

THE EFFECT OF VOID SURFACE MOTION ON THE VOID SINK STRENGTH FOR POINT-DEFECTS

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Received 31 July 1984; accepted 7 August 1984

By a generalization of an analysis due to Frank of the growing precipitate we derive an analytic sink strength for the growing void that takes account of the void surface motion in a self-consistent fashion. The lower mobility of the vacancies compared to the interstitials ensures that a growing void captures more vacancies than the usual quasi-static void. The various consequences of this void bias for vacancies are discussed in relation to the swelling of reactor materials.

1. Introduction

Void growth in irradiated materials leads to a net volume increase, or swelling, of such materials and has been the subject of much experimental and theoretical investigation since Cawthorne and Fulton [1] first observed the phenomenon in stainless steel fast reactor fuel cladding. Such voids are usually considered to be relatively neutral sinks for mobile point-defects and their growth, in such materials, occurs because interstitials are lost preferentially at the dislocations so that, in a steady-state irradiation environment, there must be a consequent excess vacancy flux into relatively neutral sinks such as voids. The dependence of the kinetics of void swelling on the physical and irradiation parameters and on the overall microstructural state of materials has been extensively studied using the rate theory model of the total evolving microstructure [2–7]. A description of the microstructure is thus provided by replacing the crystalline material, with its spatially varying local point-defect concentrations in the neighbourhood of each sink, by an effective medium in which the point-defect concentrations are homogeneous and the actual sinks are replaced by effective sinks. The effective medium is thus a lossy continuum in which the various sink types, that together define the total microstructure,

each have an associated sink strength. The sink strengths for many of the important sink types have now been obtained using the consistent embedding procedure, the fundamental basis of which has been reviewed by Brailsford and Bullough [8]. In most of these sink strength calculations a quasi-static approximation is adopted in which the motion or transient morphological changes of the sink caused by the net flux of point-defects absorbed by it are assumed to have negligible effect upon the flux itself. Rauh and Bullough [9] have recently studied the effect of dropping this restriction for the edge dislocation sink; they have shown that the climb motion of the dislocations can have a significant effect upon their sink strength at high damage rates. In particular the climb motion increases the vacancy flux into a dislocation and thereby *reduces* its effective preference (or bias) for the interstitials which, in turn, reduces the actual climb rate.

The purpose of the present paper is to present an analogous, albeit simpler, analysis for the growing void in which the actual motion of the void surface is included in a self-consistent fashion. In section 2 we present the calculation of the self-consistent growing void sink strength using an embedding model valid for low sink densities. In addition, some limiting analytic features of the solution are discussed and related to the usual quasi-static results for void swelling in the presence of a second sink. A range of numerical evaluations of the general solution are described in section 3 using physical parameters appropriate for stainless steel. Finally in section 4, we discuss the practical relevance of the present growing void sink strength, together with its

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physical limitations and suggest how its significance could be investigated in the future.

2. The sink strength for the growing void

If the effective medium [8], which represents the real material, has total sink strength k_α^2 , where henceforth the subscript α can be i (for interstitial) or v (for vacancy) to distinguish the two point-defects, we have

$$k_\alpha^2 = k_{\alpha D}^2 + k_{\alpha C}^2, \quad (1)$$

where $k_{\alpha D}^2$ is the dislocation sink strength and $k_{\alpha C}^2$ is the void (Cavity) sink strength we seek; for simplicity the only sink types deemed to be present in the microstructure will be dislocations and voids. In the usual notation we write

$$k_{\alpha D}^2 = Z_\alpha \rho_D \quad (2)$$

where $Z_\alpha (Z_i > Z_v)$ are the dislocation bias parameters defining the dislocation preference for interstitials compared to vacancies and ρ_D is the edge dislocation density. For such a homogeneous medium the steady state point-defect concentration is

$$c_\alpha = K/D_\alpha k_\alpha^2 \quad (3)$$

where K is the point-defect production (damage) rate, assumed to be equal for vacancies and interstitials, D_α is the diffusion coefficient of the point-defect α and thermal emission of point-defects from the sinks together with loss of point-defects due to bulk recombination are neglected.

