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ABSTRACT: The effect of segregation on void nucleation is investigated utilizing previous results for the capture efficiency of coated voids. First, it is shown that any segregation, whether or not it leads to actual precipitation, leads to a modification of the bias factors for any sink. Small increases of either the lattice parameters or the elastic moduli result in reduced interstitial bias factors. Second, segregations to void embryos not only changes their capture efficiencies but also the surface energy. The effect of these changes on the void nucleation rate is studied in quantitative terms. When the segregation to voids results in an increase of the local lattice parameter by 0.4% or an increase of the shear modulus by 3%, the ultimate void nucleation rate is reached. Further increases no longer enhance void nucleation. Void nucleation without segregation effects would only be possible if the dislocation bias exceeds 50%. With segregation, void nucleation is not strongly dependent on the dislocation bias.

KEY WORDS: radiation effects, void swelling, segregation, diffusion, vacancy, interstitial, alloys

Segregation to voids has a dramatic effect on the void bias as has been shown recently [1,2]. These bias changes can be traced to the composition dependence of the elastic constants, the lattice parameter, and to the Kirkendall effect. One of the major goals of the present paper is to evaluate the magnitude of the parameter changes required to significantly affect the void nucleation rate. This rate depends sensitively on the bias of both voids and dislocations. This strong dependence can in fact be shown explicitly in the void nucleation theories of Katz and Wiedersich [3] and Russell [4] by reformulating the theory in terms of the various bias factors. Accordingly, we briefly review the void nucleation theory and cast it into a somewhat different form. Radiation-induced segregation to voids

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can also affect the surface energy, and this matter is further discussed below. Results pertaining to void nucleation under neutron irradiation are then given for nickel or austenitic stainless steels.

BIAS OF VOIDS WITH SEGREGATION

Radiation-induced segregation to voids or any other sinks introduces additional drift terms in the diffusion equations for both vacancies and interstitials. It has been shown [2,5] that the diffusion fluxes to a void in a substitutional binary alloy are given by

$$J_V = -\nabla(D_V C_V) - \frac{D_V C_V}{kT} \nabla G_V^S + (D_{AV} - D_{BV}) \alpha C_V \nabla x_A \tag{1}$$

for the vacancies, and by

$$J_I = -\nabla(D_I C_I) - \frac{D_I C_I}{kT} \nabla G_I^S - (D_{AI} - D_{BI}) \alpha C_I \nabla x_A \tag{2}$$

for the interstitials. Here, C_V and C_I are the concentration of vacancies and interstitials, D_V and D_I their diffusion coefficients for migration, and D_{AV} and D_{AI} are the atomic diffusivities for the A atoms migrating via vacancies and interstitials, respectively. The last term in the above equations is the Kirkendall drift term. It is proportional to the gradient of the A atom fraction, $x_A = 1 - x_B$, and to the thermodynamic factor

$$\alpha = 1 + \partial \ln \gamma_A / \partial \ln x_A \tag{3}$$

where γ_A is the activity coefficient.

The second term in Eqs. (1) and (2) consists of several drift terms as the Gibbs free energies for the point defects in their saddle-point configuration are composed of

$$G^S = U^\sigma + U^{Im} + U^C + G^f + G^m \ . \tag{4}$$

Here, G^f and G^m are the formation and the migration energy of the point defect, U^C is the interaction with the coherency strain field, U^{Im} is the image interaction, and U^σ is the interaction with the stress field produced by the void. As discussed in a companion paper [2], G^f and G^m are dependent on the alloy composition x_A. When we model the point defect as an inclusion the energy $(G^f + G^m)$ is essentially equal to the strain or bulk relaxation energy given by

$$U^{BR} = \frac{2\kappa\mu}{3\kappa + 4\mu} \frac{v^2}{\Omega} \ , \tag{5}$$

where κ and μ are the bulk and shear modulus, respectively, v is the relaxation volume of

the point defect, and Ω is the atomic volume. By virtue of the fact that the elastic moduli depend on the alloy composition, U^{BR} becomes a function of x_A.

The interaction with the coherency strain field, U^C, depends on the local lattice parameter $a_o(x_A)$ according to

$$U^C = \frac{3\kappa 4\mu}{3\kappa + 4\mu} v n(x_A) \tag{6}$$

where:

$$n(x_A) = [a_o(x_A) - \bar{a}_o]/\bar{a}_o \tag{7}$$

is the variation of the lattice parameter with respect to its average value $\bar{a}_o$.

