Nucleation of Voids in Materials Supersaturated with Mobile Interstitials, Vacancies, and Divacancies

W.G. Wolfer and A. Si-Ahmed

May 1982

UWFDM-471

Phil. Mag. A46, 723-736 (1982).

Nucleation of Voids in Materials Supersaturated with Mobile Interstitials, Vacancies, and Divacancies

W.G. Wolfer and A. Si-Ahmed

Fusion Technology Institute
University of Wisconsin
1500 Engineering Drive
Madison, WI 53706

http://fti.neep.wisc.edu

May 1982

UWFDM-471

Phil. Mag. A46, 723-736 (1982).

Nucleation of Voids in Materials Supersaturated With Mobile Interstitials, Vacancies, and Divacancies

W.G. Wolfer and A. Si-Ahmed*

Fusion Engineering Program
Nuclear Engineering Department
University of Wisconsin-Madison
Madison, Wisconsin 53706

May 1982

UWFDM-471

---

*Present address: Centre des Sciences et de la Technologies Nucleaires, Algiers, Algeria.

Abstract

Previous void nucleation theories allow the void size to change only by one atomic volume through vacancy or interstitial absorption or through vacancy emission. To include the absorption of divacancies, the classical nucleation theory is extended to include double-step transitions between clusters. The new nucleation theory is applied to study the effect of divacancies on void formation. It is found that the steady state void nucleation rate is enhanced by several orders of magnitude compared to results with previous void nucleation theories. However, in order to obtain void nucleation rates comparable to measured ones, the effect of impurities, segregation, and insoluble gases must still be invoked.

§1. INTRODUCTION

Previous treatments of homogeneous void nucleation (Katz and Wiedersich 1971, Russell 1971) in irradiated materials were based on the assumption that only the monatomic point defects, namely vacancies and interstitials, need to be considered as mobile species. Presumably, the greater concentration of the monodefects, as compared to the dimer populations, served as the justification for this assumption. However, a closer examination of the previous void nucleation theories reveals that the nucleation rate does not only depend on the supersaturations of the mobile point defects, but also on the products of their concentrations and their corresponding diffusion coefficients. This type of dependence is in fact well known from the rate theory for void growth (Brailsford and Bullough 1972).

If the monodefects have also the highest mobility in addition to the highest concentration, it is indeed justified to neglect mobile multi-defects such as divacancies. This appears to be the case for interstitial-type point defects. For example, Johnson (1966) and Schober (1977) found by computer simulation that the migration energies for diffusion of di-, tri-, and tetra-interstitials are all slightly larger than the migration energy of mono-interstitials. On the other hand, Ehrhardt and Schlagheck (1974) have concluded from experiments that the migration energy of di-interstitials in copper could be slightly less than the one of mono-interstitials. Nevertheless, the difference is not sufficiently large to make the products of concentration and diffusivity for di-interstitials and mono-interstitials of comparable magnitude.

In the case of vacancies, the situation is different. As the reader is well aware, the migration energy of divacancies can be significantly lower than the one of monovacancies for many cubic metals. Furthermore, the binding energy of divacancies is sufficiently large so that a significant fraction of them remains bound even at high temperatures. As a result, the product of divacancy concentration and diffusion coefficient can be as large as the corresponding product for monovacancies. This has been demonstrated by Yoo (1979) in the case of void and dislocation loop growth in irradiated metals.

In spite of the recognition that divacancies play a role in void and loop growth, their effect on the nucleation process has not yet been investigated. Whereas the incorporation of divacancies in the rate theories for void and loop growth is accomplished by a simple extension of the classical rate theory (Brailsford and Bullough 1972), previous void nucleation theories cannot be simply modified to include divacancies. The reason is that all homogeneous nucleation theories as presently formulated require that a cluster can change its size by only one unit at a time. Therefore, to allow multi-step processes to occur requires a substantial extension of previous nucleation theories.

Such an extension is presented in §3 and applied to void nucleation in the presence of mono-interstitials, and mono- and divacancies. The rate equations which determine their concentrations are given in §2. The results of numerical evaluation of the new void nucleation theory and comparison with the old theory are presented in §4.

We note that the present extension of the homogeneous void nucleation theory can also be applied to other nucleation phenomena which involve nearest and next-nearest neighbor transitions between clusters. Furthermore, the method developed here can be extended to any more distant neighbor transitions, and can therefore provide a formalism to incorporate coagulation reactions into the nucleation theory.

§2. RATE EQUATIONS FOR POINT DEFECTS AND CLUSTERS

Let f(x,t) denote the number of vacancy clusters containing x vacancies. This number may change with time as a result of vacancy capture, divacancy capture, interstitial capture, or vacancy re-emission. The rates for these reactions are denoted by the symbols listed in Table 1, and they are assumed to depend on the size x of the cluster.

The detailed dependence will be given later in §4. By inspection of Fig. 1 it is easy to derive the following rate equation for the change in the cluster density f(x,t), namely

$$df(x,t)/dt = \nu(x - 2)f(x-2,t) + \beta(x - 1)f(x-1,t) - [\alpha(x) + \beta(x) + \gamma(x) + \nu(x)]f(x,t) + [\alpha(x + 1) + \gamma(x + 1)]f(x+1,t) \ . \tag{1}$$

The change in the cluster density may also be derived from a cluster current advancing clusters smaller than x to clusters larger or equal to x, minus a cluster current promoting clusters with x vacancies. The former is the net reaction I(x-1,t) crossing the dashed line in Fig. 1 just below the cluster size level x, whereas the latter is the net reaction I(x,t) crossing the dashed lined just above the size x.

This cluster current is given by

$$I(x,t) = \nu(x - 1)f(x-1,t) + a(x)f(x,t) - b(x + 1)f(x+1,t) \ . \tag{2}$$

The forward and backward reaction rates, a(x) and b(x), are defined in Table 1. By changing x to x - 1 one obtains I(x-1,t), and one can then write for Eq. (1)

Table 1. Notation for Reaction Rate Coefficients

Symbol	Reaction Rate
$\alpha(1)$	Recombination rate per mono-interstitial capture at all mobile monovacancies
$\alpha(x)$	Capture rate of mono-interstitials at a cluster containing x vacancies
$\beta(x)$	Capture rate of monovacancies at a cluster containing x vacancies
$\gamma(2)$	Dissociation rate of divacancies
$\gamma(x)$	Re-emission rate of monovacancies from a cluster containing x vacancies
$\nu(x)$	Capture rate of divacancies at a cluster containing x vacancies
$a(x)$	$= \beta(x) + \nu(x)$, forward reaction rate
$b(x)$	$= \alpha(x) + \gamma(x)$, backward reaction rate

