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Classical nucleation theory of microstructure development under cascade-damage irradiation

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Abstract

Cascade irradiation produces a significant fraction of the damage in the form of small mobile and immobile vacancy and interstitial clusters. This has led to the introduction of the Woo–Singh production bias theory. In the pursuant studies, the predominant effort that has been spent is in investigating the validity of the concept, and in its usefulness in complementing the traditional theory based on the concept of sink bias. Although plenty of theoretical and experimental results supports the concept, relatively little attention has been paid to the important area of microstructure nucleation. Within the framework of the classical theory of nucleation of overcritical precipitates from small subcritical nuclei, the nucleation processes at elevated temperatures of both voids and interstitial loops from the primary clusters are similar, and can be similarly treated. Recognizing the importance of stochastic fluctuations in the evolution of small embryos, a single-component nucleation theory is formulated using the Fokker–Planck equation, to take into account the stochastic effects of the fluxes of mobile defects, arising from the random nature of diffusion jumps and cascade initiation. Analytic solutions for the separate cases of voids and Frank loops are obtained, and the corresponding effects on the evolution of the microstructure are discussed.

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1. Introduction

Under cascade-damage irradiation, point defects are produced in the form of small mobile or immobile vacancy and interstitial clusters [1–5]. Recognition of this fact has led to the introduction of the Woo–Singh production bias [6,7] theory. Under such conditions, point defects are produced in discrete packages randomly in time and space, and both the randomness of diffusion jumps and the random initiation of cascades introduce stochastic fluctuations in the point-defect flux that arrives at any sink [8,9]. It is intuitively clear that such fluctuations are particularly important to processes involving only relatively small numbers of point defects, such as the evolution of small point-defect clusters during the nucleation of microstructure components. Indeed, to consider the growth or shrinkage of small interstitial clusters with sizes of about 5–10 atoms, the effect of fluctuations in point-defect fluxes is very important, because a cluster that has been annihilated cannot be revived, even when the time-averaged point-defect flux dictates that it must always grow. Similarly, a small void embryo, after having been annihilated by a wave of interstitials, cannot be regenerated even if a larger wave of vacancies subsequently arrives. Consideration of kinetics beyond the mean-field approximation is clearly necessary to allow an accurate physical understanding of the evolution of damage microstructure under cascade irradiation, particularly the nucleation of microstructure components, such as voids and dislocations.

The conventional approach to modeling void nucleation under irradiation is based on the classical description of the formation of small precipitates in a supersaturated solution. In this approach, small thermally unstable new-phase embryos continuously form

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and re-dissolve in the supersaturated solution, and can grow beyond the critical size via stochastic fluctuations [10–16]. Beyond the critical size, nuclei of the new phase become thermally stable, and on the average can grow directly from the supersaturated solution, without the help of the stochastic fluctuations. In most studies of void nucleation only statistical fluctuations produced by random point-defect jumps are taken into account, and the dislocation bias is the only driving force for the evolution of the damage microstructure [13,14]. In the present paper the additional effects of the cascade-induced fluctuations, and the fluctuations in the rate of vacancy emission from the void, are also taken into account.

Another important component in the microstructure development during irradiation is the generation of dislocations from loops. In the temperature range just above the annealing stage V (i.e. the peak swelling regime) the dissociation of primary vacancy clusters that are thermally less stable than primary interstitial clusters (PICs) produces a much higher flux of freely migrating vacancies to all sinks, than that of the mobile interstitials. At the same time, from the continuous generation of small clusters in cascades (particularly thermally stable and immobile PICs), it is clear that, if they are only produced and not removed/annihilated, they will become the dominant sinks at a even very low dose. Acting as recombination centres, they will suppress further development of the microstructure. Within the mean-field theory, it is difficult to rationalize the concurrence of the annihilation/removal of PICs on the one hand, and the nucleation and growth of interstitial dislocations loops, on the other, under the continuous assault of a net vacancy flux that drives the growth of voids.

One of the possible ways faulted loops may grow, despite the net vacancy flux they receive, is through the absorption of smaller interstitial clusters and loops by coalescence during climb. Numerical calculations [17] shows that the absorption of small interstitial clusters and loops, by climbing dislocations in their vicinity, can provide both the positive growth rate of larger loops, and the sufficiently high climb rate of network dislocations, to produce a swelling rate in agreement with experimentally observed values. However, the probability of finding another cluster in the immediate neighbourhood, and subsequently combining with it, decreases as the loop size decreases, and vanishes for the smallest immobile interstitial clusters. Thus, this mechanism can only account for the growth of sufficiently large interstitial loops.

From the foregoing, the resemblance between the nucleation of growing voids and Frank loops at elevated temperatures is clear. Thus, both vacancy and interstitial clusters are directly produced in collision cascades, and there are also corresponding critical sizes in both cases. Subcritical void and loop embryos are shrinking on the average during the nucleation process, and nucleation, i.e. the successful growth of a very small fraction of the embryos to supercritical size, can only be accomplished via stochastic fluctuations.

Thus, within the framework of the classical theory of nucleation of overcritical precipitates from small subcritical nuclei, the nucleation processes of both voids and interstitial loops from the primary clusters are similar, and can be similarly treated using a unified kinetic model. As we will show in this paper, an expression of general applicability can be derived to calculate the nucleation probability in both cases. Through the formulation of such a model, the present paper aims at understanding the details of the physics involved in the nucleation of voids and Frank loops at elevated temperatures when the production bias is operational.

We note that the present model only considers the nucleation of a single microstructure component, instead of the more general case of simultaneous nucleation of an interactive multi-component system, which is far more complicated, and is beyond the scope of the present work. Thus, here we only deal with the case in which the nucleation of voids and dislocation loops occurs separately in time. The simplification allows the physics of nucleation of voids and Frank loops to be followed through an analytic approach. Thus, an expression of general applicability can be derived for the nucleation probability of microstructure components growing due to stochastic fluctuations in point-defect fluxes. Cu and Mo, representing two opposite extremes with regard to the effect of surface tension on vacancy emission from voids, are chosen for investigation. Stainless steel, a concentrated alloy in which the interstitial cluster mobility is expected to be very much restricted, is a good material to consider with regard to the Frank loop nucleation. It will be shown that cascade-induced fluctuations indeed produce sufficiently high nucleation rate of growing loops to explain the experimentally observable concentrations of interstitial loops and the continuous regeneration of network dislocations.

2. Nucleation probability

The general kinetic equation for the microstructure evolution under cascade-damage irradiation, including the full statistical effects, has been derived in [9]. It is applicable to both voids and interstitial loops, by considering them as precipitates of vacancies and interstitials, respectively. Adopting the simplest approximation that takes into account the effect of fluctuations, the kinetic equation takes the form of the Fokker–Planck equation [9]

$$\frac{\partial P(n,t|n_0,t_0)}{\partial t} = -\frac{\partial}{\partial n}\left\{V(n) - \frac{\partial}{\partial n}D(n)\right\}P, \tag{1}$$

where $P(n,t|n_0,t_0)$ is the probability density that a precipitate with an initial size of n_0 point defects (embryo) at time t_0 will have a size of n point defects at a later time t, $V(n)$ is the drift velocity determined by the average rates of absorption of vacancies and interstitials by the precipitate. By definition of P, the initial condition of (1) may be written as

$$P(n,0|n_0,t_0) = \delta(n-n_0). \tag{2}$$

The diffusivity $D(n)$ in (1) governs the 'diffusive spread' of the probability P due to stochastic fluctuations in the point-defect fluxes received by the precipitate. Physically, the stochastic growth of the microstructure components is the manifestation of the 'diffusive spread'.