Disregarding the Kirkendall drift terms we find that the remaining drift is due to the force

$$\nabla G^S = \nabla U^\sigma + \nabla U^{Im} + \left(\frac{dU^C}{dx_A} + \frac{dU^{BR}}{dx_A}\right) \nabla x_A \ . \tag{8}$$

It is seen that in addition to the first two forces present when there is no segregation, two new forces arise. As shown in a companion paper [2], the Kirkendall drift term for interstitials is small compared to the ones proportional to ∇G_I^S. For vacancies, all the drift terms are relatively small. Therefore, in the present paper we shall neglect the Kirkendall drift terms. In this case, the void bias factors are given by

$$Z^o = \left\{\int_o^1 d\left(\frac{r_s}{r}\right) \exp[(G^S(r) - \bar{G}^S)/kT]\right\}^{-1} \tag{9}$$

where $\bar{G}^S$ is the average of $G^S(r)$ and r_s is the void radius.

Suppose that segregation produces a shell around the void such that in the region $r_s \leq r \leq r_m$, the interaction energy $U^C + U^{BR}$ differs from the average value by the small amount ΔU^* which we assume to be constant in the segregation shell. Then, the void bias factor can be written as [1]

$$Z^o = \left[\frac{1}{Z^b} + \frac{h}{r_s} \exp(\bar{U}/kT)[\exp(\Delta U^*/kT) - 1]\right]^{-1} \tag{10}$$

where

$$Z^b = \left\{\int_o^1 d\left(\frac{r_s}{r}\right) \exp[(U^\sigma + U^{Im})/kT]\right\}^{-1} \tag{11}$$

is the bias factor of a bare void, i.e. in the uniform material with no segregation, and

$$\exp(\bar{U}/kT) = \int_{r_s/r_m}^1 d\left(\frac{r_s}{r}\right) \exp[(U^\sigma + U^{Im})/kT] \ . \tag{12}$$

The above Eqs. (10) to (12) are almost identical to the ones derived previously by Wolfer and Mansur [1] where it was not assumed that ΔU^* is small compared to the average value of $U^C + U^{BR}$. As will be shown in this paper, only very small changes in the lattice parameter and the elastic moduli, and hence only small values of ΔU^*, are required for the shell region to significantly affect the void nucleation rate. Therefore, the much simpler derivation of the void bias factor derived here provides a more than adequate approximation. Furthermore, in the present derivation it is not essential that the segregation produces a discrete shell. For a continuous segregation gradient, ΔU^* is merely an average value for the deviation of $(U^C + U^{BR})$ from its value in the matrix.

EFFECT OF SEGREGATION, TEMPERATURE, AND CURVATURE ON THE SURFACE ENERGY

The vacancy re-emission from void nuclei is an important reaction in the nucleation process. As shown by Katz and Wiedersich [3], the re-emission rate of vacancies is proportional to $D_V C_V^o(x)$, where

$$C_V^o(x) = C_V^{eq} \frac{A_o(x-1)\, Z_V^o(x-1)}{A_o(x)\, Z_V^o(x)} \exp\{[w(x) - w(x-1)]/kT\} \tag{13}$$

is the vacancy concentration in thermodynamic equilibrium with a void containing x vacancies, and C_V^{eq} is the corresponding concentration in the perfect crystal. Furthermore,

$$A_o(x) = (4\pi)^{2/3} (3\Omega x)^{1/3} \tag{14}$$

and

$$w(x) = (4\pi)^{1/3} (3\Omega x)^{2/3} \theta(x,T) \; . \tag{15}$$

Here, $\theta(x,T)$ is the surface energy assumed to depend on segregation, temperature T, and void size x. $w(x)$ represents the energy required to form a spherical void containing x vacancies in the absence of gas.

Values for the surface energy reported for nickel and stainless steel at a temperature of 773 K are 2.28 J/m^2 and 2.10 J/m^2 [6], respectively. These values are for a clean and flat surface. If values of this magnitude were used in void nucleation calculations, the nucleation rate would be exceedingly low. Therefore, it has been customary to use much lower values [7-9] with, however, little justification. Segregation of impurity elements under thermodynamic equilibrium is driven by a reduction in surface energy, and it has been found that oxygen, sulfur, and phosphorus [10-12] lower the surface energy of iron by as much as a factor of two. Considering the low magnitude of the total surface area of void embryos, only a few appm of active surface elements are required to cause a substantial

lowering in the void surface energy. Accordingly, we select a value of $\theta_0 = 1$ J/m^2 for the energy of flat surface at 773 K for stainless steel. Since the surface energy depends on temperature, the temperature coefficient 1.8×10^{-3} J/m^2/K [6] of 304 stainless steel is employed to obtain

$$\theta_c(T) = \theta_0 + (773 - T) \times 1.8 \times 10^{-3} \text{ J/m}^2 \tag{16}$$

as the temperature dependent surface energy for a flat surface.