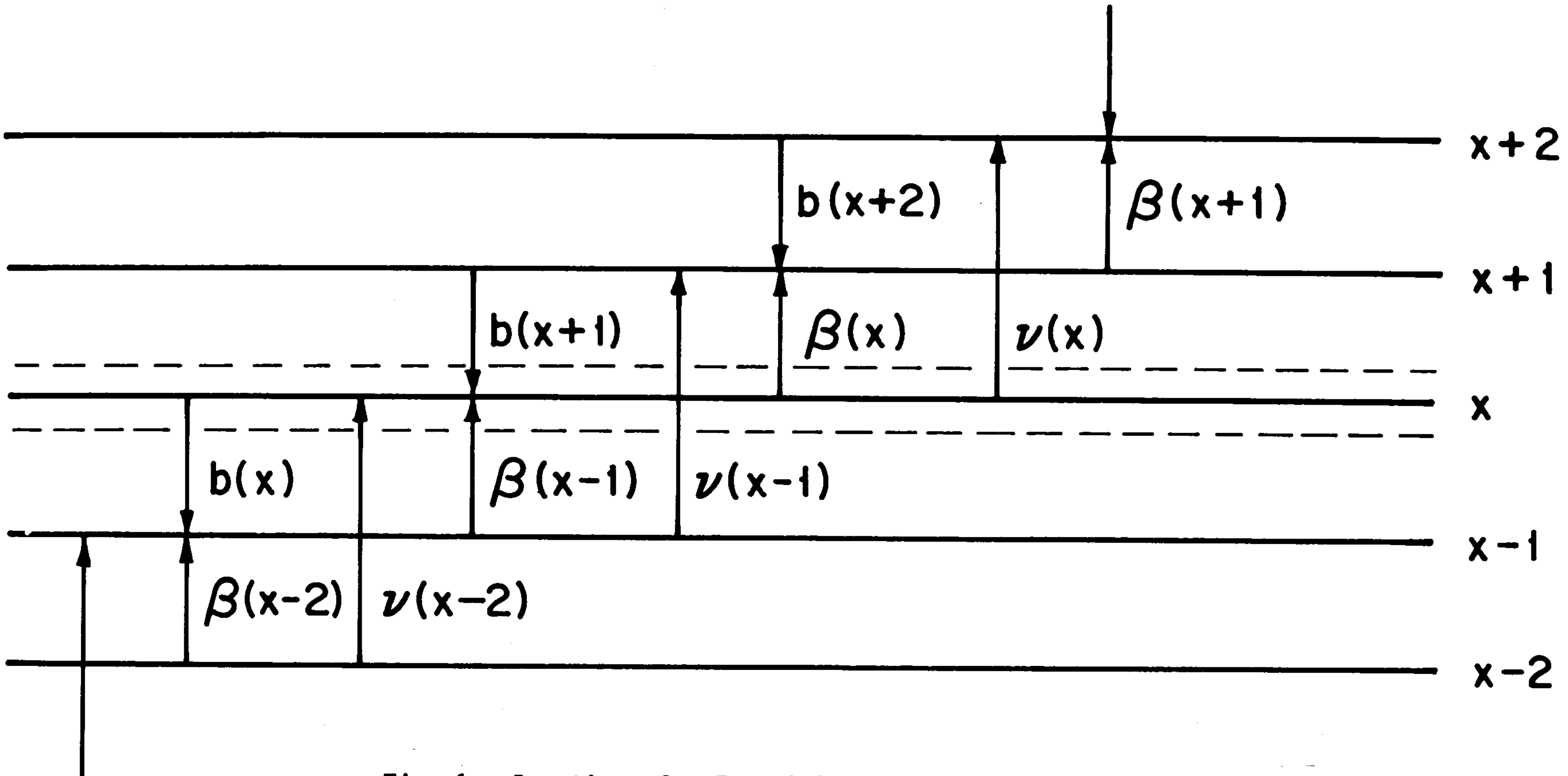

Fig. 1. Reactions for Transitions Between Vacancy Clusters.

$$df(x,t)/dt = I(x-1,t) - I(x,t)\ . \tag{3}$$

Equations (1) to (3) hold for the immobile vacancy clusters, i.e. for $x > 3$. The rate equations for the monovacancy concentration $f(1,t) = C_V$ is given by

$$\begin{aligned} df(1,t)/dt = P_V &- [\alpha(1) + \beta(1) + \nu(1)]f(1,t) + [\alpha(2) + 2\gamma(2)]f(2,t) \\ &- \sum_{x=2}^{\infty} \beta(x)f(x,t) + \sum_{x=3}^{\infty} \gamma(x)f(x,t) - L_V\ . \end{aligned} \tag{4}$$

The first term represents the rate of monovacancy formation, P_V, by radiation-induced displacements, the second term is the sum of all the capture rates for mobile point defects by monovacancies, and the third term contains the dissociation reaction and interstitial capture by divacancies. The third and last term represent the loss of monovacancies captured at vacancy clusters and other sinks, respectively. Finally, the fifth term gives the rate of re-emission of vacancies from all immobile clusters. Thermal vacancy re-emission takes place also at sinks other than vacancy clusters. Therefore, L_V is the net absorption rate for monovacancies at these sinks, and can be written as a difference of two terms, namely vacancy capture minus thermal emission rate.

The rate equation for the divacancy concentration $f(2,t) = C_{2V}$ can be expressed as

$$\begin{aligned} df(2,t)/dt = P_{2V} &+ \beta(1)f(1,t) - [\alpha(2) + \beta(2) + \gamma(2) + \nu(2)]f(2,t) \\ &+ [\alpha(3) + \gamma(3)]f(3,t) - \sum_{x=3} \nu(x)f(x,t) - L_{2V}\ . \end{aligned} \tag{5}$$

The production term P_{2V} allows for the possible generation of divacancies by the radiation-induced displacements so that $(P_V + 2P_{2V})$ is the total production rate of Frenkel pairs. The second term in Eq. (5) represents the divacancy formation by the reaction of two monovacancies. All the other terms carry a similar meaning as those in Eq. (4). However, there are no terms in Eq. (5) which represents the thermally activated re-emission of divacancies from vacancy clusters or any other sink. Although such terms could be included in principle, we can neglect them because of the large formation energy of divacancies which is equal to twice the formation energy of monovacancies minus the binding energy of divacancies.

To complete the set of rate equations, we must finally specify the one for the mono-interstitial concentration C_I. As discussed in the introduction, we may neglect the presence of mobile interstitial clusters, and write

$$dC_I/dt = P_I - \alpha(1)f(1,t) - \sum_{x=2}^{\infty} \alpha(x)f(x,t) - L_I \tag{6}$$

where

$$P_I = P_V + 2P_{2V} , \tag{7}$$

and L_I is the capture rate of interstitials at sinks other than vacancies and vacancy clusters.

