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The effect of bias factor variations on void nucleation in irradiated alloys

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Abstract

The effect of the variations of void and dislocation biases on the homogeneous nucleation of voids in irradiated structural materials is considered. It is demonstrated that in some cases a modest variation of dislocation bias can result in the orders of magnitude differences in kinetic barriers for void nucleation.

1. Introduction

This paper considers to which extent homogeneous void nucleation in an irradiated metal can be accelerated by dislocation bias modification. The underlying motivation for the study is related to the yet unexplained effect of impurities on swelling in vanadium alloys. Indeed, while pure vanadium is known for very low swelling as compared to austenitic stainless steels, the addition of 5–10 wt% of undersized solutes (such as iron [1–3] or chromium [4]) can increase the swelling rate of vanadium by two orders of magnitude, making it comparable to or even higher than that in austenitic stainless steels. One of the possible reasons for this effect might be the dislocation bias modification by impurity segregation at dislocation lines, which was observed to precede the onset of enhanced swelling in V–5%Fe [1].

The swelling enhancement can be generally ascribed to dislocation bias modification effect either on void growth, or on the void nucleation. However, the effect on void growth does not seem a proper explanation, because (i) the bias factor modification due to impurity segregation at dislocations is rather modest (increase by a factor of 2 at solute concentration ~5% [5]), and (ii) when voids are present in a bcc metal, their growth rates are expected to be of the same order of magnitude as those in stainless steels [6]. Therefore, below we concentrate on the influence of dislocation bias modification on void nucleation.

The swelling of pure vanadium is small (~3% at 100 dpa), although cavities are sometimes observed in neutron-irradiated vanadium by TEM [7,8]. The extremely high number density of these cavities indicates that these are bubbles, stabilized by internal pressure of residual gas impurities, most probably oxygen. This is in agreement with the experimental observations that comparably high amounts of bubbles arise during vanadium implantation with helium, even at temperatures noticeably above 500 °C (that of peak swelling in V–5%Fe) [9]. Even when present in the material, bubbles should first undergo a conversion to voids, in order to grow exclusively by preferential absorption of vacancies and produce high swelling rates [10]. Without continuous gas supply, the bubble-to-void conversion still requires overcoming an (appropriately reduced) kinetic nucleation barrier [11]. Here we restrict ourselves to the most severe case, when no gaseous impurity assists void nucleation, and neglect the effect of residual gases on void formation.

When considering the effect of irradiation on homogeneous nucleation, the most important parameter is the amount of vacancies available for nucleation. Since point defects nucleate as Frenkel pairs, there should exist some internal mechanisms of vacancy and interstitial partitioning between nucleated voids and other point defect sinks. At present two sources of vacancy-interstitial imbalance are considered, namely the dislocation bias for interstitial absorption and, in case of fast

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neutron and heavy ion irradiation, the direct in-cascade clustering of interstitials ('production bias' [12]). Here we restrict ourselves only to the first contribution; for a discussion of the production bias effect on nucleation the reader is referred to Ref. [13]. Indeed, the impurity-accelerated swelling was observed for other than fast neutron irradiation (e.g. during self-ion irradiation [3] or irradiation with 4 MeV nickel ions [9]) and there is no reason to expect that high-energy electrons would not induce the swelling of vanadium alloys at a rate comparable to that found for neutron irradiation.

2. Theory

Qualitatively, we should expect that the increase of the dislocation bias for interstitials should decrease the kinetic barrier for void formation, due to the increased relative amount of vacancies available for clustering, and vice versa. Since the sign of dislocation modification is opposite to the sign of impurity elastic misfit [5], a selective swelling enhancement by undersized impurities should be expected, in qualitative agreement with experimental observations [2]. On the other hand, because the solute segregation at dislocations provides very moderate dislocation bias modification, let us try a more quantitative validation of its relevance.

As already mentioned, a vacancy cluster grows in a steady-state mode provided the number of vacancies in it overcomes a certain critical value n_c. Smaller clusters have a tendency to dissolve. Mathematically it means that for a vacancy cluster (void) with the a number of vacancies n, the probability of cluster growth due to absorption or evaporation of individual point defects, $P(n,t)$, is smaller then that for its dissolution, $Q(n,t)$, when $n < n_c$, and visa versa at $n > n_c$. The critical cluster size is defined by the requirement

$$P(n_c) = Q(n_c). \tag{1}$$

In other words, formation of voids in pure metals is a classical example of kinetic barrier limited nucleation [14] and is usually considered in terms of the Fokker–Planck equation formalism (see e.g. [15–21]).