Questions have often been raised about the appropriateness of using the energy of a flat surface for small voids. The recent computer simulation work of Mruzik and Russell [13] on the energy of voids has shown that the ratio $\Theta(x)/\theta_c$ becomes equal to one for $x > 40$. For smaller values of x, $\Theta(x)$ becomes somewhat smaller than θ_c, but the ratio $\Theta(x)/\theta_c$ remains independent of temperature. Mruzik and Russell [13] performed their computer simulation for vacancy clusters with $x > 10$. By using the simulation results of Doyama and Cotterill [14,15] for the formation energy of mono-, di-, tri-, and tetra-vacancies, we can extend the values of $\Theta(x)/\theta_c$ to smaller vacancy clusters. All results can be fitted to the simple expression

$$\Theta(x,T) \cong \theta_c(T)\left(1 - \frac{0.8}{x + 2}\right) \tag{17}$$

with a 10% accuracy.

VOID NUCLEATION THEORY

The void nucleation process can be divided into three stages. During the first stage, a subcritical vacancy cluster population is built up until some of the clusters have grown to a certain critical size x* to be defined presently. After a time lag period τ, to be given below, the subcritical cluster population remains constant, and void nucleation proceeds at a constant rate I_S. After a sufficient number of voids have been formed, void nucleation is terminated. The incubation time for steady-state swelling to be reached is roughly inversely proportional to I_S.

If [3,4]

$$n(x) = C_V \exp[-\Delta G(x)/kT] \tag{18}$$

defines the constrained equilibrium cluster distribution, where

$$\Delta G(x) = -kT \sum_{j=2}^{x} \ln\{[\alpha(j) + \gamma(j)]/\beta(j - 1)\} \tag{19}$$

then the steady-state void nucleation rate is given by

$$I_S = \{ \sum_{i=1}^{N} \frac{1}{\beta(i)n(i)} \}^{-1} \tag{20}$$

where N is a sufficiently large number, and where the rate coefficients are defined as

$$\alpha(x) = (4\pi)^{2/3} (3\Omega x)^{1/3} D_I C_I Z_I^o(x) \tag{21}$$

$$\beta(x) = (4\pi)^{2/3} (3\Omega x)^{1/3} D_V C_V Z_V^o(x) \tag{22}$$

$$\gamma(x) = (4\pi)^{2/3} (3\Omega x)^{1/3} D_V C_V^o(x) Z_V^o(x) \ . \tag{23}$$

They represent the capture rate of interstitials, of vacancies, and the re-emission rate of vacancies, respectively. The first two factors in Eqs. (21) to (23) are equal to $4\pi r(x)$, where $r(x)$ is the void radius, whereas $Z_I^o(x)$ and $Z_V^o(x)$ are the bias factors for interstitials and vacancies as given above.

The lag time τ is given by [16,17]

$$\tau = I_S \sum_{i=1}^{N} \frac{1}{\beta(i)n(i)} \sum_{j=i+1}^{N} n(j) \sum_{k=j}^{N} \frac{1}{\beta(k)n(k)} \ . \tag{24}$$

The critical void size x^* is defined by the maximum of $\Delta G(x)$. When $\Delta G(x)$ has a pronounced and narrow maximum, then the time lag is given approximately by

$$\tau = \beta(x^*) \, n^2(x^*)/2I_S^2 \ . \tag{25}$$

In the present paper, the more accurate expression of Eq. (24) was used, and it was found that it gives a somewhat larger delay time than the approximate expression in Eq. (25). However, the difference is no larger than a factor of two.

The concentrations of vacancies and interstitials, C_V and C_I, are obtained in the usual manner by solving the two rate equations [8]. The solutions can be expressed in the form

$$D_V C_V = \langle Z_I \rangle D_V (F + \langle C_V \rangle) \tag{26}$$

and

$$D_I C_I = \langle Z_V \rangle D_V F \tag{27}$$

where

$$F = \frac{\langle N \rangle}{2QD_V} \{ ([1 + M]^2 + L)^{1/2} - M \} \tag{28}$$

$$M = 1 + QD_V \langle C_V \rangle / \langle N \rangle \langle Z_I \rangle \tag{29}$$

$$L = 4PQ/\langle N \rangle^2 \langle Z_I \rangle \langle Z_V \rangle \ . \tag{30}$$

Here, <N> is the total sink strength, P is the production rate of vacancies and interstitials, and

$$Q = 4\pi R_c / D_V \tag{31}$$

is a coefficient related to the recombination rate. R_c is the recombination radius of Frenkel pairs which was recently shown to be about twice the lattice parameter [18].

In addition, the above equation contains the sink-averaged bias factors

$$\langle Z_{I,V} \rangle = \sum_S N_S A_S Z^S_{I,V} / \langle N \rangle \tag{32}$$

and the sink averaged thermal vacancy concentration

$$\langle C_V \rangle = \sum_S N_S A_S Z^S_V C^S_V / \langle Z_V \rangle \langle N \rangle \ . \tag{33}$$

Here, $N_S A_S$ is the strength of the sink of type "s" which has the bias factors Z^S_V and Z^S_I for vacancies and interstitials, respectively. Finally, the thermal vacancy concentration in equilibrium with the sink "s" is C^S_V.