To obtain the exact consistent sink strength of the growing voids for interstitials and vacancies we must attempt to follow the embedding prescription [8] and identify at time t one of the voids embedded in the effective medium of (unknown) radius $r_C(t)$. The point defect concentration around such a void satisfies the conservation equation:

$$\frac{D_\alpha}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial c_\alpha}{\partial r} \right) + K - D_\alpha k_\alpha^2 c_\alpha = \frac{\partial c_\alpha}{\partial t} \quad (4)$$

where the centre of the void is located at the origin, $r = 0$ of a spherical coordinate system. In the effective medium away from the vicinity of the identified void the spatial and explicit transient variation of c_α must vanish and, from (3)

$$c_\alpha = c_\alpha^\infty = K/D_\alpha k_\alpha^2 \quad \text{as } r \rightarrow \infty. \quad (5)$$

For simplicity we may treat the void as an ideal sink for either point-defect and thus

$$c_\alpha = 0 \quad \text{at } r = r_C(t). \quad (6)$$

We can specify a small value (or zero) for the radius of the void at $t = 0$:

$$r_C(t) = r_C^0 \quad \text{at } t = 0. \quad (7)$$

Finally the consistent velocity of the void surface is given by the net vacancy flux into the void:

$$\dot{r}_C(t) = \left[D_v \frac{\partial c_v}{\partial r} - D_i \frac{\partial c_i}{\partial r} \right]_{r=r_C(t)}. \quad (8)$$

By equating the point-defect loss rate to the identified void

$$4\pi r_C^2(t) D_\alpha \frac{\partial c_\alpha}{\partial r} \Big|_{r_C(t)} \quad (9)$$

to the corresponding loss rate in the effective medium

$$k_{\alpha C}^2 D_\alpha c_\alpha^\infty / C_C, \quad (10)$$

where C_C is the volume concentration of voids, we obtain the implicit equation for the required growing void sink strength [4,8]

$$k_{\alpha C}^2 = \frac{4\pi r_C^2(t) C_C}{c_\alpha^\infty} \left[\frac{\partial c_\alpha}{\partial r} \right]_{r_C(t)}. \quad (11)$$

It is clear that to solve (4) when the boundary condition (6) is specified on a surface $r = r_C(t)$ whose value has to be consistently deduced by integrating (8) is a mathematically formidable problem; this is particularly true also when the $k_{\alpha C}^2$ component of k_α^2 in (4) is the quantity we seek from (11). To obtain a solution of these equations we can simplify the conservation eq. (4) by removing the explicit source and sink terms ($K - D_\alpha k_\alpha^2 c_\alpha$) with the knowledge that this approximation will yield a sink strength result that is correct to lowest order in the sink density [8]. When the void growth is neglected, as in the usual quasi-static approximation, eq. (4) becomes

$$\frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dc_\alpha}{dr} \right) = 0 \quad (12)$$

which, from (5), (6) and (11) yields the first-order neutral sink strength:

$$k_{\alpha C}^2 = k_C^2 = 4\pi r_C C_C. \quad (13)$$

The adoption of this approximation thus ensures that our final result will yield the correction to the lowest order void sink strength due to its growth and its use must therefore be restricted with this in mind. As discussed by Brailsford and Bullough [8] the approximation is only valid when the sink-sink interactive correction terms are negligible; that is when

$$k_\alpha r_C \ll 1. \quad (14)$$

Replacing eq. (4) by the dilute sink density form:

$$\frac{D_\alpha}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial c_\alpha}{\partial r} \right) = \frac{\partial c_\alpha}{\partial t} \quad (15)$$

and considering the growth of the voids from zero radius ($r_C^0 = 0$ in (7)) we can then proceed to obtain a consistent solution of (15) that satisfies (5), (6), (7) and (8). To do so we follow a previous analysis of the growing spherical precipitate due to Frank [10] and assume a trial function for $r_C(t)$ of the explicit form

$$r_C(t) = A\sqrt{t}. \quad (16)$$

We now seek a *constant* value for A such that the solution of (15) satisfies the boundary conditions (5), (6) and (7) together with the growth velocity condition (8). If A is constant then (16) automatically satisfies the initial condition (7), with r_C^0 set to zero. The required solution of (15) for the point-defect concentration c_α around the growing void may be obtained by replacing the variables (r, t) by the single variable s , such that

$$s = r/\sqrt{t}. \quad (17)$$

With this transformation eq. (15) is replaced by the ordinary differential equation

$$\frac{d^2 c_\alpha}{ds^2} + \left(\frac{s}{2D_\alpha} + \frac{2}{s} \right) \frac{dc_\alpha}{ds} = 0. \quad (18)$$

