac{dr}{dt} = \frac{\Omega}{r} [Z_V^V(r) D_V C_V - Z_i^V(r) D_i C_i - Z_V^V(r) D_V C_V^V(r)]. \quad (49)$$

Substituting the C's from Eqs. (44) and (45) gives

$$\begin{aligned} \frac{dr}{dt} = \frac{\Omega}{r} & \left\{ \frac{Z_V^V(r) D_V}{2R \sum_j K_j^V} \left(\sum_j K_i^j \sum_j K_j^V - R G_T \right) \left\{ \left[1 + \frac{4R(G + G_T) \sum_j K_j^V \sum_j K_i^j}{\left(\sum_j K_i^j \sum_j K_j^V - R G_T \right)^2} \right] - 1 \right\} \right. \\ & \left. - \frac{Z_i^V(r) D_i}{2R \sum_j K_j^i} \left(\sum_j K_j^V \sum_j K_i^j + R G_T \right) \left\{ \left[1 + \frac{4R G \sum_j K_j^i \sum_j K_j^V}{\left(\sum_j K_i^j \sum_j K_j^V + R G_T \right)^2} \right]^{1/2} - 1 \right\} - Z_V^V(r) D_V C_V^V(r) \right\}. \end{aligned} \quad (50)$$

To illustrate a special case of Eq. (50), we restrict the point defect loss to dislocations and voids and neglect vacancy-interstitial mutual recombination and thermal emission. This gives

$$\frac{dr}{dt} = \frac{\Omega G L}{r} \frac{\left(Z_i^d Z_V^v - Z_i^v Z_V^d \right)}{\left(4\pi Z_i^v(r) r N_V + Z_i^d L \right) \left(4\pi Z_V^v(r) r N_V + Z_V^d L \right)}. \quad (51)$$

As usual, we set $Z_i^d = (1 + \epsilon') Z_V^d$, where ϵ' measures the preference of dislocations for interstitials over vacancies. Substituting Eqs. (47) and (48) into (51) and integrating, we obtain for $Z_V^d L \gg 4\pi Z_V^v(r) r N_V$ (i.e., dislocations the dominant sink),

$$r = - \frac{\Omega G w_i}{L D_i Z_V^d (1 + \epsilon') (1 - p)} t - \frac{D_i (1 + \epsilon')}{w_i} \ln \left[1 - \frac{w_i r}{(1 + \epsilon') D_i} \right], \quad (52)$$

while for $Z_V^d \ll 4\pi Z_V^v(r) r N_V$ (i.e., voids the dominant sink),

$$\frac{r^4}{4} - \frac{D_i (1 + \epsilon') r^3}{3w_i} - \frac{D_i^2 (1 + \epsilon')^2 r^2}{2w_i^2} - \frac{D_i^3 (1 + \epsilon')^3}{w_i^3} r - \frac{D_i^4 (1 + \epsilon')^4}{w_i^4} \ln \left[1 - \frac{\bar{r} w_i}{D_i (1 + \epsilon')} \right] = \frac{\Omega G L Z_V^d t}{(4\pi N_V)^2 (1 + p)}. \quad (53)$$

In these equations L (the dislocation density) may be a function of dose. Nevertheless, it has been possible to integrate these equations with respect to dose provided that N_V , the void number density, does not change with dose and the dislocation density varies with time according to $L = L_0 t^p$, where L_0 is a constant and p is a number (experimentally, p usually lies between 0 and 0.5).

When $w_i r / D_i (1 + \epsilon') \ll 1$ (as we have already assumed), Eq. (52) can be approximated as

$$r \approx \left[\frac{2\Omega G}{L Z_V^d (1 - p)} \right]^{1/2} t^{1/2} \quad \text{dislocations dominant} \quad (54)$$

Similarly, Eq. (53) becomes

$$r \approx \left[\frac{5\Omega G L Z_V^d D_i (1 + \epsilon')}{w_i (4\pi N_V)^2 (1 + p)} \right]^{1/5} t^{1/5} \quad \text{voids dominant} \quad (55)$$

These forms are obtained by expanding Eqs. (52) and (53) and neglecting higher order terms.