With the above definitions, the nucleation barrier of Eq. (19) can be written in the following form

$$\Delta G(x)/kT = \sum_{j=2}^{n} \ln \{1 + B(j) \frac{D_I D_I}{D_V C_V} + \frac{C^o_V(j) - \langle C_V \rangle}{C_V}\} \tag{34}$$

where

$$B(j) = \frac{A_o(j)\ Z^o_I(j)}{A_o(j-1)\ Z^o_V(j-1)} - \frac{\langle Z_I \rangle}{\langle Z_V \rangle} \tag{35}$$

may be interpreted as the net bias of a void containing j vacancies. Equation (34) displays in a very clear manner that there are two contributions to the nucleation barrier. The last term, $[C^o_V(x) - \langle C_V \rangle]/C_V$, makes a contribution only when the thermal vacancy concentration $C^o_V(x)$ in equilibrium with the void is both larger than $\langle C_V \rangle$ as well as not negligible compared with the radiation-enhanced vacancy concentration C_V. The latter condition is satisfied only at high temperatures. The second term, $B(j)D_I C_I / D_V C_V$, is important at all temperatures and contributes to the nucleation barrier $\Delta G(x)$ whenever the void bias factor ratio, the first term in Eq. (35), exceeds the average bias factor ratio, the second term in Eq. (35).

Segregation to voids affects both contributions to $\Delta G(x)$. First, as argued above the surface energy and hence $C^o_V(x)$ is lowered by segregation of active surface elements.

Second, the net bias of the void can also be reduced dramatically by segregation. As shown in the following section, this effect is important at all temperatures.

RESULTS

The following results for the void nucleation were obtained with the parameters listed in Table 1 and the ones used in Ref. [1]. These parameters are applicable to nickel, the 300 series of austenitic stainless steel, and fcc alloys of Fe, Ni and Cr. For the void capture efficiencies, previous results were used [1]. The results for the bare void case (i.e., voids with no segregation shell), are compared with the results of voids with a segregation shell having either a thickness of 0.1 or 0.2 of the void radius. The shells were assumed to have either a different lattice parameter, or a different shear modulus. Although both properties are expected to vary simultaneously, for the purpose of comparing their effects, we have studied separately the changes of both properties.

The effect of a lattice parameter change in the void shell on the nucleation barrier $\Delta G(x)$ is shown in Fig. 1 for an irradiation temperature of 773 K and a shell thickness of 0.1 of the void radius. It is seen that bare voids possess a large critical size of about 400 vacancies, and a large nucleation barrier. A substantial reduction of both nucleation parameters can be achieved when the void shell has a larger lattice parameter. On the

Table 1. Parameters Used in the Calculations

The other parameters can be found in Ref. [1].

Parameters	Symbol	Value
Lattice parameter	a_o	3.639×10^{-10} m
Equilibrium vacancy concentration	C_V^{eq}	$\frac{1}{\Omega} \exp[1.5 - E_V^f/kT]$
Sink averaged thermal vacancy concentration	$\langle C_V \rangle$	C_V^{eq}
Vacancy diffusion coefficient	D_V	$1.53 \times 10^{-6} \exp[-E_V^m/kT][m^2/s]$
Vacancy formation energy	E_V^f	2.563×10^{-19} J
Vacancy migration energy	E_V^m	2.24×10^{-19} J
Cascade survival coefficient	e	0.25
Dislocation sink strength	$\langle N \rangle$	2×10^{13} m^{-2}
Production rate of Frenkel pairs	P	$[7.5 \times 10^{-26} \phi e]/\Omega m^{-3} s^{-1}$
Fast flux	ϕ	$2 \times 10^{19} n/m^2/s$
Atomic volume	Ω	$a_o^3/4$