§3. STEADY STATE NUCLEATION

Nucleation phenomena are characterized by three stages. During the first stage a supersaturation of the mobile species is established by external means. The supersaturation drives then the evolution of a subcritical cluster distribution which reaches a quasi-stationary distribution during the second stage. In this stage, clusters may grow due to fluctuations beyond the critical size and become stabilized and members of the supercritical cluster population. In the third and final stage, the further nucleation of supercritical clusters ceases because the supersaturation drops or is depleted, or because the large population of the supercritical clusters competes more effectively for the mobile species to sustain their continued growth.

Nucleation theory deals with the second stage where a stationary subcritical cluster population $f_S(x)$ is assumed to exist. The cluster current $I(x,t)$ becomes then also independent of time, and independent of x as will be shown below.

Let us introduce first two formal definitions for the sake of convenience. Although $\nu(o)$ and $f_S(o)$ are devoid of any physical meaning we shall assume that

$$\nu(o) = f_S(o) = 0 \; . \tag{8}$$

Next we introduce a constraint cluster size distribution of the first kind by the recursive relationship

$$b(x + 1)n(x + 1) = a(x)n(x) + \nu(x - 1)n(x - 1) \quad , \quad x > 1 \tag{9}$$

and set

$$n(1) = C_V \; , \tag{10}$$

i.e. equal to the monovacancy concentration. For the stationary distribution $f_S(x)$ it follows then from Eq. (3) that $I_S(x - 1) = I_S(x)$ for $x > 2$, and hence $I_S(x) = I_S$ = constant. With the definition for I, Eq. (2), and the relationship (9), we can then derive

$$I_S = \nu(x - 1)n(x - 1)\Big[\big(f_S(x - 1)/n(x - 1)\big) - \big(f_S(x + 1)/n(x + 1)\big)\Big] \\ + a(x)n(x)\Big[\big(f_S(x)/n(x)\big) - \big(f_S(x + 1)/n(x + 1)\big)\Big] \; . \qquad (11)$$

Let us further introduce the quantity

$$\Delta(x) = \big(f_S(x)/n(x)\big) - \big(f_S(x + 1)/n(x + 1)\big) \qquad (12)$$

and write

$$\begin{aligned} I_S &= \nu(x - 1)n(x - 1)\Delta(x - 1) + [\alpha(x)n(x) + \nu(x - 1)n(x - 1)]\Delta(x) \\ &= \nu(x - 1)n(x - 1)\Delta(x - 1) + b(x + 1)n(x + 1)\Delta(x) \; . \end{aligned} \qquad (13)$$

Equation (13) is of the same form as in the nucleation theory with only near-neighbor transitions between clusters.

The case $I_S \equiv 0$ defines now a constrained distribution N(x) of the second kind by the recursive relationship

$$[(a,x)n(x) + \nu(x - 1)n(x - 1)]N(x) = \nu(x - 1)n(x - 1)N(x - 1) \qquad (14)$$

valid for $x > 2$, and normalized for convenience such that $N(1) \equiv 1$.

In terms of the functions $\Delta(x)$ and $N(x)$ we may write for the stationary nucleation current

$$I_S = [a(x)n(x) + \nu(x - 1)n(x - 1)][(\Delta(x - 1)/N(x - 1)) - (\Delta(x)/N(x))] \; . \quad (15)$$

In the usual manner, this equation is divided by the first factor and then summed over x. If a sufficiently large x_M exist for which $\Delta(x_M)$ becomes zero, then the steady state nucleation rate is given by

$$I_S = [1 - f_S(2)/n(2)]/[\sum_{x=2}^{x_M} [a(x)n(x) + \nu(x - 1)n(x - 1)]^{-1}] \quad (16)$$

where we used the fact that

$$f_S(1) = C_V = n(1) \; . \quad (17)$$

Equation (16) reproduces the result of previous nucleation theories when no divacancies are assumed to exist. In this case

$$f_S(2) = C_{2V} \quad (18)$$

is zero and all $\nu(x)$ vanish.

It is possible to define in a formal sense an activation energy for nucleation of a cluster of size x by

$$n(x) = n(1) \exp[-\Delta G(x)/kT] \; . \quad (19)$$

With the recursive relationship (9) we obtain then

$$\Delta G(x) = -kT \sum_{y=1}^{x-1} \ln[(a(y)/b(y+1)) + (\nu(y-1)/b(y+1))(n(y-1)/n(y))] \ . \tag{20}$$

The maximum of the activation energy for nucleation $\Delta G(x)$ determines now the critical cluster size x^*. Around this maximum

$$\Delta G(x^* - 1) \cong \Delta G(x^*) \cong \Delta G(x^* + 1) \ .$$

This condition is equivalent to

$$a(x^*) + \nu(x^* - 1) \cong b(x^* + 1) \ . \tag{21}$$

To satisfy this equation precisely will in general give a non-integer number for the critical size. Therefore x^* is the integer which satisfies the relationship (21) as closely as possible.

With the solution for I_S we can finally obtain the stationary subcritical cluster size distribution from the relationship

$$f_S(x+1)/n(x+1) = f_S(x)/n(x) + [(f_S(2)/n(2)) - 1][1 - \sum_{y=2}^{x} [a(y)n(y) + \nu(y-1)n(y-1)]^{-1} / \sum_{y=2}^{x_M} [a(y)n(y) + \nu(y-1)n(y-1)]^{-1}] \tag{22}$$

valid for $x > 2$. The above equations must be solved in a recursive manner. First, the steady state rate equations for mono-interstitials, monovacancies and divacancies are solved. Then $f_S(1) = C_V$, $f_S(2) = C_{2V}$, and $n(1) = C_V$.

Equation (9) gives then n(2) so that

$$f_S(2)/n(2) = C_{2V}/n(2) = (C_{2V}/C_V)[[\alpha(2) + \gamma(2)]/[\beta(1) + \nu(1)]] \ . \tag{23}$$

All the parameters are now known to start the recursive iterations to obtain n(x) and $f_S(x)$ for increasing x.