The solution of this equation and hence of the original eq. (15), that satisfies both (5) and (6) is, in terms of the r and t variables

$$c_\alpha = c_\alpha^\infty \left\{ 1 - \frac{F(r/\sqrt{t}, D_\alpha)}{F(A, D_\alpha)} \right\}, \quad (19)$$

where

$$F(x, y) = \frac{\exp(-x^2/4y)}{x} - \frac{1}{2} \sqrt{\frac{\pi}{y}} \operatorname{erfc}(x/2\sqrt{y}). \quad (20)$$

The value of A must now be both constant and consistent with the growth velocity eq. (8); substitution of (19) into (8) yields the required relation for A

$$\frac{A}{2} = c_v^\infty \sqrt{D_v} \psi(A/\sqrt{D_v}) - c_i^\infty \sqrt{D_i} \psi(A/\sqrt{D_i}) \quad (21)$$

where

$$\psi(z) = \frac{\exp(-z^2/4)}{z} / \left\{ \exp(-z^2/4) - \frac{z\sqrt{\pi}}{2} \operatorname{erfc}(z/2) \right\}. \quad (22)$$

The existence of the relation (21) for A confirms the validity of the trial solution (16) and that (19) is the

required solution for the point-defect concentration around the growing void.

The required growing void sink strengths now follow from (11) and (19) and may be written in the form

$$k_{\alpha C}^2 = k_C^2 \xi_\alpha \quad (23)$$

where

$$\xi_\alpha = (A/\sqrt{D_\alpha}) \psi(A/\sqrt{D_\alpha}) \quad (24)$$

are the growth correction factors on the first order neutral void sink strength k_C^2 , given by (13). The explicit relation for A now follows by substituting the c_α^∞ concentrations given, from (1), (2), (5) and (23), by

$$c_\alpha^\infty = K/D_\alpha (Z_\alpha \rho_D + k_C^2 \xi_\alpha), \quad (25)$$

into the relation (21) to yield

$$\begin{aligned} A & \left[Z_v \rho_D + k_C^2 (A/\sqrt{D_v}) \psi(A/\sqrt{D_v}) \right] \\ & \times \left[Z_i \rho_D + k_C^2 (A/\sqrt{D_i}) \psi(A/\sqrt{D_i}) \right] \\ & = 2K\rho_D \left[(Z_i/\sqrt{D_v}) \psi(A/\sqrt{D_v}) \right. \\ & \quad \left. - (Z_v/\sqrt{D_i}) \psi(A/\sqrt{D_i}) \right]. \end{aligned} \quad (26)$$

Before presenting results obtained by solving eq. (26) numerically it is instructive to replace (26) by an approximate form valid when the argument of the function ψ , defined by (22), is small. It is then easily shown that

$$\psi(z) \approx \frac{1}{z} / \left[1 - \frac{\sqrt{\pi}}{2} z + \frac{z^2}{2} \right] \text{ when } z < 2/\sqrt{\pi} \quad (27)$$

which, when substituted into (26) yields the quadratic equation for A :

$$\begin{aligned} & (Z_v \rho_D + k_C^2)(Z_i \rho_D + k_C^2) A^2 \\ & - K\rho_D \sqrt{\pi} (Z_v/\sqrt{D_v} - Z_i/\sqrt{D_i}) A \\ & - 2K\rho_D (Z_i - Z_v) = 0. \end{aligned} \quad (28)$$

Since $D_i \gg D_v$, the required positive real root of (28) is accurately given by:

$$A = \frac{K\rho_D \sqrt{\frac{\pi}{D_v}} Z + \left[\frac{K^2 \rho_D^2 \pi Z^2}{D_v} + 8k_0^4 K\rho_D (Z_i - Z_v) \right]^{1/2}}{2k_0^4} \quad (29)$$

where Z_i and Z_v have been replaced by Z when the small difference between them is unimportant and

$$k_0^2 = Z\rho_D + k_C^2. \quad (30)$$

Two extremes can be identified from (29):

(i) if the vacancies are very mobile (D_v large) then

the growth of the voids will have a negligible effect on their sink strength and (24) yields

$$A = A_b = [2K\rho_D(Z_i - Z_v)/k_0^4]^{1/2}, \quad (31)$$

where the subscript b indicates 'bias' dominated value.

(ii) Conversely when the vacancies are not very mobile (D_v small) the void growth will lead to extra vacancy capture and the value of A will increase towards

$$A = A_g = K\rho_D \sqrt{\frac{\pi}{D_v}} Z/k_0^4, \quad (32)$$

where the subscript g indicates 'growth' dominated value.

