STEADY-STATE SIZE DISTRIBUTION OF VOIDS IN METALS UNDER CASCADE IRRADIATION.-
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OBJECTIVE

The theory of radiation damage in metallic materials predicts that under cascade-irradiation conditions the voids should approach a steady state, which is characterised by a maximum mean void size. It is shown in this Letter that the steady-state concentrations of voids of different size are described by the Gaussian distribution with the maximum size mentioned above to be the most probable value. The evolution of voids towards the steady state is analysed.

SUMMARY

The steady state of the void population predicted by the theory under cascade-irradiation conditions has been analyzed. The following conclusions have been drawn.

1) The theoretical steady-state SDF of voids is described by a Gaussian distribution, which is quite narrow, in contrast to much bigger spread of void sizes observed.
2) At high void density, when $\alpha >> 1$, the irradiation dose required to reach the steady state is higher than those at which void lattices are observed. Hence, the void size saturation of randomly distributed voids and in the lattice are not related to each other.
3) At low void density, when $\alpha \leq 1$, the irradiation dose required to reach the steady state is relatively small and might be a reason for the incubation period of swelling frequently observed.

PROGRESS AND STATUS

I. Introduction

Since the prediction [1] and discovery [2] of swelling in metals in nuclear reactors, much effort has been made to formulate a theory of the phenomenon. The 'Production Bias Model' (PBM) in its modern form succeeds in explaining several striking observations, for example, enhanced swelling rates near grain boundaries and in materials with small grain size and under neutron compared to electron irradiation [3-6]. The model owns its success to the recognition of two distinguishing features of defect production by high-energy recoils. First, that clusters of self-interstitial atoms (SIAs) are formed directly in displacement cascades, the fact revealed both experimentally [7] and in molecular dynamics (MD) simulations [8,9], and, second, that these clusters execute one-dimensional diffusion [9-13], a phenomenon proposed in [9,14] for the explanation of the void-lattice formation [15,16].

The model predicts that, if a random distribution of voids is maintained, a steady state should establish at high irradiation doses, which is characterised by a maximum void size, r_{m}, above which the net vacancy flux to voids is negative. This is because the cross-section of the interaction of three-dimensionally (3-D) diffusing vacancies with voids is proportional to the void radius r, while that of the 1-D migrating SIA clusters to r^2. As a result, above some critical radius, the latter becomes higher than the former. It has been shown that $r_{\mathrm{m}} \approx 2\pi r_{\mathrm{d}} / Z_{\mathrm{v}}$, where r_{d} is the dislocation capture radius for the SIA clusters and Z_{v} is the capture efficiency of dislocations for vacancies [3]. Note that this expression does not include the dependence on the dislocation bias for point defects: $B = Z_{\mathrm{i}} / Z_{\mathrm{v}} - 1$, where Z_{i} is the capture efficiency of dislocations for single SIAs, which accounts for one of the main driving forces for the void growth [1].

In this Letter we derive the dependence of the critical void radius on the dislocation bias factor and an equation for the steady-state size distribution function (SDF) of voids and analyse how voids approach the steady state.

II. Steady-state size-distribution function

Let us assume that the primary damage produced in cascades consists of 3-D mobile single vacancies and SIAs and 1-D mobile SIA clusters. In addition, let us assume that the void nucleation stage is over and the mobile defects interact only with existing voids of the number density N and dislocations of the density ρ. Then, according to the PBM (see, *e.g.* [6]), the rate of swelling is equal to the difference in arrival rates of vacancies, j_{v}, single SIAs, j_{i}, and SIAs in clusters, j_{cl}, to voids

$$\frac{dS}{d\phi} = j_{\mathrm{v}} - j_{\mathrm{i}} - j_{\mathrm{cl}} = \frac{4\pi rN}{4\pi rN + Z_{\mathrm{v}}\rho} - (1-\varepsilon_{\mathrm{i}}^{\mathrm{g}})\frac{4\pi rN}{4\pi rN + Z_{\mathrm{i}}\rho} - \varepsilon_{\mathrm{i}}^{\mathrm{g}}\frac{\pi r^2 N}{\pi r^2 N + \pi\rho r_{\mathrm{d}}/2}, \quad (1)$$

where $S = 4\pi r^3 N/3$, r is the mean radius of voids, $\varepsilon_{\mathrm{i}}^{\mathrm{g}}$ is the fraction of the SIAs produced in the clustered form and ϕ is the irradiation dose. The irradiation dose is in displacements per atom (dpa) and accounts for the fraction of defects that survived intra-cascade recombination, ε_S; hence it corresponds to the dpa calculated using the NRT standard procedure [17] and multiplied by ε_S.

Eq. (1) cannot be used below some temperature, when the vacancy or SIA diffusivity is so low that recombination reactions between mobile species are important. At higher temperature, the defect nobilities do not enter the analysis and the swelling is determined by partitioning of defects between sinks, as in Eq. (1). Since the recombination between cascade-produced mobile vacancies and SIAs in clusters have not been studied so far, any estimates are not available. Another limitation is due to neglecting vacancy emission from voids, which requires high vacancy super-saturation and thus restricts the analysis to temperatures below ~(0.5÷0.6) T_{m}, where T_{m} is the melting temperature, depending on the dose rate. Also, it is assumed that the void nucleation stage is separated from the growth stage due to reduced thermal stability of void nuclei. The validity of this assumption and the nucleation itself is in fact one of the fundamental unresolved problems, which is closely connected with the validity of conventional assumption of homogeneous spatial distribution of defects in the system and is analysed in [18,19].