$D(n)$ depends on the average point-defect fluxes, the cascade properties, and can be expressed in the following form [8,9,18]:

$$D(n) = D^{s}(n) + D^{c}(n) + D^{e}(n), \tag{3}$$

where the superscripts s, c and e refer to contributions due to the random migratory jumps, random cascade initiation and random emission of point defects, respectively.

Since small clusters consisting of two or three point defects are mobile [19,20], there is a minimum size $n_{\min}$ below which the cluster cannot be considered a precipitate anymore. This allows us to write down the left boundary condition for the kinetic equation (1) in the form

$$P(n = n_{\min}, t|n_0,t_0) = 0. \tag{4}$$

Neglecting the probability for a sufficiently large supercritical precipitate of the size n_{m} to shrink to a subcritical size, the right boundary condition can be written as [10,12,15]

$$P(n = n_{\mathrm{m}} \gg n_{\mathrm{cr}}, t|n_0,t_0) = 0. \tag{5}$$

The existence of a critical size n_{cr} in this sense, in the case of interstitial loops, is not as obvious as in the case of voids. This issue will be addressed in the following section. At this juncture, it is sufficient to note that, even in cases where the average loop growth rate is negative for all loop sizes, this boundary condition is still valid, because sufficiently large loops will unfault, join the network, and, consequently, disappear.

With this boundary condition, the probability $P_{\mathrm{m}}(t)$ for a subcritical precipitate to become supercritical during the time period (t_0,t) is given by

$$P_{\mathrm{m}}(t) = -\left[\frac{\partial}{\partial n}\int_{t_0}^{t} D(n)P(n,t|n_0,t_0)\,\mathrm{d}t\right]_{n=n_{\mathrm{m}}}. \tag{6}$$

From the Fokker–Planck equation (1), the initial condition (2) and the boundary conditions (4) and (5), the following conservation law can be derived:

$$P_0(t\to\infty) + P_{\mathrm{m}}(t\to\infty) = 1, \tag{7}$$

where $P_0(t)$ is the probability for an embryo to shrink below the minimum size $n_{\min}$ and disappear, i.e.

$$P_0(t) = \left[\frac{\partial}{\partial n}\int_{t_0}^{t} D(n)P(n,t|n_0,t_0)\,\mathrm{d}t\right]_{n=n_{\min}}. \tag{8}$$

Eq. (7) has the obvious physical meaning that an embryo, once generated, may either shrink away or grow to become supercritical.

Finally, the probability $P_{\mathrm{m}} \equiv P_{\mathrm{m}}(t\to\infty)$ for a small embryo to attain supercriticality is given by [21]

$$P_{\mathrm{m}} = \int_{n_{\min}}^{n_0} \exp\left\{-\int_{n_{\min}}^{n}\frac{V(n')}{D(n')}\mathrm{d}n'\right\}\mathrm{d}n \Big/ \int_{n_{\min}}^{n_{\mathrm{m}}} \exp\left\{-\int_{n_{\min}}^{n}\frac{V(n')}{D(n')}\mathrm{d}n'\right\}\mathrm{d}n. \tag{9}$$

Expression (9) will be used in the following for the consideration of nucleation of both voids and growing interstitial loops.

3. Void nucleation

Within the mean-field theory and a spherical approximation for the voids, the drift term in Eq. (1), which governs the average number of vacancies in the void, has the following conventional form:

$$V(n) = \frac{3n^{1/3}}{a^2}(D_v C_v - D_i C_i - D_v C_{\mathrm{s}}^{\mathrm{e}}(n)), \tag{10}$$

where D_j and C_j $(j = i, v)$ are the diffusion coefficient and the concentration of point defects, respectively, $a = (3\Omega/4\pi)^{1/3}$, and Ω is the atomic volume. The mean equilibrium concentration $C_{\mathrm{s}}^{\mathrm{e}}(n)$ of vacancies in the neighbourhood of a void of radius $R_{\mathrm{c}}(n) = an^{1/3}$ can be written as

$$C_{\mathrm{s}}^{\mathrm{e}}(n) = C_\infty \exp\left(\frac{2\gamma_{\mathrm{s}}\Omega}{kTR_{\mathrm{c}}}\right) \approx C_\infty\left(1 + \frac{2\gamma_{\mathrm{s}}\Omega}{kTR_{\mathrm{c}}}\right). \tag{11}$$

Here C_∞ is the equilibrium vacancy concentration, γ_{s} is the surface tension coefficient, k is the Boltzmann constant, and T is the absolute temperature. We note that the approximation in (11) is only valid for sufficiently large void sizes.

In the case of voids the corresponding expressions for the diffusivities are given by [8,9,18]

$$D^{s}(n) = \frac{3n^{1/3}}{2a^2}(D_v(C_v - C_{\mathrm{s}}^{\mathrm{e}}(n)) + D_i C_i), \tag{12}$$

$$D^{c}(n) = \frac{3n^{2/3}}{4a}\left[\frac{G_v\langle N_{dv}^2\rangle}{k_v N_{dv}} + \frac{G_i\langle N_{di}^2\rangle}{k_i N_{di}}\right], \tag{13}$$

$$D^{e}(n) = \frac{9D_v C_s^{e}(n) n^{2/3}}{2a^2}. \tag{14}$$

Here G_j is the effective generation rate of free point defects, N_{dj} and $\langle N_{dj}^2\rangle$ are the average number and the average square number of free point defects generated in a single cascade, respectively, and k_j^2 is the total sink strength for point defects of the type j.

For temperatures at which the vacancy emission from voids is not negligible, the critical void size n_{cr}, at which the drift velocity $V(n_{cr})$ vanishes, is much larger than n_{min} According to [21], expression (9) for the probability of void nucleation can be approximated by

$$P_m \cong \sqrt{\frac{\beta}{2\pi a^3 n_{cr}}\frac{(D_v C_v - D_i C_i)}{D(n_{cr})}} \times \exp\left[\int_{n_{min}}^{n_{cr}} V(n)/D(n)\,dn\right](n_0 - n_{min}), \tag{15}$$

where $\beta = 2\gamma_s\Omega/kT$. It is important to note that D^{e} in (14) is proportional to the surface area of the void [18,22], as it is natural to expect in the case of emission. In contrast, the contribution of the vacancy emission to the diffusion term in the kinetic equation (1) $D(n)$ is treated conventionally like the emission contribution to the average void growth rate, and is assumed to be proportional to the void radius instead [12–14]. This issue has been discussed in detail in [22], and the corresponding expression for D^{e} had been derived in [18].

The difference between the present and conventional treatments of vacancy emission from the voids is clear when we neglect the cascade-induced fluctuations. In this case, the corresponding total diffusion coefficient becomes

$$D^{s}(n) + D_{conv}^{e}(n) = \frac{3n^{1/3}}{2a^2}(D_v(C_v + C_s^{e}(n)) + D_i C_i) \tag{16}$$

and the term proportional to $n^{2/3}$ is absent. Referring to (15) and recalling that $V(n)$ is negative for $n < n_{cr}$, it is clear that the fluctuations in the vacancy emission rate would produce a void nucleation probability higher than conventionally, as long as

$$\frac{D^{e}(n_{cr})}{(3n_{cr}^{1/3}/2a^2)(D_v(C_v + C_s^{e}(n_{cr})) + D_i C_i)} \geqslant 1. \tag{17}$$

By definition of the critical size, from Eqs. (10) and (14), the left-hand side of inequality (17) is equal to $3(n_{cr})^{1/3}(D_v C_v - D_i C_i)/2D_v C_v$. The ratio $(D_v C_v - D_i C_i)/D_v C_v$ is related to the mean net vacancy flux to the void embryos. The operation of production bias at elevated temperatures predicts that this ratio is, to a good approximation, given by the fraction of interstitials produced in cascades in the form of immobile clusters, ε_i [6]. The last inequality is then satisfied when $n_{cr}^{1/3} \geqslant 2/(3\varepsilon_i)$. Thus, for a critical void size larger than ~20 (i.e. with $\varepsilon_i \cong 0.25$–0.4), the conventional vacancy emission treatment under-predicts the survival probability of the void nucleus. We note that this range of values of ε_i is consistent with MD results (see e.g. [5]).