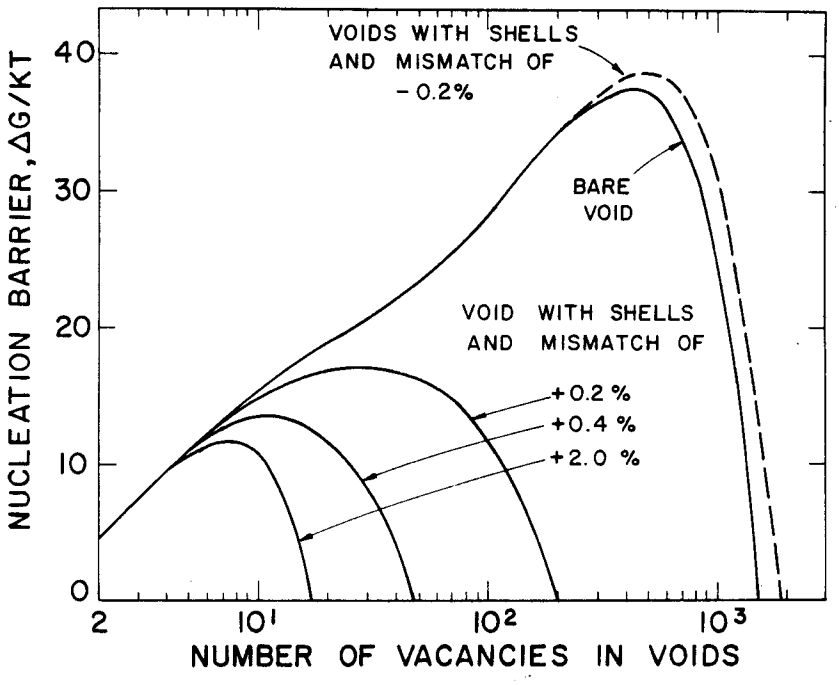

Fig. 1. Nucleation barrier as a function of the lattice parameter mismatch $(a_s - a_0)/a_0$, where a_s is the lattice parameter in the segregation shell and a_0 the average lattice parameter. The shear modulus difference is zero, the irradiation temperature is 773 K, and the shell thickness is 0.1 of the void radius.

other hand, a reduction of the lattice parameter in the void shell has a relatively small effect on the nucleation barrier. Therefore, a positive mismatch in the segregation shell leads to a drastic increase in the nucleation rate, whereas a negative mismatch further reduces the nucleation rate as compared to the rate obtained for bare voids. As seen in Fig. 2, void nucleation rates of 10^{15} m^{-3} sec^{-1} can only be obtained with a positive mismatch of at least 0.2% at irradiation temperatures below about 773 K. Nucleation rates of this magnitude are required in order to be compatible with experimental results from fast neutron irradiations. As pointed out earlier [14], void nucleation at higher temperatures requires furthermore the presence of helium or other insoluble gases.

An increase of the shear modulus within the segregation shell is equally effective in increasing the void nucleation rate. However, in order to obtain comparable results, the shear modulus must be larger by at least about 1% compared to the average value in the matrix. This is demonstrated by the results in Fig. 3. The nucleation rate is plotted against temperature for three different values of $(1 - g) = (\mu_s - \mu_m)/\mu_s$, where μ_s and μ_m

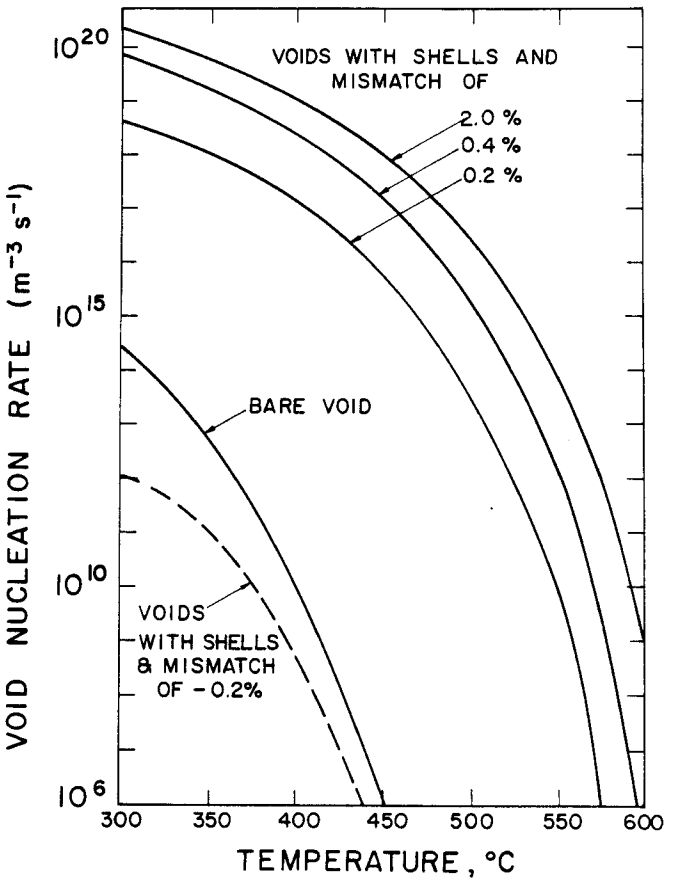

Fig. 2. Steady state void nucleation rate as a function of temperature and lattice parameter mismatch. The other parameters are the same as in Fig. 1.

are the shear moduli of the shell and the matrix, respectively. Although not shown in Fig. 3, a decrease in the shear modulus of the shell results in void nucleation rates somewhat less than for bare voids.