When the volume concentration of voids is C_C the expected void swelling rate must be

$$\frac{d}{dt} \left(\frac{\Delta V}{V} \right) = 4\pi r_C^2 \dot{r}_C C_C \quad (33)$$

and thus, from (16)

$$\frac{d}{dt} \left(\frac{\Delta V}{V} \right) = 2\pi A^3 t^{1/2}. \quad (34)$$

It is easily seen that (34), with A given by (31) is indeed the usual quasi-static swelling rate [4] when the effects of void growth are neglected together with the thermal emission and bulk recombination of the point-defects. In the other extreme, when void growth effects dominate over the bias growth (34) and (32) also lead to a non-zero swelling rate. We thus conclude that once void growth begins it will continue in the presence of a second sink even if that second sink is completely neutral! This conclusion and its possible physical relevance will be further discussed in the final section.

3. Numerical results

To explore the consequences of the present sink strengths for the growing voids we have solved the exact eq. (26) for A by an accurate numerical iteration procedure using the physical parameters, reasonably appropriate to stainless steel [11], given in table 1. In all the calculations we have assumed a small void radius of 1 nm and a fixed void concentration of 10^{21} m^{-3} and varied the dislocation density, temperature and damage rate. Fig. 1 shows the variation of A^2 with dislocation density at a damage rate of 10^{-6} dpa/s for the temperatures 300°C and 500°C. It is clear from (16) that the quantity A^2 is a measure of the mobility of the surface of a growing void and because its value is much less than that of the vacancy diffusion coefficient in this temperature range ($D_v = 2.9 \times 10^{-17} \text{ m}^2/\text{s}$ at 300°C

Table 1

Physical and irradiation parameters for 316 stainless steel used to obtain the results in figs. 1-9

Interstitial diffusion coefficient, $D_i = D_i^0 \exp(-E_i^m/k_B T)$

$$D_i^0 = 10^{-7} \text{ m}^2/\text{s}$$

$$E_i^m = 0.2 \text{ eV}$$

Vacancy diffusion coefficient, $D_v = D_v^0 \exp(-E_v^m/k_B T)$

$$D_v^0 = 6 \times 10^{-5} \text{ m}^2/\text{s}$$

$$E_v^m = 1.4 \text{ eV}$$

Average cavity radius $r_C = 1 \text{ nm}$

Cavity number density $C_C = 10^{21} \text{ m}^{-3}$

Range of dislocation densities $\rho_D = 10^{13} - 10^{16} \text{ m}^{-2}$

Range of temperatures $T = 300 - 500^\circ\text{C}$

Range of damage rates $K = 10^{-3} - 10^{-6} \text{ dpa/s}$

Dislocation bias parameters $Z_i = 1.10, Z_v = 1.00$

and $D_v = 4.5 \times 10^{-14} \text{ m}^2/\text{s}$ at 500°C) the motion of the void surface can have only a small effect at this low damage rate. This almost quasi-static behaviour results in the near temperature independence of the curves and the position of the maximum in A^2 at about $\rho_D = 10^{13} \text{ m}^{-2}$, when the sink strengths of the voids ($4\pi r_C C_C \approx 1.2 \times 10^{13} \text{ m}^{-2}$) and dislocations are equal [4]. Fig. 2 shows the same results for the higher damage rate of 10^{-3} dpa/s ; it is immediately noticeable that the magnitude of A^2 is now much larger and that as it ap-

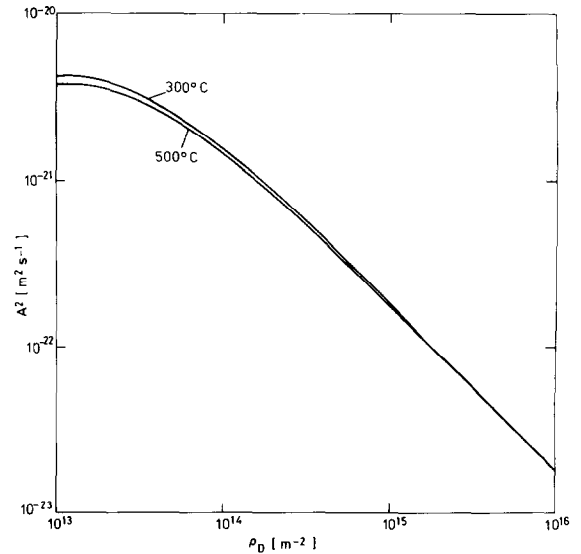


Fig. 1. The variation of A^2 , given by solving (26), with dislocation density ρ_D for the two temperatures specified. The damage rate $K = 10^{-6} \text{ dpa/s}$ and the physical parameters in table 1 have been used for all the figures.