A clear illustration of the foregoing conclusion is offered in the case of large critical void size. Using approximation (11) for $C_s^{e}(n)$, the diffusion coefficient in the void size space are described by the probability P_m [21]:

$$P_m \cong \sqrt{\frac{\beta}{6\pi R_{cr} n_{cr}}\frac{(D_v C_v - D_i C_i)}{D_i C_i(1 + dn_{cr}^{1/3})}}(n_0 - n_{min}) \times \exp\left\{-3\frac{v}{d}\left[\frac{(n_{cr}^{2/3} + n_0^{2/3})}{2} - n_{cr}^{1/3} n_0^{1/3} + \frac{n_{cr}^{1/3} - n_0^{1/3}}{d} - \frac{1 + dn_{cr}^{1/3}}{d^2}\ln\left(\frac{1 + dn_{cr}^{1/3}}{1 + dn_0^{1/3}}\right)\right]\right\}, \tag{18}$$

where

$$v = \frac{(D_v C_v - D_i C_i)(1 - \exp(-\beta/R_{cr}))}{D_i C_i} \tag{19}$$

and

$$d = d^{c} + d^{e} \tag{20}$$

with

$$d^{c} = \frac{aG_i}{4D_i C_i}\left[\frac{G_v\langle N_{dv}^2\rangle}{G_i N_{dv} k_v} + \frac{\langle N_{di}^2\rangle}{N_{di} k_i}\right], \tag{21}$$

$$d^{e} = \frac{3(D_v C_v - D_i C_i)}{2D_i C_i}. \tag{22}$$

In the conventional treatment, the corresponding P_m is also given by (18), but taking the limit $d \to 0$, and

$$v = \frac{(D_v C_v - D_i C_i)(1 - \exp(-\beta/R_{cr}))}{D_v C_v}. \tag{23}$$

Comparing the curves for $d = 0$ with those for $d \neq 0$ in Fig. 1, it can be seen that the conventional treatment of vacancy emission results in a contribution to the diffusion coefficient $D(n)$ that underestimates the survival probability of the void embryo by several orders of magnitude. We note that the value of $\varepsilon_i = 0.4$ or $D_i C_i/D_v C_v \cong (1 - \varepsilon_i) \cong 0.6$ is used in Fig. 1, to unambiguously demonstrate the difference between the two approaches, recognizing that the results depend exponentially on the parameters. Note also that for a relatively low total sink strength ($k_j^2 < 10^{15}$ m^{-2}), d^{c} can be neglected ($d^{c} < 1.5 \times 10^{-1}$, $N_{dj} \cong 50$), and in this case

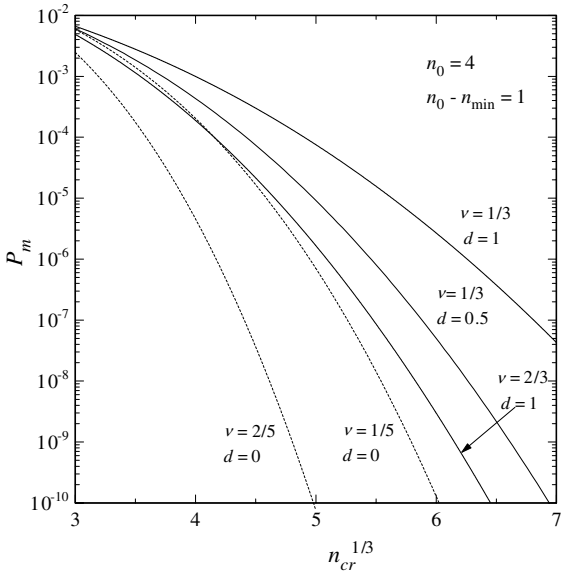

Fig. 1. Probability for a small void nucleus to become supercritical, plotted as a function of critical void size at different values of the parameters ν and d, calculated using Eq. (18). Dashed lines correspond to the probability in the limit $d \to 0$.

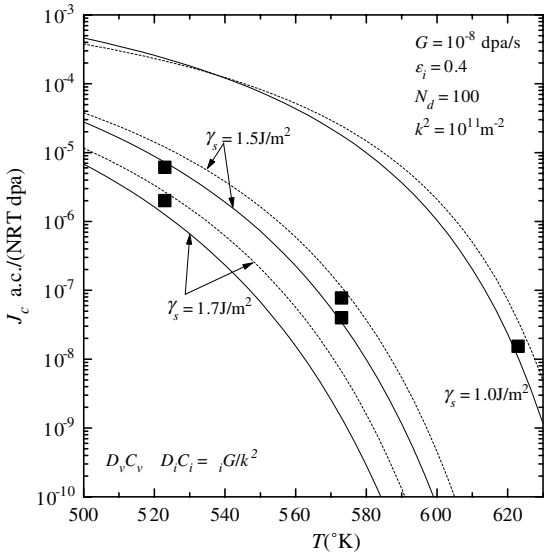

Fig. 2. Rates of void nucleation in annealed copper at different values of void surface energies. Corresponding nucleation probability P_m is calculated using expression (15). The parameter d^e is equal to 1.0 (—) and 0.5 (- - -). Experimental points are obtained from Refs. [23–25] by dividing the experimental values of void concentration by the corresponding irradiation doses.

the value of d is dominated by the vacancy emission term d^e.

We can also compare the present results calculated with Eq. (15), without assuming a large critical void radius [21], with experimental void nucleation rate for annealed pure copper with low dislocation density ($\sim 10^{11}$ m^{-2}), and neutron-irradiated up to doses of 10^{-4}–10^{-2} NRT dpa [22–24].

To make this comparison we need to know the rate of formation of small void embryos (subcritical) in the form of vacancy clusters with n_0 vacancies. A small immobile three-dimensional vacancy cluster can be formed directly in a collision cascade [26] or through the agglomeration of several single vacancies present in a solid solution. Thus, if the formation of small void nuclei through the consecutive agglomeration of single vacancies can be neglected, the rate J_c of void nucleation under cascade-damage irradiation may be estimated by multiplying the probability P_m with the average rate of cascade production:

$$J_c \cong \frac{G}{N_d} P_m, \tag{24}$$

where G is the effective generation rate of point defects in cluster and free form, and N_d is the average total number of point defects generated in a single cascade.

The void nucleation rate calculated with (24) is shown in Fig. 2. Using a surface tension coefficient of $\gamma_s = 1.7$ J/m^2 for pure copper, the agreement between calculated and experimental values is good at 523 K, but deteriorates at higher temperatures. Since the experimental void size distribution is bimodal at higher temperatures, with the position of the first peak weakly dependent on irradiation dose [25], the presence of solute elements that stabilize the voids against shrinkage and collapse into loops or stacking-fault tetrahedra is likely. According to [25], the presence of dissolved oxygen in the copper sample under investigation may reduce the surface tension coefficient γ_s to below 1.0 J/m^2. With the reduced surface energy, the void nucleation probability is much increased, and a good agreement between theoretical and experimental results can be reestablished (Fig. 2).