As indicated earlier, the initial increase in either the lattice parameter or the shear modulus within the segregation shell causes the largest increase in the void nucleation rate. The more detailed investigation into this effect gives results as shown in Figs. 4 and 5. It is seen that a positive lattice parameter mismatch greater than 0.4% and a shear modulus difference $(1 - g) > 0.03$ no longer result in further increases of the void nucleation rate. The reasons for this saturation behavior can be traced to the action of the segregation shell as a barrier for migration of point defects. A positive lattice parameter mismatch or a stiffer shell creates an activation barrier mainly for interstitials. Once the barrier reaches a certain height, the interstitials are prevented from entering the void, and any further increase in the barrier height will no longer increase

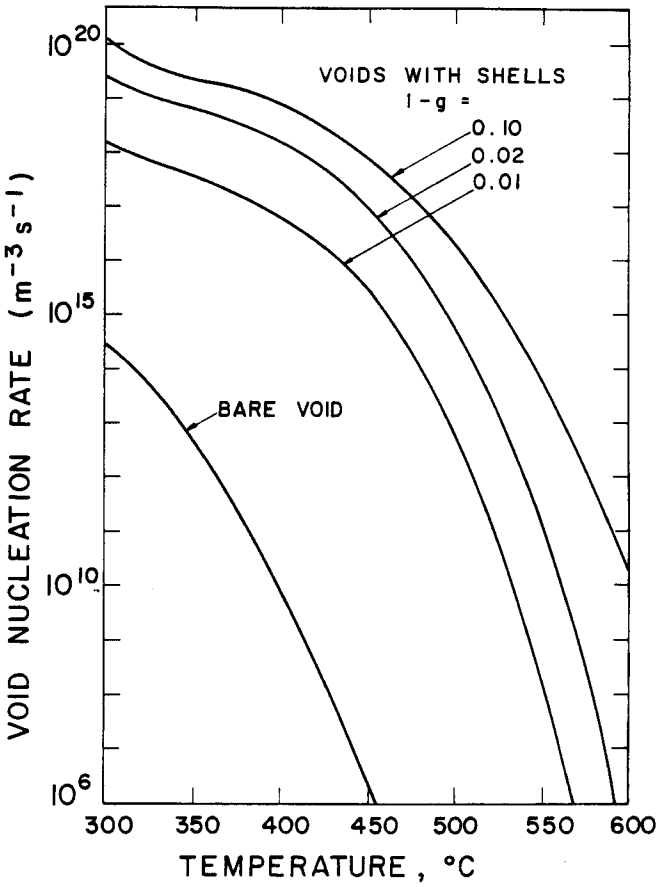

Fig. 3. Steady state void nucleation rate as a function of temperature and the shear modulus difference $(1 - g) = (\mu_s - \mu_m)/\mu_s$, where μ_s is the shear modulus of the segregation shell and μ_m the shear modulus of the matrix. The lattice parameter mismatch is zero, and the shell thickness is 0.1 of the void radius.

its effectiveness in blocking the interstitial flow. On the other hand, the segregation shell remains permeable to vacancies.

Segregation to voids not only influences the rate of void nucleation but also the time lag to reach steady state nucleation. A demonstration of this influence is given in Fig. 6. It should be noted that the delay time τ is different from the time required to reach the steady state swelling. The latter is often referred to as the incubation time for swelling. It is inversely proportional to the steady state nucleation rate I_S, and is several orders of magnitude larger than τ. Therefore, high void nucleation rates imply short incubation times for swelling.

It is commonly assumed that preferential absorption is both the driving force for void nucleation and void growth. This is certainly true for voids with no bias. However, bare voids possess a substantial image interaction with interstitials, and hence, also an interstitial bias. As a result, the dislocation bias would have to exceed the bare void bias for nucleation to occur. This is clearly shown in Fig. 7 which gives the void nucleation

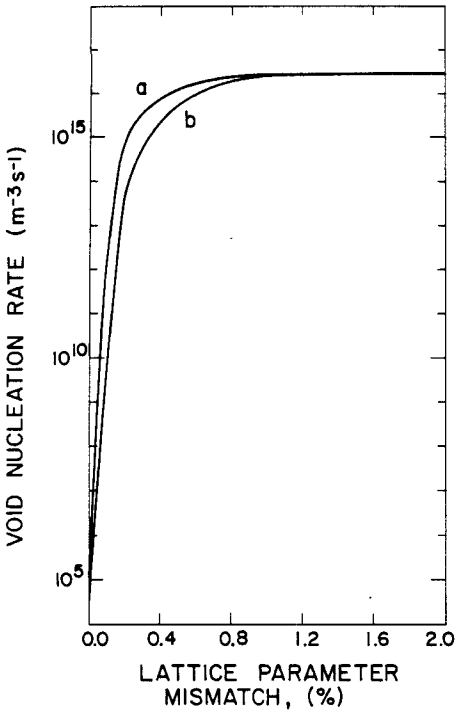

Fig. 4. Steady state void nucleation rate as a function of the lattice parameter mismatch, and for a shell thickness of (a) 0.2, or (b) 0.1 of the void radius. The shear modulus difference is zero, and the irradiation temperature is 773 K.