§4. RESULTS

To evaluate the void nucleation rate I_S, the reaction rate coefficients must be specified. Assuming that these reactions are diffusion controlled, they are given by

$$\alpha(x) = (4\pi)^{2/3}(3\Omega x)^{1/3}D_I C_I Z_I^o(x) \tag{24}$$

$$\beta(x) = (4\pi)^{2/3}(3\Omega x)^{1/3}D_V C_V Z_V^o(x) \tag{25}$$

$$\gamma(x) = (4\pi)^{2/3}(3\Omega x)^{1/3}D_V C_V^o(x) Z_V^o(x) \tag{26}$$

$$\nu(x) = (4\pi)^{2/3}(3\Omega x)^{1/3}D_{2V} C_{2V} Z_{2V}^o(x) \tag{27}$$

when x > 2. The above results are obtained by treating the vacancy cluster of size x as a spherical cavity of volume $x\Omega$. The bias factors Z_I^o, Z_V^o, and Z_{2V}^o take into account the effect of the interaction of the migrating point defect with the void. These factors have been obtained previously (Wolfer and Mansur 1980) for interstitials and vacancies. We assume in the following that $Z_{2V}^o = Z_V^o$.

The rate coefficient for vacancy emission depends on the vacancy concentration in thermal equilibrium with the cluster of size x. This concentration is given by

$$C_V^o(x) = C_V^{eq} \left(A^o(x - 1)Z_V^o(x - 1)/A_V^o(x)Z^o(x)\right) \exp\{[w(x) - w(x - 1)/kT\} \qquad (28)$$

where

$$A^o(x) = (4\pi)^{2/3}(3\Omega x)^{1/3} \qquad (29)$$

and

$$w(x) = (4\pi)^{1/3}(3\Omega x)^{2/3}\Theta(x,T) \qquad (30)$$

is the energy of the spherical void. The surface energy $\Theta(x,T)$ contains corrections for small cluster sizes and varying temperatures, as discussed previously (Si-Ahmed and Wolfer 1982), and it can be written into the form

$$\Theta(x,T) = \Theta_0(T)\left(1 - \ (0.8/x + 2)\right) \qquad (31)$$

where $\Theta_0(T)$ is the plane surface energy.

For the reaction rates among the mobile species, the following expressions are used. The recombination rate can be written as

$$\alpha(1)f_S(1) = 4\pi R_{IV}(D_I + D_V)C_I C_V \ , \qquad (32)$$

the divacancy formation rate as

$$\beta(1)f_S(1) = 4\pi R_{VV}\ 2D_V C_V^2\ , \tag{33}$$

and the trivacancy rate as

$$\nu(1)f_S(1) = 4\pi R_{V2V}(D_V + D_{2V})C_V C_{2V}\ . \tag{34}$$

The formation rate of vacancies by the reaction of interstitials and divacancies is given by

$$\alpha(2)f_S(2) = 4\pi R_{I2V}(D_I + D_{2V})C_I C_{2V}\ . \tag{35}$$

The recombination radius R_{IV} has recently been evaluated by Wolfer and Si-Ahmed (1981) and found to be about equal to twice the lattice parameter a_o at elevated temperatures. Estimates for the other reaction radii have been obtained by Si-Ahmed (1981), and the appropriate values are listed in Table 2.

The divacancy dissociation rate coefficient $\gamma(2)$ is obtained by considering thermal equilibrium. In this case, detailed balance requires that

$$\gamma(2)\ C_{2V}^{eq} = 4\pi R_{VV} D_V (C_V^{eq})^2$$

where (Seeger and Mehrer 1970)

$$C_{2V}^{eq} = 6\Omega(C_V^{eq})^2\ \exp[(E_{2V}^b + T\Delta S_{2V})/kT]$$

Table 2

Parameter	Definition	Value
e	fraction of displaced atoms escaping the collision cascade	0.25
P_I	interstitial production rate	e*dpa/s
P_V	vacancy production rate	$P_I(1 - \varepsilon_{2V})$
P_{2V}	divacancy production rate	$P_I\varepsilon_{2V}/2$
ε_{2V}	fraction of vacancies escaping the collision cascade as divacancies	0 to 0.2
R_{IV}	recombination radius	2 a_o
R_{I2V}	reaction radius between an interstitial and a divacancy	2 a_o
R_{VV}	reaction radius between vacancies	1.5 a_o
$D_V{}^{\dagger}$	vacancy migration coefficient	$1.53 \times 10^{-6} \exp[-E_V^m/kT]$ m^2/s
$D_{2V}{}^{\dagger}$	divacancy migration coefficient	$1.0 \times 10^{-7} \exp[-E_{2V}^m/kT]$ m^2/s
$D_I{}^{\ddagger}$	interstitial migration coefficient	$8.0 \times 10^{-7} \exp[-E_I^m/kT]$ m^2s
E_V^m †	vacancy migration energy	2.24×10^{-19} J (1.4 eV)
E_{2V}^m †	divacancy migration energy	1.31×10^{-19} J (0.82 eV)
E_I^m ‡	interstitial migration energy	2.4×10^{-20} J (0.15 eV)
C_V^{eq} †	equilibrium vacancy concentration	$\exp[1.5 - E_V^f/kT]/\Omega$
E_V^f †	vacancy formation energy	2.56×10^{-19} J (1.6 eV)

Table 2. (Continued)

Parameter	Definition	Value
Ω	atomic volume	$a_0^3/4$
a_0	lattice parameter	0.352 nm
$\Theta_0(T)$ §	surface energy for austenitic stainless steel	$1 + (773 - T) * 1.8 \times 10^{-3}$ J
E_{2V}^{b} ‡	divacancy binding energy	4.8×10^{-20} J (0.3 eV)
ΔS_{2V} †	entropy change upon divacancy formation	1.8

† Seeger and Mehrer (1970)

‡ Yoo (1979)

§ Murr (1975); Si-Ahmed and Wolfer (1982) is the equilibrium divacancy concentration. Hence, with $R_{VV} = 1.5\ a_o$, we obtain

$$\gamma(2) = \frac{16\pi}{3a_o^2} D_V \exp[-(E_{2V}^b + T\Delta S_{2V})/kT] \ . \tag{36}$$

For the loss of the mobile point defects to dislocations, a sink strength of $2 \times 10^{13}\ m^{-2}$ was chosen together with a bias factor of 1.2 for preferential interstitial absorption.

Equations (4), (5), and (6) were solved numerically for the steady state concentrations C_V, C_{2V}, and C_I assuming that the loss to the subcritical cluster population is negligible. This was confirmed subsequently after having obtained the steady state nucleation rate and the subcritical distribution.

Figures 2 and 3 show the steady state nucleation rates for bare voids, i.e. without the formation of a segregation shell. The parameters used and listed in Table 2 are applicable to nickel alloys and austenitic stainless steels. The solid lines are based on the new void nucleation theory, whereas the dashed curves are results of the previous nucleation theory in which divacancies are neglected. We note first that the inclusion of divacancies in the nucleation process increases the void nucleation rate by 3 to 6 orders of magnitude depending on temperature and displacement rate. If a certain fraction of vacancies becomes already clustered as divacancies during the short-term annealing processes in the collision cascade, this has a noticeable but secondary effect on the void nucleation rate.