In the opposite extreme to the case of Cu considered the foregoing, we consider void nucleation in the case when vacancy emission is negligible. A typical case can be found in molybdenum at temperatures below $0.35T_m \cong 1015$ K [21]. If the void growth rate does not depend on vacancy emission from voids, we have shown that [21], (9) reduces to

$$P_m(n_0) = \int_{n_{min}}^{n_0} f(n)\,dn \Big/ \int_{n_{min}}^{\infty} f(n)\,dn, \tag{25}$$

where

$$f(n) = \exp\left\{ -\frac{3\alpha_s^2}{2\alpha_c^3}\left(\frac{\alpha_c}{\alpha_s} n^{1/3} - 1\right)^2 - \frac{3\alpha_s^2}{\alpha_c^3} \ln\left(\frac{\alpha_c}{\alpha_s} n^{1/3} + 1\right)\right\}, \tag{26}$$

$$\alpha_s = \frac{D_v C_v + D_i C_i}{2(D_v C_v - D_i C_i)}, \tag{27}$$

$$\alpha_c = \frac{a}{4(D_v C_v - D_i C_i)} \left[\frac{G_v \langle N_{dv}^2 \rangle}{k_v N_{dv}} + \frac{G_i \langle N_{di}^2 \rangle}{k_i N_{di}} \right]. \tag{28}$$

Here α_s represents effects coming from the random point-defect jumps, and α_c from the random cascade initiation.

In the limit $\alpha_c \to 0$, $P_m(n_0)$ in (25) can be approximated by

$$P_m(n_0) = 1 - \exp(-(n_0 - n_{min})/\alpha_s). \tag{29}$$

In the other limit $\alpha_s \to 0$,

$$P_m(n_0) = 1 - \frac{(3/2\alpha_c)^{1/2} n_0^{1/3} \exp(-3n_0^{2/3}/2\alpha_c) + \sqrt{\pi} Q(\sqrt{3/\alpha_c} n_0^{1/3})}{(3/2\alpha_c)^{1/2} n_{min}^{1/3} \exp(-3n_{min}^{2/3}/2\alpha_c) + \sqrt{\pi} Q(\sqrt{3/\alpha_c} n_{min}^{1/3})}, \tag{30}$$

where

$$Q(x) = \frac{1}{\sqrt{2\pi}} \int_x^\infty \exp(-x^2/2)\, dx. \tag{31}$$

According to Eqs. (25), (29) and (30), even when the void embryos are growing on the average independent of their sizes, not all them can survive to finally become supercritical.

Since voids are usually the dominant sink for point defects in Mo [27–30] the parameter α_s can be approximately written as

$$\alpha_s = \frac{k_c^2 (D_v C_v + D_i C_i)}{2\, dS/dt} \cong \left(\frac{dS}{dGt} \right)^{-1}, \tag{32}$$

where k_c^2 is the void sink strength. At temperatures under consideration, the void swelling rate $dS/d(Kt)$ varies between 1×10^{-4} and 3×10^{-4} per NRT dpa ($G/K \cong 0.3$) [28], and α_s takes on values between 1×10^3 and 3×10^3. Values of $P_m(n_0)$ from (25), where $n_0 = n_{min} + 1$ is the minimum number of vacancies in the void embryo, are plotted in Fig. 3. The dashed lines in the figure are the void nucleation probabilities in the two limiting cases, according to (29) and (30). These equations give the same nucleation probability when [21]

$$\frac{\alpha_c}{\alpha_s} = \frac{3}{(2\pi\alpha_s)^{1/3}}. \tag{33}$$

When the value of α_s falls in the range between 1×10^3 and 3×10^3, the above ratio is about 0.1. Since

$$\frac{\alpha_c}{\alpha_s} \cong \frac{ka}{4} \left[\frac{G_v \langle N_{dv}^2 \rangle}{G N_{dv}} + \frac{G_i \langle N_{di}^2 \rangle}{G N_{di}} \right], \tag{34}$$

this value of α_c/α_s corresponds to a value of the total sink strength $k^2 \approx 3 \times 10^{15}$ m^{-2} ($N_{dj} \cong 50$, $G_j/G \cong 0.5$).

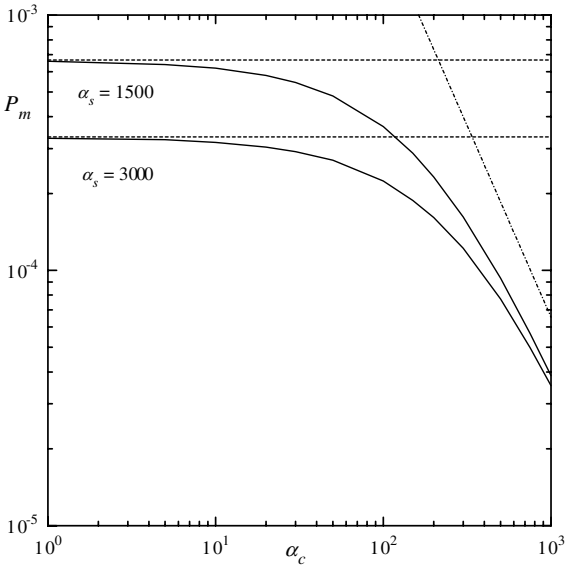

Fig. 3. Probability $P_m(n_{min} + 1)$ of void nucleation in molybdenum as a function of parameter α_c at different values of parameter α_s. Dashed lines correspond to the probabilities given by Eqs. (29) (horizontal lines) and (30).

When the void sink strength is much below this value, the rate of void nucleation is independent of α_c, and is determined by the average void growth rate. With the increase in the total sink strength, the nucleation rate starts to drop very fast as α_c/α_s increases, because cascade-induced fluctuations significantly reduce the survival probability for the void embryos. As can be seen from Fig. 3, for temperatures at which vacancy emission from the voids is negligible, an irradiation dose of several NRT dpa is required to produce a void density of the order of 10^{23} m^{-3}. This is in agreement with the experimental observation [28].

In this connection, the shrinkage of rather large voids with the positive average growth rate is also of particular interest. While this kind of void shrinkage is not expected under the conventional theory of void swelling, it follows from our consideration. The probability $P_0(n)$ for a void of the size n to completely dissolve is given by the conservation law (7) and Eq. (25). It is plotted as a function of void radius in Fig. 4, for different values of parameters α_s and α_c. It can be seen that, even in the absence of vacancy emission, there is a chance of higher than 50% that voids with a diameter as large as 5 nm may shrink away. Thus, the thermal stability, or even a positive growth rate of the ensemble average, cannot guarantee the survival of a void in the course of its evolution. Only those voids for which $P_0(n)$ is substantially less than one will grow with time.