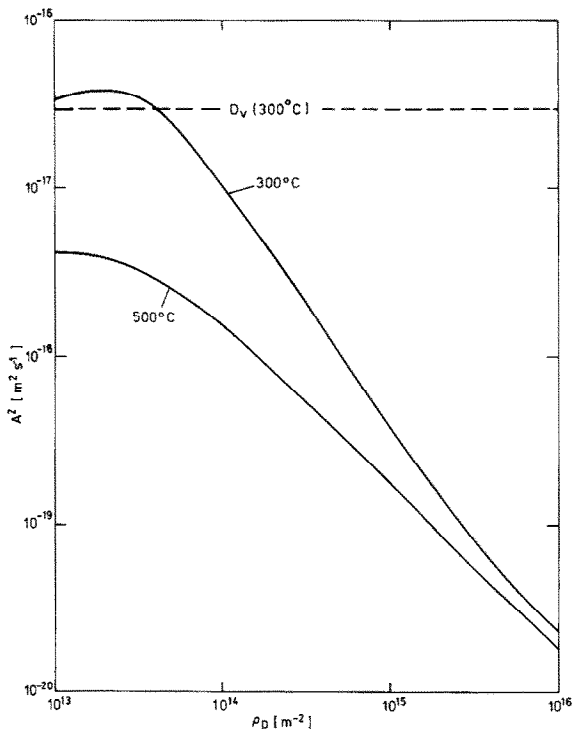


fig. 2. As for fig. 1 but for a damage rate $K = 10^{-3}$ dpa/s; the value of the vacancy diffusion coefficient D_v at 300°C is indicated.

proaches the value of D_v the curves both change shape and exhibit a marked temperature dependence. To further clarify this behaviour figs. 3 and 4 show the variations of the ratio A^2/A_b^2 for the low (10^{-6} dpa/s) and high (10^{-3} dpa/s) damage rates respectively. This ratio is a measure of the “self enhancement” of the void surface mobility and we see the enhancement is considerable at the higher damage rate in fig. 4 with a strong temperature dependence especially at low dislocation densities.

The consequent variations of the growth correction factors ξ_α for the growing void sink strengths, as given by (23) and (24), are depicted in figs. 5, 6 and 7 by presenting the percentage increase of the void sink strength due to growth, $(\xi_\alpha - 1) \times 100$, again for the two temperature 300°C and 500°C . We see, from fig. 5, that for typical reactor conditions ($K = 10^{-6}$ dpa/s) the increases for the vacancies are always small, typically small fractions of a per cent. On the other hand, from fig. 6, it is clear that for accelerator conditions ($K = 10^{-3}$ dpa/s) increases of a few per cent or greater can occur. For comparison with these sink strength increases for vacancies we show the corresponding sink strength increase, $(\xi_i - 1) \times 100$, for the interstitials in fig. 7 for the high damage rate (10^{-3} dpa/s); even for this high damage rate the interstitial mobility is sufficiently high to ensure that the increase is always quite negligible compared with that for the vacancies. We conclude that void surface motion does not significantly affect the interstitial flux into the voids.

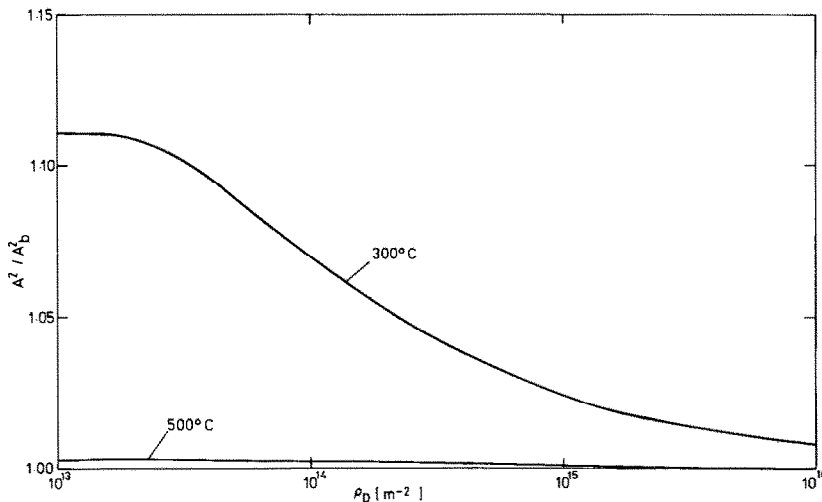


Fig. 3. The variation of the ratio $(A/A_b)^2$, where A_b is given by (31), with dislocation density ρ_D for the two temperatures specified. The damage rate $K = 10^{-6}$ dpa/s.