Accordingly, the evolution of void ensemble follows three well-defined stages, namely the incubation period (when no supercritical voids are present), the nucleation stage with intensive production of supercritical clusters at a nearly constant rate I, and the growth stage, when the already existing voids grow, but practically no new voids are produced.

For our purpose here only the first two stages are important. According to the general treatment of barrier nucleation theory [14], both the duration of the incubation stage (that is, the swelling incubation time t_c) and the void nucleation rate J can be expressed in terms of the so-called kinetic nucleation barrier $\varphi(n_c)$, namely

$$t_i \propto \exp(\varphi(n_c)) \tag{2}$$

and

$$J \propto \exp(-\varphi(n_c)), \tag{3}$$

while the nucleation barrier is determined by the ratio of point defect absorption and description probabilities,

$$\varphi(n_c) = \int_{n_0}^{n_c} \ln\left(\frac{Q(r)}{P(r)}\right) \mathrm{d}n, \tag{4}$$

where n_0 is the smallest size of an immobile vacancy cluster (that is, one should choose $n_0 \geqslant 2$).

There are at least two physical reasons for the existence of a critical void size. Most often it is related to the vacancy evaporation from voids, since, according to the Gibbs–Thomson law, the thermal vacancy concentration at void surfaces exceeds that in the bulk of material. However, at the temperatures relevant for operation regimes of fusion and fission reactors, the thermal solubility of vacancies in most structural materials is too low to provide any barrier for void nucleation. Nonetheless, at these temperatures the growth of small voids remains hindered due to the intrinsic void bias [22], stipulated by the elastic void interaction with point defects (a brief review of various mechanisms of such interaction see e.g. in [22,23]).

Correspondingly, the kinetic coefficients of void growth and dissolution are determined by the absorption of, respectively, vacancies and interstitial atoms created by irradiation and one can express the rates of individual void increase and decrease in terms of diffusion currents of vacancies and interstitials as

$$P(n) = 4\pi R Y_v(n) D_v C_v \quad \text{and} \quad Q(n) = 4\pi R Y_i(n) D_i C_i, \tag{5}$$

where Y_α is the void bias factor due to the interaction of point defects of the type α ($\alpha = i$ for interstitials and v for vacancies) with the elastic field of the void, D_α is the diffusion coefficient and C_α – the mean-field concentration of corresponding point defects. At temperatures where the point defect recombination is negligible as compared to loss at sinks, point defect concentrations are defined by the balance equations

$$G - k_\alpha^2 D_\alpha C_\alpha = 0, \tag{6}$$

where G is the point defect generation rate and k_α^2 – the sink strength for α-type point defects. At the nucleation stage the contribution from nucleated voids to the sink strengths is negligible. In most cases the sink strength is determined by the dislocation network and thus

$$k_\alpha^2 \simeq Z_\alpha \rho_d, \tag{7}$$

where Z_α is the dislocation bias factor for α-type point defects and ρ_d – the dislocation density. Then the ratio P/Q can be written down as

$$\frac{P(n)}{Q(n)} = \frac{Z_i Y_v(n)}{Z_v Y_i(n)}. \tag{8}$$

For all known mechanisms of void interaction with point defects this is a monotonically growing function of void size [23], which can fall below unity at small void sizes, where the void bias for interstitials is strong, and which tends to $Z_i/Z_v > 1$ as the void size becomes sufficiently large. The critical void size is defined by the relation

$$Y_i(n_c)Z_v = Y_v(n_c)Z_i. \tag{9}$$

At the experimentally expected values of critical void size (corresponding to void radius R_c of several nanometers) the dominant contribution to the void bias factors comes from the diffusion anisotropy of point defects in the void elastic field and the following approximation for Y_α holds [23]

$$Y_\alpha = 1 + \frac{R_\alpha}{R}, \tag{10}$$

where the void radius R is related to n as $4\pi R^3/3\Omega = n$, Ω is the atomic volume, and R_α is determined by the components of the elastodiffusion tensor of point defects in the saddle points of their diffusion jumps. In this case the critical radius can be explicitly obtained in the form

$$R_c = \frac{R_i Z_v - Z_i R_v}{Z_i - Z_v} = \frac{1}{B}[R_i - (1+B)R_v], \tag{11}$$

where $B = (Z_i - Z_v)/Z_v$ is the dislocation bias. It is seen that the nucleation barrier appears only when $R_i Z_v > R_v Z_i$. The estimates of R_c for the parameter values typical for copper and α-iron are given in Table 1.