We have shown in the foregoing that cascade-induced fluctuations lead to a large increase (several times) in the probability of void shrinkage. Since the

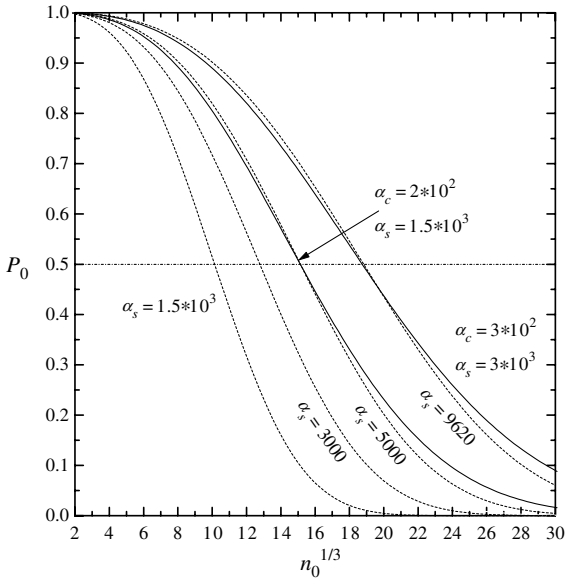

Fig. 4. Probability of void dissolution as a function of void size at different values of parameters α_s and α_c. Probability $P_0 = \exp(-(n_0 n_{\min})/\alpha_s)$ is shown by the dashed lines.

probability of shrinkage for smaller voids is much higher than that for the larger ones, a possible effect associated with the shrinkage, may be identified as stochastic void coarsening. As shown in [31] stochastic void coarsening can also be interpreted as a non-equilibrium phase transition in the void ensemble induced purely by the stochastic fluctuations.

Experimental observation of the void coarsening effect has been reported in neutron-irradiated molybdenum at 450 °C, where the overall void number density drops from 2.8×10^{23} to 1.2×10^{23} m^{-3} during the medium-dose irradiation. This was observed to be due to a general reduction in the number of voids, mostly with diameters $\leqslant 3$ nm ($n_0^{1/3} \leqslant 11.2$) [32]. At the same time, larger voids continue to grow, so that a positive swelling rate $\leqslant 2\times10^{-2}$% per NRT dpa can be maintained. Vacancy emission from voids is not likely to play an important role in the void shrinkage at such irradiation temperatures [21]. On the other hand, voids of these sizes and concentration have a sink strength k_c^2 equal to 5.3×10^{15} m^{-2}, corresponding to a ratio of $\alpha_c/\alpha_s \cong 0.13$. It follows from Fig. 4 that stochastic fluctuations in point-defect fluxes can lead to a reduction in the number of voids with diameter $\cong$3 nm by 4–5 times.

4. Nucleation of interstitial loops

4.1. Loop nucleation probability

Taking into account the absorption of smaller interstitial clusters and loops by coalescence during the climb, the average loop growth rate $V(n)$ can be written as [17]

$$V(n) = V_i^{\mathrm{pd}}(n) + V_i^{\mathrm{cl}}(n), \tag{35}$$

where V_i^{pd} is the drift velocity given by the conventional expression for the growth rate of interstitial loops due to point-defect absorption:

$$V_i^{\mathrm{pd}}(n) = \frac{2\pi r_i(n)}{\Omega}(Z_i D_i C_i - Z_v D_v C_v). \tag{36}$$

Here Z_j $(j = i, v)$ is the reaction constant between the dislocation and point defects, $r_i(n) = (n_i\Omega/\pi b)^{1/2}$ is the loop radius, and b is the Burgers vector.

For elevated temperatures, we may neglect the contribution from the vacancy clusters, and write

$$V_i^{\mathrm{cl}}(n) = \frac{2\pi r_i(n)}{\Omega}\int_{n_{\min}}^{n} n' f_{i\mathrm{l}}(n', t) W(n', n)\,\mathrm{d}n' \tag{37}$$

to represent the rate of change of loop sizes due to the absorption of smaller interstitial loops by coalescence [17]. Here $f_{i\mathrm{l}}(n, t)$ is the distribution function of interstitial loops, which is related to the total loop number density $N_{i\mathrm{l}}(t)$ by

$$N_{i\mathrm{l}}(t) = \int_{n_{\min}}^{n_{\max}} f_{i\mathrm{l}}(n, t)\,\mathrm{d}n, \tag{38}$$

where $n_{\max}$ is the size of loops, at which they unfault and join the network.

The coalescence between the loops of sizes n' and n $(n' < n)$ is described by the reaction constant $W(n', n)$ [17],

$$W(n', n) = \frac{4r_i(n')}{\lambda_{\mathrm{d}}}[D_{\mathrm{l}}(n) + (D_{\mathrm{l}}^2(n) + V_{\mathrm{l}}^2(n)\lambda_{\mathrm{d}}^2/4)^{1/2}], \tag{39}$$

where λ_{d} is the mean free path of the dislocation between two consecutive reactions with clusters, V_{l} is the average loop climb velocity, and $D_{\mathrm{l}}(n)$ is the climb 'diffusion coefficient' due to the fluctuating point-defect fluxes, which is proportional to the loop diffusion coefficient in the size space $D(n)$. Similar to (12) and (13), $D(n)$ has two components [8]

$$D^{\mathrm{s}}(n) = \left(\frac{\pi n}{\Omega b}\right)^{1/2}(Z_v D_v C_v + Z_i D_i C_i), \tag{40}$$

$$D^{\mathrm{c}}(n) = \frac{N_{\mathrm{d}} G n}{4b}\left[Z_v^2\frac{\langle N_{\mathrm{d}v}^2\rangle}{k_v N_{\mathrm{d}}^2} + Z_i^2\frac{\langle N_{\mathrm{d}i}^2\rangle}{k_i N_{\mathrm{d}}^2}\right]. \tag{41}$$

Vacancy emission in $D(n)$ is neglected, because for a small interstitial loop the process involves an associated increase in the loop size, which is energetically unfavorable. Note also that in (9) the reaction distance

between the coalescing loops is assumed to be equal to the radius of smaller loop $r(n')$.

Further, using the mean value theorem, we can write down the probability P_m given by Eq. (9) as follows [33]:

$$P_m = \int_{n_{min}}^{n_0} \exp\left\{ -\int_{n_{min}}^{n} \frac{V_i^{pd}(n')}{D_i(n')} dn' \right\} dn \Big/ \int_{n_{min}}^{n_m} \exp\left\{ -(1-\alpha^*)\int_{n_{min}}^{n} \frac{V_i^{pd}(n')}{D_i(n')} dn' \right\} dn, \tag{42}$$

where

$$\alpha^* = -\int_{n_{min}}^{n^*} \frac{V_i^{cl}(n')}{D(n')} dn' \Big/ \int_{n_{min}}^{n^*} \frac{V_i^{pd}(n')}{D(n')} dn' = -\frac{V_i^{cl}(n_1)}{V_i^{pd}(n_1)} \quad (n_0 < n_1 < n^* < n_m). \tag{43}$$

Since at elevated temperatures there is a net vacancy flux to the interstitial loops, α^* is positive. It can be also shown that $\alpha^* < 1$ [33]. Physically, this inequality means that for subcritical interstitial loops, which can grow beyond the critical size only through stochastic fluctuations, the absorption of clusters due to coalescence only results in some reduction in the average net vacancy flux.

In general, the integrals in (42) can be expressed in terms of special functions. To simplify our analysis, however, we consider only the two limiting cases: $D^s \to 0$, and $D^c \to 0$. In the limit $D^c \to 0$, the probability P_m reduces to

$$P_m = (1-\alpha^*)\frac{e^{-vn_0/d_s} - e^{-vn_{min}/d_s}}{e^{-vn_m(1-\alpha^*)/d_s} - e^{-vn_{min}(1-\alpha^*)/d_s}} e^{\alpha^* vn_{min}/d_s}, \tag{44}$$

where

$$v = V_i^{pd}(n)/n^{1/2}; \quad d_s = D^s(n)/n^{1/2}. \tag{45}$$

At elevated temperatures v is negative, and, consequently, the probability for a small interstitial cluster to grow beyond ctiticality to a size n_m decreases exponentially with increasing n_m.