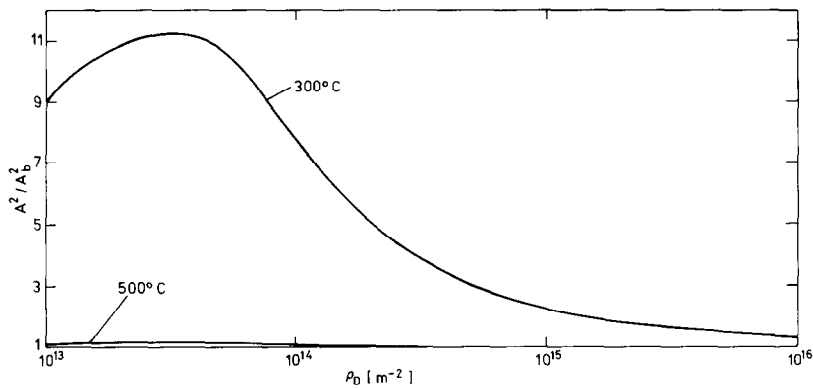


Fig. 4. As for fig. 3 but for a damage rate $K = 10^{-3}$ dpa/s.

Finally, in figs. 8 and 9 we present the variation of the percentage fraction of the total swelling rate, given by (34), due to the void surface motion, $(1 - (A_b/A)^3) \times 100$, for the respective low and high damage rates used previously. Again results for the two temperatures 300°C and 500°C are given. At the lower, reactor, damage rate in fig. 8 the swelling enhancement due to the void surface motion is restricted to a few per cent whereas at the higher, accelerator, damage rate in fig. 9 very large swelling enhancements obtain, particularly at

the lower temperature. The practical relevance and validity of these swelling results will be discussed in the next section; it will suffice here to emphasize that these swelling curves are presented only for their interest as accurate evaluations of the simple theoretical expressions, such as (26), in the present paper and are not intended to have validity to any physical system over the complete range of ρ_D and T used here.

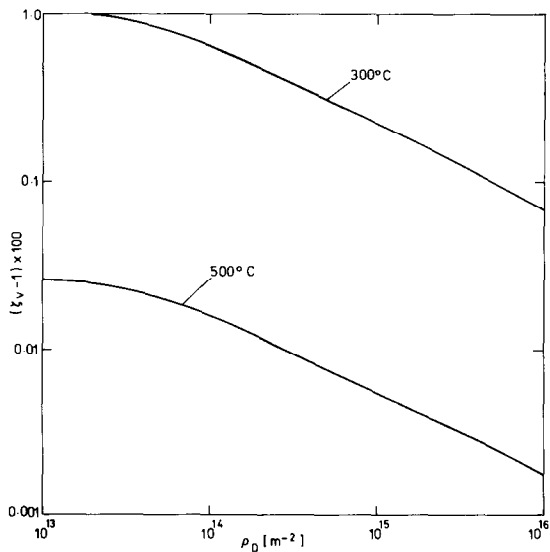


Fig. 5. The variation with ρ_D of the percentage increase of the void sink strength for vacancies, $(\xi_v - 1) \times 100$, due to the motion of the void surface for the two temperatures specified. The damage rate $K = 10^{-6}$ dpa/s.

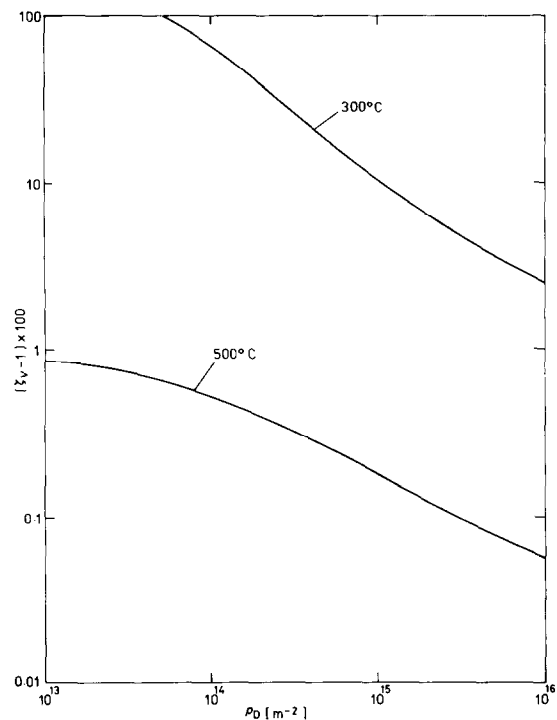


Fig. 6. As or fig. 5 but for a damage rate $K = 10^{-3}$ dpa/s.