It has been shown previously³ that when both interstitial and vacancy are surface-reaction controlled to the same extent (i.e., $w_v / D_v = w_i / D_i$), then under the same assumptions as above

$$r = \frac{\Omega G w_v}{L Z_v^d (1-p)} \left(\frac{Z_i^d D_i w_v}{Z_v^d D_v w_i} - 1 \right) t \quad (56)$$

dislocations dominant

$$r = \left[\frac{5\Omega G D_v Z_v^d L}{w_v (4\pi N_v)^2 (1+p)} \right]^{1/5} \left(\frac{Z_i^d D_i w_v}{Z_v^d D_v w_i} - 1 \right)^{1/5} t^{1/5}, \quad (57)$$

voids dominant

while if both interstitial and vacancy are diffusion controlled

$$r = \left(\frac{2\Omega G}{Z_v^d L (1-p)} \right)^{1/2} \left(\frac{Z_i^d}{Z_v^d} - 1 \right)^{1/2} t^{1/2} \quad (58)$$

dislocations dominant

$$r = \left(\frac{\Omega G Z_v^d L}{4\pi^2 N_v^2 (1+p)} \right)^{1/4} \left(\frac{Z_i^d}{Z_v^d} - 1 \right)^{1/4} t^{1/4} \quad (59)$$

voids dominant

Thus, comparing Eqs. (57) and (55) we see that when the interstitial alone is surface-reaction controlled and the voids are the dominant sink, the kinetics are the same as if both defects are surface-reaction controlled except that the radial growth rate is larger by the factor $(Z_i^d/Z_v^d - 1)^{-1/5}$. Similarly, by comparing Eqs. (58) and (54) we see that when the interstitial alone is surface-reaction controlled and the dislocations are the dominant sink then the kinetics are the same as if both defects are diffusion controlled except that the radial growth rate is larger by the factor $(Z_i^d/Z_v^d - 1)^{-1/2}$. Z_i^d/Z_v^d is normally taken to be 1.01 to 1.1. Thus, the growth rate of a coated void is always expected to be much larger than that of the uncoated void. Again, this is the consequence of the repulsive image interaction produced by the void coating reducing the void capture efficiency for the interstitial while not significantly altering the capture efficiency for the vacancy.

CONCLUSIONS

From the foregoing we conclude that the induced image interaction between point defects and vacancy clusters or voids may have a significant effect on swelling during irradiation. Using the accurate interaction energies, it is found that the void has a strong preference for interstitials over vacancies, thus contradicting the usual assumption that voids are neutral sinks, but that when the void is coated with a shell of segregated material this preference is decreased or reversed for typical parameter values. Several consequences follow from this. The critical nucleation energy is decreased, and the nucleation rate is increased substantially for coated versus uncoated void embryos. Nucleation computations show that void number densities of the order observed are more likely if the embryos are surrounded by solute shells. Void growth computations show that growth rates are significantly more rapid for coated voids, also due to the preference of the coated void for vacancies.

Void coatings are expected to occur widely based on thermodynamic requirements for adsorption at surfaces as well as impurity drag due to radiation-induced point defect fluxes. Thus, these concepts suggest interpretations of swelling behavior in terms of the properties of segregated impurity shells. These ideas may be tested experimentally. For example, by introducing into a metal or alloy, say in the Fe-Cr-Ni system, impurities known to segregate (e.g., carbon, oxygen, or silicon) on the one hand, and on the other hand introducing impurities known to trap segregants either in the matrix or by precipitation reactions (e.g., Ti or Zr), one should be able to produce large differences in swelling behavior.

With regard to additional implications, it is recommended that the common practice of taking voids as neutral sinks in models of swelling materials be reevaluated. In future work the present analysis could be used in models of the evolving microstructure where solute segregation builds up and results in dynamically changing void capture efficiencies for point defects.

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