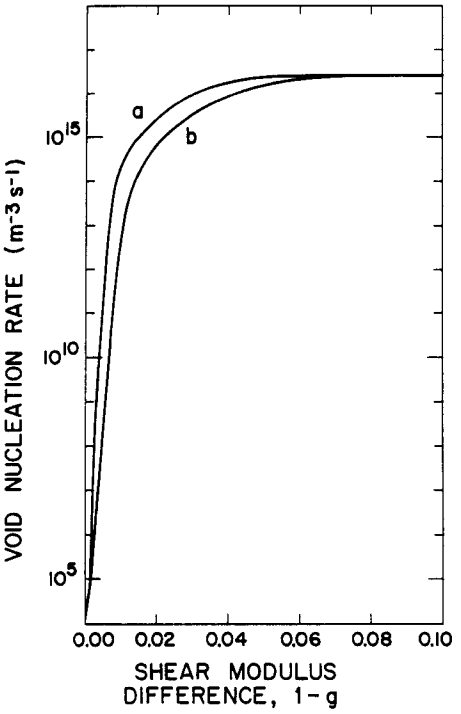

Fig. 5. Steady state void nucleation as a function of the shear modulus difference. The lattice parameter mismatch is zero, and the other parameters are the same as in Fig. 4.

rate with no segregation shell both as a function of the dislocation sink strength and as a function of the bias factor ratio $\langle Z_I \rangle / \langle Z_V \rangle$. It is seen that $\langle Z_I \rangle / \langle Z_V \rangle$ must exceed the value of at least 1.5 before substantial nucleation of bare voids becomes possible. A dislocation bias ratio larger than 1.5 is believed to be unrealistic, and this is a further indication that segregation to voids and formation of an interstitial barrier must accompany the void nucleation process.

Once a segregation shell has formed, the dislocation bias has a relatively minor effect on the void nucleation rate. This is shown in Fig. 8 for an irradiation temperature of 773 K and for different dislocation sink strengths. In fact, it can be seen that the dislocations could even be neutral sinks, and void nucleation with segregation could still proceed. Of course, a net bias does exist even in this case, but now the voids with shells have a bias against interstitials.

CONCLUSIONS

Segregation of impurities and major elements of alloys to voids can change the void

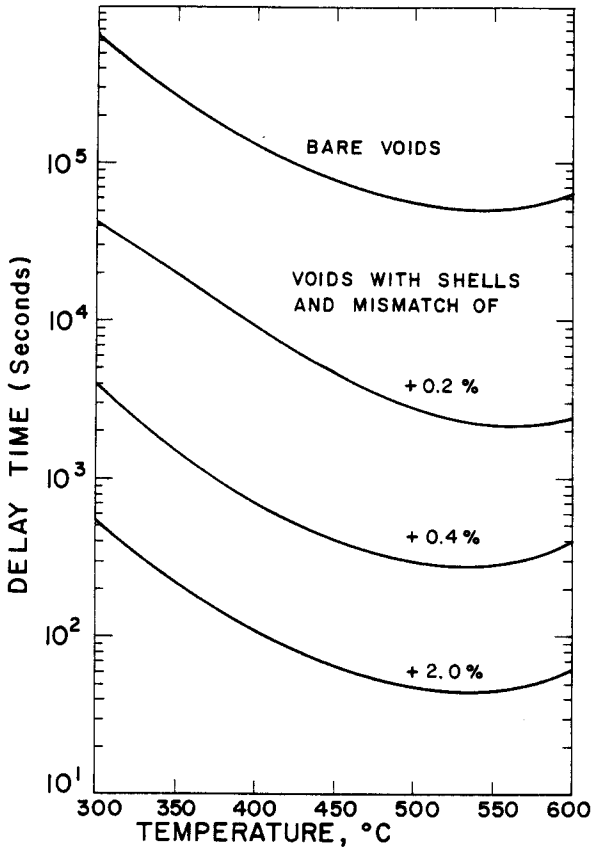

Fig. 6. Delay time as a function of temperature and lattice parameter mismatch. The parameters are the same as in Fig. 1.

nucleation rate dramatically. First, segregation of impurities lowers the surface energy. In order to obtain void nucleation rates as experimentally observed, the surface energy has to be about one half of the energy of a clean metal surface. The second effect of segregation is due to the void bias modification. Whereas a bare void has a significant bias due to the image interaction when it is small, segregation cannot only eliminate this interstitial bias but actually reverse it. The conditions for this to occur are that segregation has to slightly increase the lattice parameter around the void, leading to a compressive coherency strain field around the void which provides an interstitial barrier. The increase of the local lattice parameter need be no more than about 0.2%.

When segregation leads to an increase of the shear modulus around the void, an interstitial barrier is again created due to the fact that the interstitial formation energy increases with the shear modulus. Substantial enhancement of void nucleation results when the local modulus increase is about 2%.