In spite of the substantial enhancement of the void nucleation rate due to divacancies, the computed rates for bare voids are still too low to be

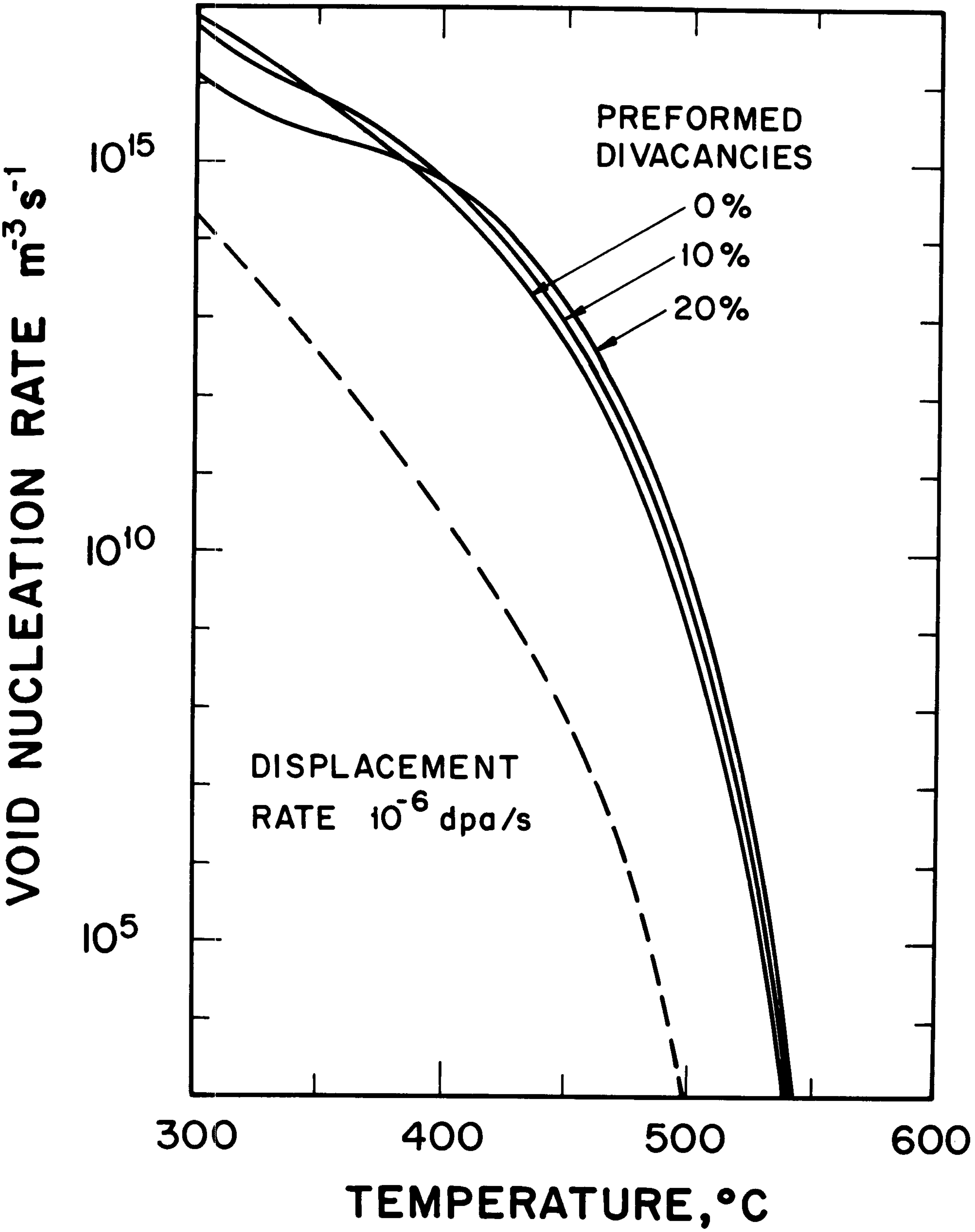

Fig. 2. Void Nucleation Rate Without Segregation at 10^{-6} dpa/s.