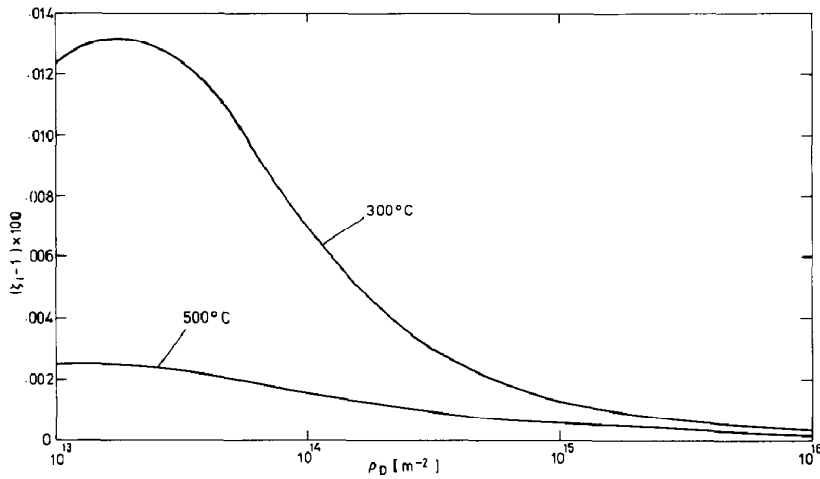


Fig. 7. The variation with ρ_D of the percentage increase of the void sink strength for interstitials, $(\xi_1 - 1) \times 100$, due to the motion of the void surface for the two temperatures specified. The damage rate $K = 10^{-3}$ dpa/s.

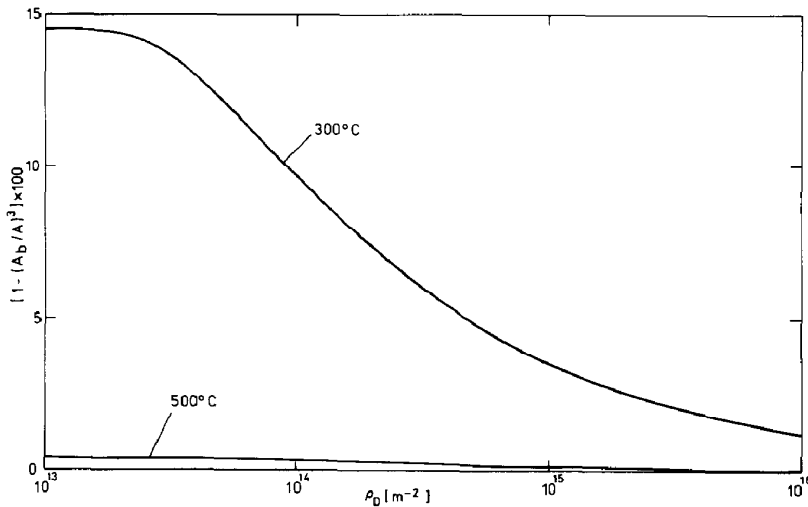


Fig. 8. The variation with ρ_D of the percentage fraction of total swelling due to the motion of the void surface, $[1 - (A_b/A)^3] \times 100$, for the two temperatures specified. The damage rate $K = 10^{-6}$ dpa/s.

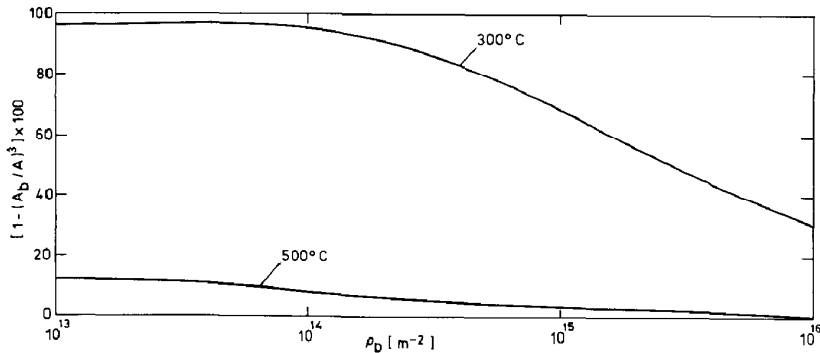


Fig. 9. As for fig. 8 but for a damage rate $K = 10^{-3}$ dpa/s.