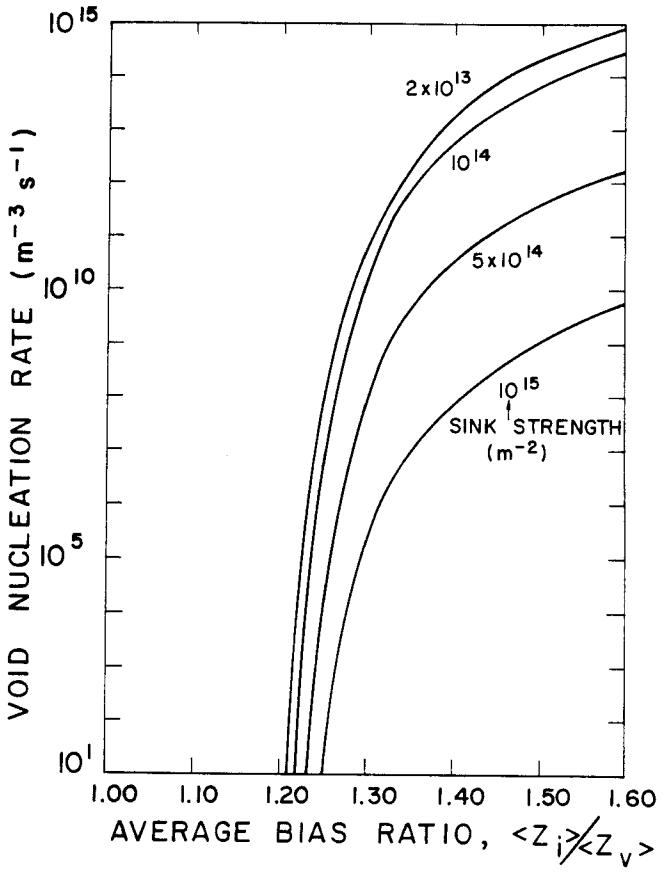

Fig. 7. Steady state void nucleation rate with no segregation shell as a function of the dislocation bias factor ratio and the dislocation sink strength. The irradiation temperature is 773 K.

In order to obtain comparable void nucleation rates without the segregation effects, a dislocation bias of 50% or greater would be required. Furthermore, void nucleation would depend very sensitively on the precise dislocation bias. On the other hand, when segregation effects are included in the void nucleation theory, the dislocation bias has only a minor effect on the nucleation rate. As a result, the evolution of the dislocation network during irradiation is not expected to play a major role in the incubation time for void swelling. Rather, the microchemical evolution connected with radiation-induced segregation appears to be the major controlling factor in void nucleation.

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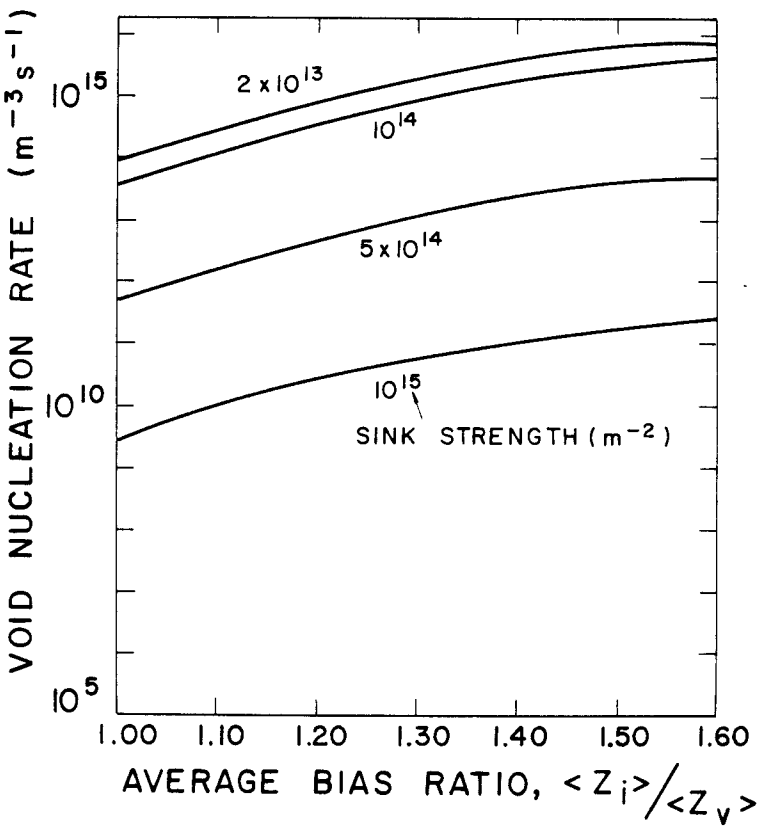

Fig. 8. Steady state void nucleation rate as a function of the dislocation bias factor ratio, and the dislocation sink strength. The segregation shell has a lattice parameter mismatch of 0.4%, the irradiation temperature is 773 K, and the other parameters are the same as in Fig. 1.

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