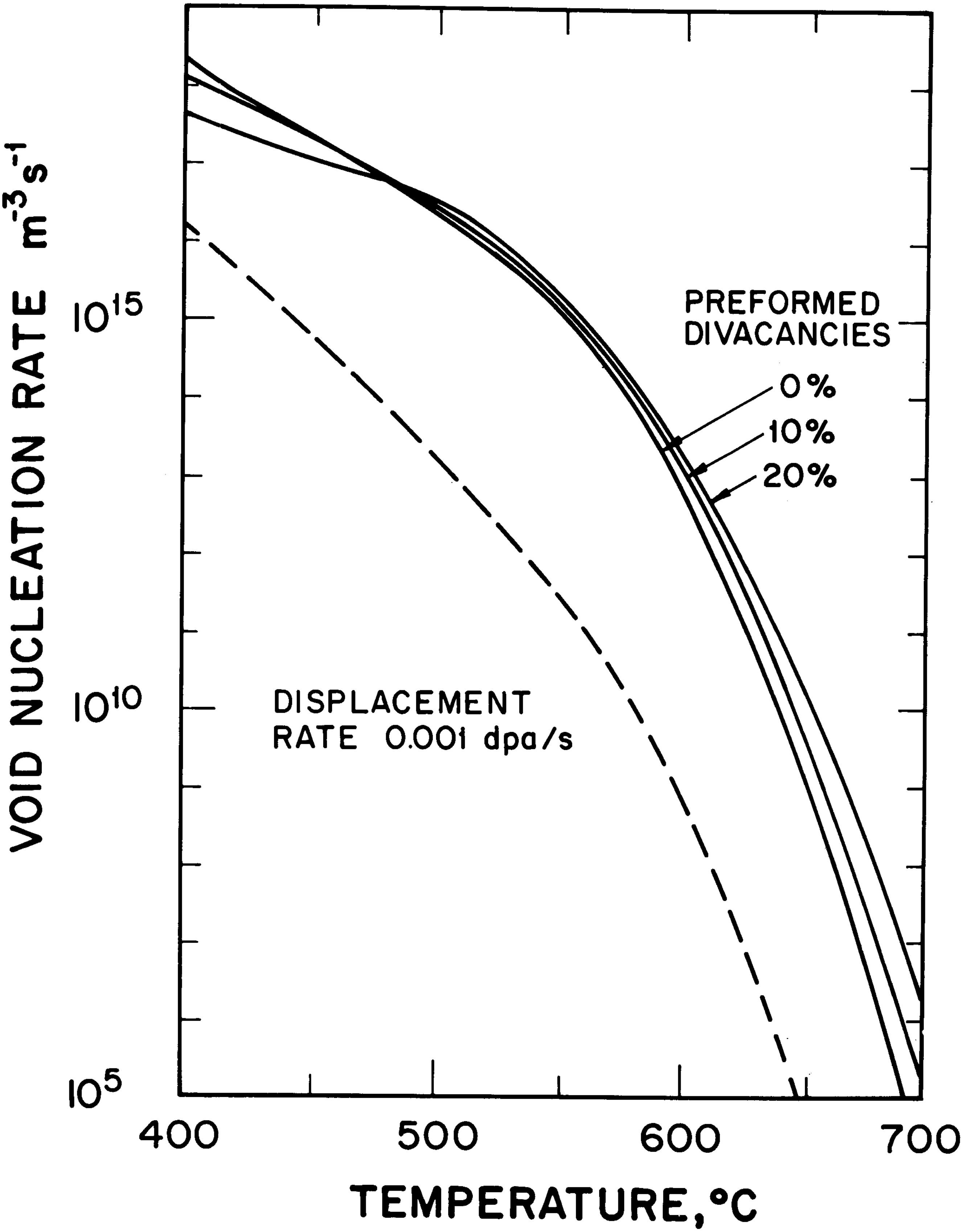

Fig. 3. Void Nucleation Rate Without Segregation at 10^{-3} dpa/s.

comparable to experimentally observed rates. Therefore, it is still necessary to invoke the presence of impurities which segregate to the subcritical clusters. As described in previous publications (Wolfer and Mansur 1980, Si-Ahmed and Wolfer 1982) we may model the effect of segregation by assuming, for example, that the region of segregation forms a shell with an elastic modulus slightly larger than the surrounding matrix. Other changes of lattice properties may also be assumed. For the present study, however, it was assumed that the relative shear modulus difference between shell and matrix is $1 - g = 0.02$. The computed nucleation rates are shown in Figs. 4 and 5 for displacement rates of 10^{-6} dpa/s and 10^{-3} dpa/s, respectively. Again, the inclusion of divacancies in the nucleation theory results in an increase in void formation rate. However, the enhancement relative to the previous nucleation theory is only one to three orders of magnitude. Segregation, on the other hand, increases the rate of void nucleation by about 6 to 8 orders of magnitude.

§5. DISCUSSION

Mobile divacancies were previously shown (Yoo 1979) to increase the void growth rate in the lower temperature range of swelling. This increase can be explained by the behavior of the effective vacancy diffusion coefficient

$$D_V^{eff} = (D_V C_V + 2D_{2V} C_{2V})/(C_V + 2C_{2V}) \qquad (37)$$

as a function of temperature. As shown in Fig. 6, it is seen that the divacancies with their higher mobility enhance the effective diffusion at lower temperatures. This situation is similar to the effect of fast-diffusing solute atoms which also increase the effective vacancy diffusion. A recent

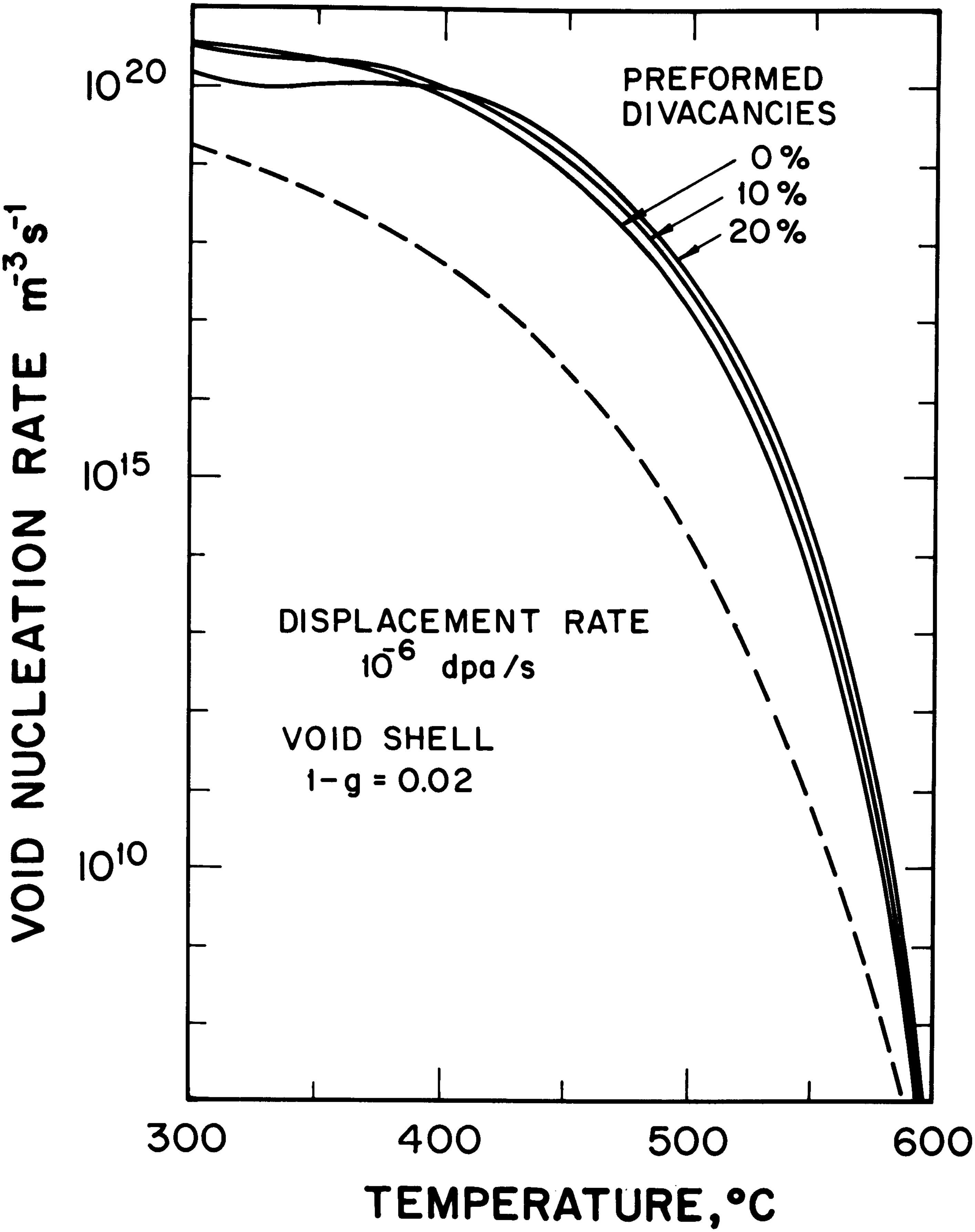

Fig. 4. Void Nucleation Rate With Segregation at 10^{-6} dpa/s.