In the opposite limit, $d_s \to 0$,

$$P_m = \frac{(1-\alpha^*)^2 e^{2\alpha^* vn_{min}^{1/2}/d_c}\left[\left(1+\frac{2vn_0^{1/2}}{d_c}\right)e^{-2vn_0^{1/2}/d_c} - \left(1+\frac{2vn_{min}^{1/2}}{d_c}\right)e^{-2vn_{min}^{1/2}/d_c}\right]}{\left(1+\frac{2(1-\alpha^*)vn_m^{1/2}}{d_c}\right)e^{-2(1-\alpha^*)vn_m^{1/2}/d_c} - \left(1+\frac{2v(1-\alpha^*)n_{min}^{1/2}}{d_c}\right)e^{-2(1-\alpha^*)vn_{min}^{1/2}/d_c}} \tag{46}$$

with

$$d_c = D^c(n)/n. \tag{47}$$

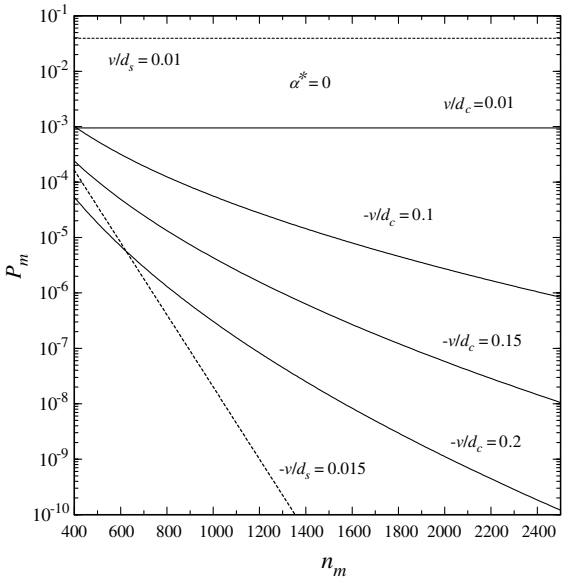

Fig. 5. Probability of nucleation of supercritical loop as a function of the loop size at different values of the ratios v/d_c and v/d_s (- - -).

When v is negative, the exponential factor that governs the probability for a small cluster to eventually become supercritical is proportional to $n_m^{1/2}$, i.e. not to the area of supercritical loop as in the case of $d_c \to 0$, but to its radius. As a result, the presence of cascade-induced fluctuations increases the survival probability of PICs by several orders of magnitude, even in cases where parameter d_c is much less than d_s (see Fig. 5), and is an essential factor in the nucleation of dislocation loops.

4.2. Critical cluster density

At elevated temperatures, under the operation of production bias, there is a net vacancy flux to the interstitial loops and clusters ($V_i^{pd} < 0$). In this case, only interstitial loops with sizes larger than critical, can have a positive average growth rate. From (35) the loop critical size n_{cr} is determined by the equation

$$V_i^{pd}(n_{cr}) + V_i^{cl}(n_{cr}) = 0. \tag{48}$$

Since the value of $V_i^{cl}(n)$ depends on the amount of interstitials accumulated in clusters, Eq. (48) also defines

the cluster density, at which the coalescence of supercritical loops with smaller clusters and loops starts to take place. As shown in [33], the following condition is necessary to satisfy (48):

$$N_{\mathrm{d}} Z_i^{3/2} \rho_{il}^{5/6} \left[\frac{\langle N_{\mathrm{d}i}^2 \rangle}{N_{\mathrm{d}i}^2} + \frac{Z_v^{3/2}}{Z_i^{3/2}} \frac{1}{(1-\varepsilon_i)^2} \frac{\langle N_{\mathrm{d}v}^2 \rangle}{N_{\mathrm{d}v}^2} \right] \times \left(1 + \frac{4(n_0^{3/2} - n_{\min}^{3/2}) \rho_{il}^{1/3}}{3(n_0 - n_{\min})} \left(\frac{2\Omega}{\pi b^3} \right)^{1/3} \left(\frac{\Omega b}{4\pi} \right)^{1/6} \right)^{1/3} \cong 4b \frac{(1-\alpha)\varepsilon_i (n_0 - n_{\min})}{n_0 (1-\varepsilon_i)^2} \left(\frac{4\pi}{\Omega b} \right)^{2/3} \left(\frac{\pi b^3}{2\Omega} \right)^{1/3}, \quad (49)$$

where ρ_{il} is the total loop line density, the parameter α is positive but less than unity. It has a physical meaning similar to α^* in Eq. (42) [33].

Considering the dislocation as a point-defect sink, Z_i is a function of the loop line density ρ_{il} (dashed lines for different reaction radii in Fig. 6) [34]. At the same time, Z_i is also a function of the loop line density ρ_{il} through (49) (solid lines for different values of α in Fig. 6). Material parameters for stainless steel used in the present calculation are the same as previous ones [17]. Intersections of the two sets of curves represent solutions of Z_i and ρ_{il} that satisfy the two equations simultaneously,

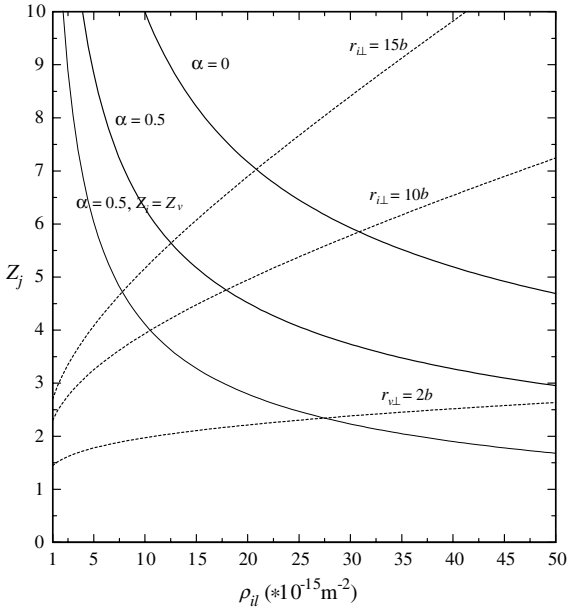

Fig. 6. Reaction constant Z_i (—) between dislocations and mobile interstitials as a function of dislocation line density that satisfies Eq. (49). Dashed lines are for the reaction constants calculated with the conventional expression $Z_j = 4\pi/\ln(1+1/\rho_{jl} r_{j\perp}^2)$ [34], where $r_{j\perp}$ is the effective reaction radius between dislocations and point defects. Low solid line corresponds to the case when Z_i is taken to be equal to Z_v in the second term on the left-hand side of Eq. (49).

which gives the critical values of ρ_{il}. It can be seen from this that interstitial loops start to grow by coalescence when the total loop line density ρ_{il} reaches a value around 2×10^{16} m^{-2}. Values of the reaction constant $Z_i \approx 5$–6 that follow from Fig. 6 also agree with the previous estimations of this constant at the corresponding dislocation line densities [35]. Note that at such line densities the last term on the right-hand sides of (49) is approximately equal to 1 and can be neglected.

It is important to remember that the value of Z_i depends on the total dislocation line density (see solid lines of Fig. 6). Electron irradiation does not generate small interstitial clusters continuously in cascades, and the total dislocation line density is orders of magnitude less than that under cascade-damage irradiation producing immobile SIA clusters. As a result, under electron irradiation dislocation bias is much less and takes the values conventionally used in the literature.