4. Discussion

In this paper we have used an embedding procedure to obtain a simple expression for the sink strength of a growing void. The essential feature of the study is probably the realization that such a self-consistent solution for the growing spherical sink exists when its growth is determined by the fluxes of more than one point-defect type. In this sense it represents a generalization of the original analysis due to Frank [9] of the growing spherical precipitate when only one point-defect type was involved in the precipitation process. Of course, the present calculation could be further generalized to involve growth arising from the collective segregation of more than two point-defect types. It is, however, important to identify the inadequacies of the present analysis in relation to void growth processes in real irradiated materials. We have stated in the text that the embedding model used is only valid when sink-sink interactive effects are negligible, that is when the inequality (14) is satisfied, and thus any results we obtain can only be valid as corrections to the "dilute" quasi-static void sink strength (13). It is to satisfy (14) that we deliberately present results for small voids of radius 1 nm; for this radius the condition (14) is satisfied for the complete range of ρ_D employed. For larger voids the upper limit of ρ_D would have to be lowered to comply with (14). However this tendency for the model to be inaccurate at high ρ_D is somewhat alleviated by the fact that the growth effects we are examining are in any case minimal when ρ_D is large.

In addition to the restriction (14) on the validity of the results it is important to emphasize that point-defect loss due to bulk recombination has been neglected both in the derivation of the growing void sink strength in section 2 and in the subsequent evaluation of the swelling increase due to the void growth in figs. 8 and 9. The error in the sink strength calculation is difficult to estimate but it arises because its value depends on the actual *net* flux of vacancies arriving at the void through eq. (8); thus, in contrast to the usual quasi-static embedding calculations of sink strengths, the simultaneous presence of both point-defect types is implicitly understood and recombination loss should be included. The only mitigation we can claim stems from the fact that when such non-linear recombination loss has been included in quasi-static sink strength calculations its effect is small [11]. In addition the present sink strengths are probably satisfactory if their use is restricted to swelling situations when the point-defect loss occurs

predominantly at the sinks rather than by bulk recombination. Thus using a recombination parameter given by [12] $\alpha = 10^{20} D_i s^{-1}$ we can estimate the value of ρ_D , for each temperature and damage rate in figs. 8 and 9, above which the curves are valid. Since no recombination has been included in the evaluations of these curves they can only be valid when $2K > \alpha c_i^\infty c_v^\infty$ where c_i^∞ and c_v^∞ are given in terms of A and the other parameters by (25). This comparison of total point-defect loss to the sinks with potential recombination loss yields the following conclusions: For the damage rate of 10^{-6} dpa/s, ρ_D must exceed $5 \times 10^{14} m^{-2}$ at $300^\circ C$ and $10^{13} m^{-2}$ at $500^\circ C$; at the higher damage rate of 10^{-3} dpa/s, ρ_D must exceed $10^{16} m^{-2}$ at $300^\circ C$ and $5 \times 10^{14} m^{-2}$ at $500^\circ C$. It follows that the results in figs. 8 and 9 have limited validity and the percentage increase of swelling due to the motion of the void surface is probably always less than 5%.

Although we have presented results showing the temperature sensitivity of the void growth on the swelling our basic rate theory model for the swelling used here is extremely primitive. Thus not only have we neglected bulk recombination but we have also omitted any temperature variations of the sink densities themselves. The curves in figs. 8 and 9 are thus not, in any sense, intended to represent predictions for stainless steel but are presented merely to provide insight into the kind of modifications the motion of the growing void surface could have on the total swelling of irradiated materials. It is interesting to speculate on the effect of including both the void surface motion as discussed here and the climb motion of the dislocations as discussed by Rauh and Bullough [9] in the same swelling analysis. The motion of void surface increases the growth rate of the void because the void surface captures more relatively sluggish vacancies, on the other hand the climb motion of the dislocation reduces the net interstitial flux to the dislocations and hence retards the void growth rate. Presumably if the two effects are simultaneously present the overall effect of such sink motions could be minimal and strong support for the accuracy of the quasi-static approach might be forthcoming.

Finally we have noted that the void surface motion provides a net vacancy bias for the growing void and thus we see that, even if the second sink is neutral, once void growth has commenced it should be self-driven by its own vacancy bias arising solely from the differences in point-defect mobility. Such an effect could well give significant assistance to the growth of the vacancy clusters during the initial nucleation and growth stage.

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