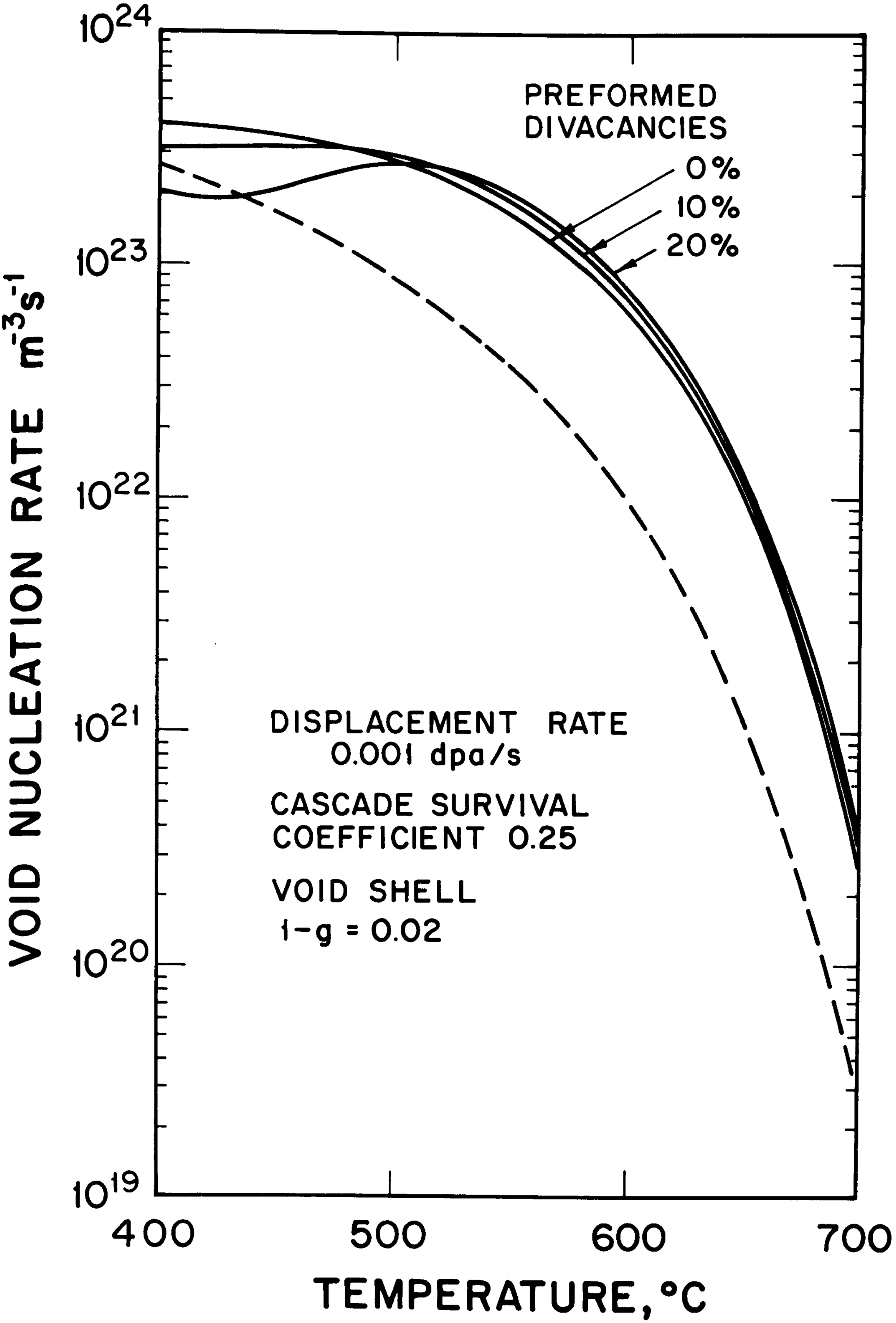

Fig. 5. Void Nucleation Rate With Segregation at 10^{-3} dpa/s.