It can also be shown [33] that this line density is mainly contributed from interstitial loops with sizes of $\sim \langle n_i \rangle$ where

$$\langle n_i \rangle \cong \frac{1}{2\rho_{il}^{2/3}} \left(\frac{\pi b^3}{2\Omega} \right)^{2/3} \left(\frac{4\pi}{\Omega b} \right)^{1/3}. \quad (50)$$

Here ρ_{il} is determined by Eq. (49). Since $\rho_{il} = 2\pi \langle r_i \rangle N_{il} \Omega$, the number density Q_{il} of interstitials accumulated in the matrix in the form of clusters and loops is given by

$$Q_{il} = \langle n_i \rangle N_{il} \cong \frac{\rho_{il}^{2/3} b^{4/3}}{2\sqrt{2}}. \quad (51)$$

When the total line density of interstitial loops is about 2×10^{16} m^{-2}, $\langle n_i \rangle \cong 1.3 \times 10^2$ and $Q_{il} \cong 3 \times 10^{-3}$. Eq. (50) also means that most loops encountered by the climbing loop segments contain about $\langle n_i \rangle$ interstitials. Consequently, the critical loop size n_{cr} is also around this value, and interstitial loops with sizes $n_{\mathrm{m}} \gg \langle n_i \rangle$ can be considered to be supercritical.

In addition, the ratio d_{c}/v can be estimated, [33]

$$-\frac{d_{\mathrm{c}}}{v} \cong \frac{N_{\mathrm{d}}(1-\varepsilon_i)^2 n_0}{4\varepsilon_i (n_0 - n_{\min})} \left(\frac{\Omega \rho_{il}}{4\pi b} \right)^{1/2} Z_i^{3/2} \left[1 + \frac{Z_v^{3/2}}{Z_i^{3/2}(1-\varepsilon_i)^2} \right] \cong 10. \quad (52)$$

According to (36), (40), (45), (47) and (52), the ratio $d_{\mathrm{s}}/d_{\mathrm{c}} < 1$. Thus, nucleation of growing interstitial loops under cascade-damage conditions is indeed mainly the result of cascade-induced fluctuations. From Fig. 5, the probability of nucleation of growing loops with sizes $n_{\mathrm{m}} \gg \langle n_i \rangle$ is of the order of 10^{-5}–10^{-6} ($n_{\mathrm{m}} \approx 1500$–$2500$). PICs are generated in collision cascades at a rate of $\varepsilon_i G/n_0$ Then, the rate of nucleation of interstitial loops J_{l}, as given by the ratio $\varepsilon_i G P_{\mathrm{m}}/(\Omega n_0)$, is approximately equal to 10^{21}–10^{22} loops/m^3/(NRT dpa) ($G/K \approx 0.2$, where K is the nominal NRT dpa rate). Thus, the

experimentally observed concentrations of growing interstitial loops would require a characteristic irradiation dose of about one NRT dpa.

5. Swelling in annealed metals

According to the foregoing, at elevated temperatures interstitial loops start growing by coalescence when the total interstitial cluster content of the matrix Q_{il} reaches a value of $\sim 10^{-3}$. Thus, an initial dose is required for the accumulation of this amount of interstitials in small clusters and loops. In the absence of a dislocation network at an early stage of irradiation, each interstitial accumulated in clusters and loops is matched by a vacancy in vacancy clusters and voids. Thus, in annealed metals at elevated temperatures, considering the short lifetime of the vacancy clusters, the accumulation of unpaired vacancies will most likely take place at the voids. The rate of vacancy accumulation in voids dS/dt is given by the conventional equation

$$\frac{dS}{dt} = k_c^2(D_v C_v - D_i C_i) \cong \frac{k_c^2 G}{Z_v \rho_{il}}\left(1 - \frac{Z_v(1-\varepsilon_i)}{Z_i}\right). \qquad (53)$$

The approximation in (53) assumes the dominance of the sink strength of small interstitial clusters and loops, when the equal amounts of vacancies and interstitials are accumulated respectively in voids and in clusters and loops [36]. Using the conservation law $S(t) = Q_{il}(t)$, similar to [36], we can obtain the following approximate expression for the number of defects accumulating in voids and clusters:

$$S(t) = Q_{il}(t) \cong C N_c^{2/5}([1 - Z_v(1-\varepsilon_i)/Z_i]Gt)^{3/5}, \qquad (54)$$

where N_c is the void density,

$$C = (10\langle n_i\rangle \Omega/3 Z_v \langle r_i\rangle)^{3/5}/(4\pi/3)^{1/5}. \qquad (55)$$

The total interstitial content of the matrix Q_{il} as a function of dose is plotted for stainless steel at elevated temperatures in Fig. 7 for different values of the void concentration. It can be seen that irradiation dose of several NRT dpa is required before significant interstitial loop nucleation and dislocation structure development starts to occur. Note that, at temperatures above 550 °C voids are often associated with coarse precipitates of certain radiation-induced and radiation-modified phases [37]. Since these precipitates effectively increase the sink strength for point defects of the associated voids, Eq. (54) is not applicable to such cases.

It was shown in an earlier work that after incubation, further evolution of the dislocation structure does not lead to an increase in the total dislocation line density [17]. The continuous absorption of small interstitial clusters and loops through coalescence, by the super-

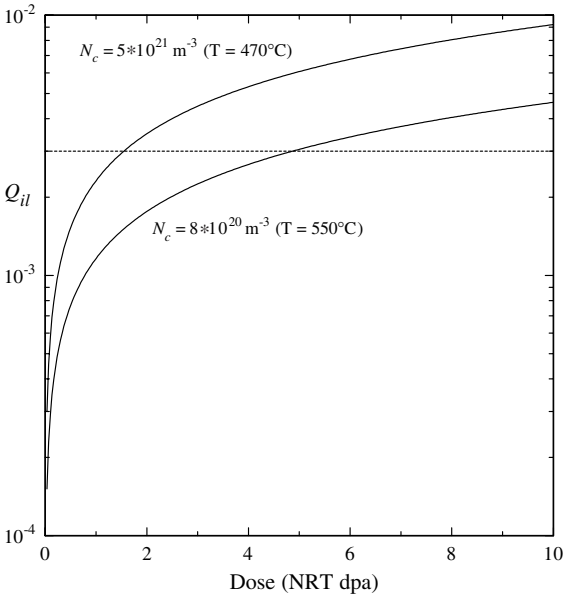

Fig. 7. Dose dependence of the total interstitial content Q_{il} in annealed metal at elevated temperatures given by Eq. (54) at different values of the void density N_c.

critical loops, as well as by climbing network dislocations, prevents further accumulation of interstitials in clusters. In turn, interstitial loops growing beyond the maximum size n_{max} join the dislocation network, the density of which is stabilized via the mutual annihilation of the climbing dislocations. Thus, knowing that the steady-state total dislocation line density ρ_{il} is about 2×10^{16} m^{-2}, we can calculate void swelling rate from Eq. (53). Rewriting,

$$\frac{dS}{dt} = \frac{k_{cv}^2 G}{Z_v \rho_{il} + k_{cv}^2} - \frac{(1-\varepsilon_i)k_{ci}^2 G}{Z_i \rho_{il} + k_{ci}^2}, \qquad (56)$$

where $k_{cj}^2 = 4\pi N_c R_c(1 + R_c k_j)$ is a more accurate expression for the void sink strength [38], $R_c = (3S/4\pi N_c)^{1/3}$ is the average void radius. Note that when the initial swelling is small, the approximation $k_{cj}^2 \cong 4\pi N_c R_c$ as used in the derivation of Eq. (54), is justified. In Fig. 8, we plot the void swelling rate as a function of swelling, showing the upper limit $\varepsilon_i G$. This agrees well with typical behaviour of the swelling rate observed in neutron-irradiated stainless steel [39]. We note that the results in Fig. 8 represents a conservative estimate that corresponds to a high dislocation line density ($\rho_{il} \approx 2\times 10^{16}$ m^{-2}), and smaller values of $\varepsilon_i = 0.2$ and $G/K = 0.2$. More detailed discussion on the void swelling and related microstructure development under cascade-damage irradiation has been presented in [17], based on similar results obtained from numerical calculations.