The kinetic nucleation barrier in this case is obtained by the direct integration using (5) and (10) and has the form

$$\varphi(R_c) = \frac{4\pi}{3}\left\{R_i^3 \ln\left(1 + \frac{R_c}{R_i}\right) - R_v^3 \ln\left(1 + \frac{R_c}{R_v}\right) + R_c(R_i - R_v)\left(\frac{R_c}{R_v} - R_i - R_v\right)\right\}, \tag{12}$$

where the small terms proportional to $R_0/R_c \ll 1$ are neglected. The dependencies of the critical radius R_c and the nucleation barrier $\varphi(R_c)$ on the dislocation bias B are shown in Fig. 1. The increase of dislocation bias results in a decrease of both the critical radius and the nucleation barrier $\varphi(R_c)$. Correspondingly, the incubation time for swelling decreases, while the nucleation rate grows. In particular, when the dislocation bias is not too high, the slope of the nucleation barrier is sharp and even a rather modest increase of B can decrease the nucleation barrier by a factor of 3–4. Having in mind the exponential dependence of t_i and J on the kinetic barrier value, this can explain one–two orders of magnitude variation of the void nucleation rate. Since the values of both dislocation and void bias factors are quite different in different materials (as demonstrated in Table 1), it may well turn out that vanadium falls in the parameter range, where void nucleation is highly sensitive to material properties.

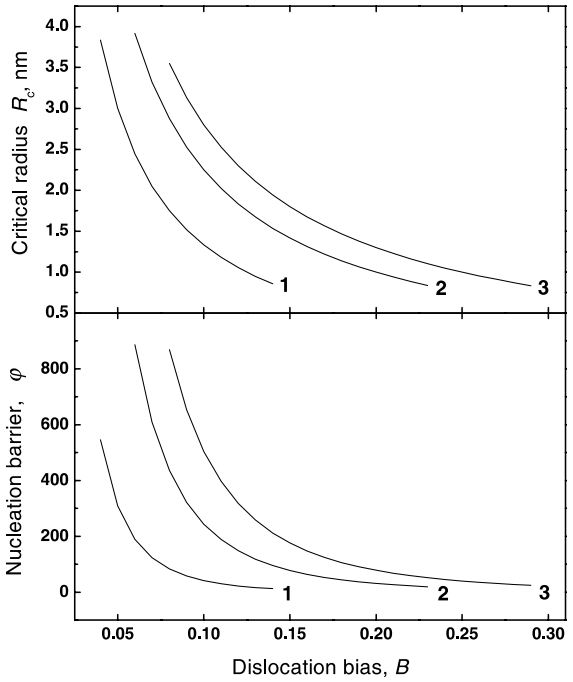

Fig. 1. The critical radius R_c (a) and the nucleation barrier $\varphi(R_c)$ (b) as functions of the dislocation bias B for $R_i = 0.5$ nm and $R_v = 0.33$ nm (1), 0.25 nm (2) and 0.2 nm (3).

Table 1
The values of the void bias parameters, the estimated dislocation bias and the critical void radius in copper and α-iron

	R_i (nm)	R_v (nm)	B	R_c (nm)
α-iron	0.7	−0.17	0.3	3.1
Copper	0.23	−0.28	0.13	4.2
Ref.	[23]	[23]	[6]	

3. Conclusions

To conclude, the estimates above demonstrate that the modification of dislocation bias due to impurity segregation at dislocations allows, in a certain parameter range, a decrease in swelling incubation time and an increase in void nucleation rate by one–two orders of magnitude. This result might provide a possible explanation of the experimentally observed noticeable swelling increase in vanadium alloyed with several percent of undersized impurity atoms.

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