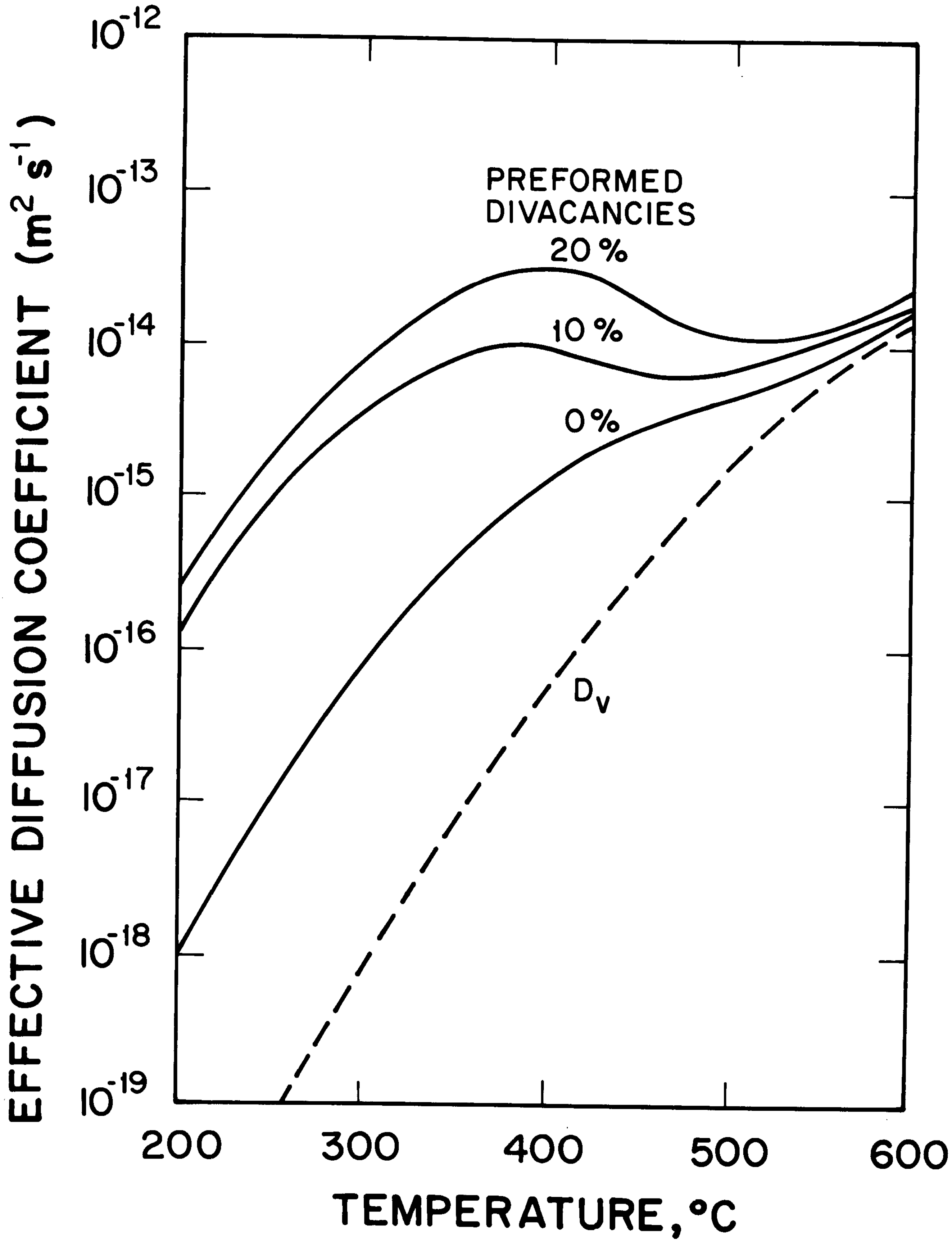

Fig. 6. Effective Vacancy Diffusion Coefficient.

study by Garner and Wolfer (1981) has however shown that when solute atoms increase the vacancy diffusion, the void nucleation rate is reduced. The reason for this result is that the vacancy supersaturation drops as the vacancies become more mobile.

One must therefore conclude that the effect of divacancies on void nucleation cannot simply be explained on the basis of an effective vacancy diffusion coefficient alone.

The nucleation rate can also be increased when the fluctuations in the growth rate of critical clusters are enhanced. In order to explore this possibility we assume that the critical size $x^* >> 1$. Therefore, in the vicinity of the critical size, we may consider the cluster distribution function $f(x,t)$ and the reaction rates to be continuous functions of x. We expand then all functions of $(x - 2)$, $(x - 1)$, and $(x + 1)$ in Eq. (1) into Taylor series, and obtain the so-called Kramers-Moyal expansion for the Eq. (1). Truncation after the second derivative leads to the Fokker-Planck equation

$$\partial f(x,t)/\partial t = -\partial/\partial x \left\{\tilde{F}(x) - (\partial/\partial x)\tilde{D}(x)\right\}f(x,t) \qquad (38)$$

where

$$\tilde{F}(x) = \beta(x) + 2\nu(x) - \alpha(x) - \gamma(x) \qquad (39)$$

is proportional to the average net growth of clusters with size x, and

$$\tilde{D}(x) = \frac{1}{2}\left[\beta(x) + 4\nu(x) + \alpha(x) + \gamma(x)\right] \qquad (40)$$

is the diffusion coefficient for the random walk of clusters in the size space. For the critical size $x = x^*$, it is seen that Eq. (39) is almost identical to Eq. (21), particularly when $x^* \gg 1$. Therefore, the critical size can also be determined by the alternate equation $\tilde{F}(x^*) \cong 0$.

In the vicinity of the critical size

$$\tilde{D}(x^*) \cong \beta(x^*) + 3\nu(x^*) = (4\pi)^{2/3}(3\Omega x^*)^{1/3} Z_V^o(x^*)[D_V C_V + 3D_{2V}C_{2V}] \, . \qquad (41)$$

If $C_V^t = C_V + 2C_{2V}$ denotes the total vacancy concentration, we can write

$$D_V C_V + 3D_{2V}C_{2V} = D_V^{eff} C_V^t + D_{2V}C_{2V}$$

which shows that $D(x^*)$ is greater than what would be expected based on the concept of effective vacancy diffusion alone.

We can therefore conclude that divacancies increase the void nucleation rate because their absorption enhances the growth fluctuations of critical clusters, and hence, their chance to pass from the subcritical to the supercritical size range. This effect more than compensates the opposite reduction of void nucleation by the increased vacancy mobility, as our numerical results demonstrate.

That the role of divacancies on nucleation differs from their role of void growth is further illustrated by the effect of preformed divacancies. These divacancies are thought to be formed following the temperature decay in the collision cascades. Since little information is available, various preformed fractions were considered in this paper and by Yoo (1979). Whereas their effect on void growth is pronounced, void nucleation is insensitive to preformed divacancies. Si-Ahmed (1981) has also investigated the effect of larger, immobile vacancy clusters formed during the short term annealing in collision cascades. It was found that their effect on void nucleation is negligible. The present result is therefore a further indication that differences in the primary defect production obtained in different radiation environments is not an important factor in void nucleation.

Acknowledgment

This work has been supported by the Division for Basic Energy Sciences, U.S. Department of Energy under contract ER-78-S-02-4861 with the University of Wisconsin.

References

Brailsford, A. D. and Bullough, R., 1972, J. Nucl. Mater. 44, 121.

Ehrhart, P. and Schlagheck, U., 1974, J. Phys. F: Metal Phys. 4, 1589.

Garner, F. A. and Wolfer, W. G., 1981, J. Nucl. Mater. 102, 143.

Johnson, R. A., 1966, Phys. Rev. 152, 629.

Katz, J. L. and Wiedersich, H., 1971, J. Chem. Phys. 55, 1414.

Murr, L. E., 1975, "Interfacial Phenomena in Metals and Alloys" (New York: Addison-Wesley).

Russell, K. D., 1971, Acta Met. 19, 753.

Schober, H. R., 1977, J. Phys. F: Metal Phys. 7, 1127.

Seeger, A. and Mehrer, H., 1970, "Vacancies and Interstitials in Metals," ed. by A. Seeger, D. Schumacher, W. Schilling, and J. Diehl (Amsterdam: North-Holland), p. 1.

Si-Ahmed, A., 1981, Doctoral Thesis, Univ. of Wisconsin.

Si-Ahmed, A. and Wolfer, W. G., 1982, Proc. of the 11th Internat. Symp. Effects of Rad. on Mater., to be published.

Wolfer, W. G. and Mansur, L. K., 1980, J. Nucl. Mater. 91, 265.

Wolfer, W. G. and Si-Ahmed, A., 1981, J. Nucl. Mater. 99, 117.

Yoo, M. H., 1979, Phil. Mag. A 40, 193.