The actual swelling strain in the irradiated metal is realized through dislocation climb. Thus, under

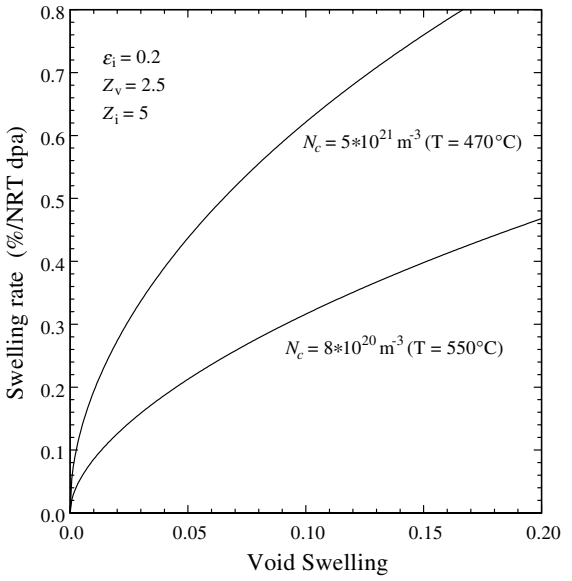

Fig. 8. Swelling rate as a function of swelling according to Eq. (56).

steady-state conditions the net rate of vacancy accumulation in voids must be equal to the net interstitial absorption by the climbing network dislocations, i.e.

$$\frac{dS}{dt} = V_N b \rho_N, \tag{57}$$

where V_N is the climb velocity of network dislocations.

Network dislocation line density ρ_N increases when interstitial loops join the network. At the same time, the climbing network dislocations also annihilate each other when segments of opposite signs meet and react, for example, by the dipole mechanism [40]. Adopting the simple recovery model for the dislocation in the absence of external stress [40], one can write down the equation for the network evolution

$$\frac{d\rho_N}{dt} = 2\pi r_i(n_{max}) J_N(n_{max}) - V_N \rho_N^{3/2}. \tag{58}$$

Here $J_N(n_{max})$ is the flux in the size space of interstitial loops, which grow beyond the maximum size n_{max} and join the dislocation network. From Eqs. (57) and (58), under steady-state conditions ($d\rho_N/dt \cong 0$), we obtain

$$J_N(n_{max}) \cong \frac{\rho_N}{2\sqrt{\pi} b} \frac{dS}{dt}. \tag{59}$$

In deriving Eq. (59) we assume, as usual, that $\pi r_i^2 \times (n_{max}) \cong 1/\rho_N$.

Typical steady-state density of network dislocations in stainless steel is about 5×10^{14} m^{-2} [39]. Thus, for the maximum swelling rate (~1%/NRT dpa [39]) experimentally observable at elevated temperatures to be realized the flux of interstitial loops joining the network has to be about 6.8×10^{21} loops/m^3/(NRT dpa), which is consistent with the rate of nucleation of supercritical loops calculated in the previous paragraph. The rate of nucleation analytically obtained in Section 4 also agrees well with the results of the numerical calculations of $J_N(n_{max})$ [17].

6. Conclusion

In this paper, we have shown that, within the framework of the classical theory of nucleation of overcritical precipitates from small subcritical nuclei, the nucleation processes of both voids and interstitial loops from the primary clusters are similar. Based on this framework, a kinetic model for single-component nucleation is formulated, which is applicable to both the nucleation of voids and dislocation loops at elevated temperatures and under the operation of the production bias. This model, like the production bias model, is based on general physical principles, and is not material specific. The relative simplicity in this model allows an analytic approach to be followed. General expressions for the nucleation probability have been derived, containing explicit dependence on material properties and irradiation conditions, thus allowing their applications to a wide class of materials and conditions, from which loop and void nucleation can be investigated.

In the conventional approach, nucleation theory is usually considered with the statistical fluctuations accounted for solely by random point-defect jumps. The diffusion coefficient in the size space of the embryo subject to such fluctuations is related to the sum of the average point-defect fluxes, and is linearly related to the radius of the embryo. The neglected fluctuations in the vacancy emission rate, and those caused by random cascade initiation, on the other hand, have diffusion coefficients proportional to the surface area of the nucleus. It is found that at elevated temperatures and when the sink density is low, fluctuations in the rate of vacancy emission from voids is the dominant factor that governs void nucleation. In this regard, the conventional assumption that fluctuations in the vacancy emission rate are linearly proportional to the void radius seriously underestimates the void nucleation rate.

It is also found that the effects of cascade-induced fluctuations become important when the total sink strength for point defects is higher than, say, 10^{15} m^{-2}. When the nucleation rate is high and the average void growth rate is low, cascade-induced fluctuations may become a dominating factor on the void evolution, as the total sink strength increases due to the nucleation and growth of voids. Under such circumstances, the majority of voids, particularly the smaller ones, will shrink away, while the largest ones maintain their growth. The resulting void coarsening is not caused by

vacancy emission from the voids, but by the stochastic fluctuations in the point-defect fluxes received by the void.

Application of the present approach to void nucleation in annealed pure copper at elevated temperatures neutron-irradiated to doses of 10^{-4}–10^{-2} NRT dpa showed reasonable agreement with experimental results. The theory has also been applied to neutron-irradiated molybdenum at temperatures at which vacancy emission from voids is negligible. The calculated void nucleation rates also show reasonable agreement with experimental results.

At elevated temperatures only sufficiently large (supercritical) interstitial loops are able to grow through coalescence with smaller clusters and loops. This starts to become important when the total line density of small clusters and loops reaches a high value of 2×10^{16} m^{-2}. In the presence of voids of sufficiently high concentration, an incubation dose of several NRT dpa is required to accumulate a sufficient interstitial content in the matrix ($Q_{il} \cong 3 \times 10^{-3}$) for the development of the dislocation structure to start. The interstitial content of $Q_{il} \sim 10^{-3}$, required for the interstitial loop growth at elevated temperatures, is in agreement with the corresponding values obtained from the numerical calculations [17].

After creation, PICs can only grow beyond the critical size via stochastic fluctuations, which is mostly cascade-induced at a sink density of $\sim10^{16}$ m^{-2}. Since the probability of nucleation of a supercritical loop from a small shrinking interstitial cluster decreases exponentially with its size, inclusion of the effect of the cascade-induced fluctuations on the loop diffusion coefficient leads to several orders of magnitude of increase in the nucleation probability. The calculated rate of nucleation of growing interstitial loops based on the presently derived nucleation probability is sufficient ($\sim10^{22}$ loops/m^3/NRT dpa) to account for the experimentally observed number densities of interstitial loops at a dose of one NRT dpa. This rate is also sufficient to account for the observed swelling rates about 1%/NRT dpa.

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