It can readily be obtained from Eq. (1) that the maximum mean void radius, which corresponds to zero swelling rate, is

$$r_{\mathrm{m}} \approx \frac{2\pi r_{\mathrm{d}}}{Z_{\mathrm{v}}}\left(1 + \frac{1-\varepsilon_{\mathrm{i}}^{\mathrm{g}}}{\varepsilon_{\mathrm{i}}^{\mathrm{g}}}B\right), \quad (2)$$

where we omitted higher order terms in B. (For a comprehensive analysis see in a separate paper [18].) According to Eq. (2), r_{m} increases with decreasing $\varepsilon_{\mathrm{i}}^{\mathrm{g}}$, so that there is no saturation of swelling in the limit of $\varepsilon_{\mathrm{i}}^{\mathrm{g}} = 0$, *i.e.* for electron irradiation, as expected. It worth mentioning that a more rigorous analysis predicts an unlimited swelling even at finite $\varepsilon_{\mathrm{i}}^{\mathrm{g}}$ somewhat smaller than *B* [18].

In order to derive the steady-state SDF of voids, $f(x) = f(x, \phi = \infty)$, where the number of vacancies in a void of radius r_x is $x = 4\pi r_x^3/3\Omega$, Ω is the atomic volume, we consider the Smoluchowski (continuity) equation for the diffusion of voids in the size space:

$$\lim_{\varphi\to\infty}\frac{\partial f(x,\phi)}{\partial\phi} = \frac{d}{dx}\left\{-V(x)f(x) + \frac{d}{dx}\left[D(x)f(x)\right]\right\} = 0. \quad (3)$$

Here $V(x) = j_{\mathrm{vx}} - j_{\mathrm{ix}} - j_{\mathrm{clx}}$ is the velocity and $D(x) = (j_{\mathrm{vx}} + j_{\mathrm{ix}} + j_{\mathrm{clx}})/2$ is the diffusion coefficient, where $j_{kx} = j_k r_x / rN$ ($k = \mathrm{v,i}$) and $j_{\mathrm{clx}} = j_{\mathrm{cl}} r_x^2 / r^2 N$. Eq. (3) represents the familiar master equation

for the evolution of the SDF of voids *via* absorption and emission of different defects in the diffusion limit of large void size (see, *e.g*. [20,21]). Its solution that provides zero flux of voids (the term in curly brackets) is

$$f(x) = A\exp\left[\int dxV(x)/D(x)\right]/D(x), \tag{4}$$

where A is a normalising constant. We expect that, in most cases, the SDF will be a narrow function around $x_{\mathrm{m}} = 4\pi r_{\mathrm{m}}^3/3\Omega$, such that $\left|x/x_{\mathrm{m}} - 1\right| << 1$. With this condition, it is readily obtained that, to a first approximation, $D(x)$ is a constant and

$$\frac{V(x)}{D(x)} \approx \lambda\varepsilon_{\mathrm{i}}^{\mathrm{g}}\left(1 - \frac{r_x}{r_{\mathrm{m}}}\right) \approx \frac{\lambda\varepsilon_{\mathrm{i}}^{\mathrm{g}}}{3}\left(1 - \frac{x}{x_{\mathrm{m}}}\right), \tag{5}$$

where

$$\lambda = 1 + \frac{1-\varepsilon_{\mathrm{i}}^{\mathrm{g}}}{\varepsilon_{\mathrm{i}}^{\mathrm{g}}}\frac{B}{1+\alpha}, \tag{6}$$

$\alpha = 4\pi r_{\mathrm{m}} N / Z_{\mathrm{v}}\rho$ and only the first order term in B is retained. By substituting Eq. (5) into Eq. (4), one obtains the SDF as Gaussian distribution centred on the most probable size, x_{m}:

$$f(x) \approx C_0\sqrt{\frac{\lambda\varepsilon_{\mathrm{i}}^{\mathrm{g}}}{6\pi x_{\mathrm{m}}}}\exp\left[-\frac{(x-x_{\mathrm{m}})^2}{6x_{\mathrm{m}}/\lambda\varepsilon_{\mathrm{i}}^{\mathrm{g}}}\right], \tag{7}$$

where $C_0 \approx \int dxf(x)$ is the total void concentration. The SDF has a half-width at half-maximum of $\sigma = \sqrt{x_{\mathrm{m}} 6\ln 2/\lambda\varepsilon_{\mathrm{i}}^{\mathrm{g}}}$ and is thus narrow: $\sigma << x_{\mathrm{m}}$ for reasonable values of $\varepsilon_{\mathrm{i}}^{\mathrm{g}}$ (~0.5 according to MD studies of displacement cascades in Fe and Cu for the primary knock-on atom energy $E_{\mathrm{PKA}} \approx$ 10keV [22]). We note that the experimentally observed spreads of void sizes are obviously much bigger and reasons for this are discussed below.

Figure 1 shows the SDF calculated using Eq. (7) for B=0.04, α=1, $\varepsilon_{\mathrm{i}}^{\mathrm{g}}$=0.25, 0.5 and 1 (open symbols and connecting lines). In this figure, $x_{\mathrm{m}}^1 \approx 10^4$ corresponds to $r_{\mathrm{m}}^1 \approx \pi$ nm and is the most probable void size for $\varepsilon_{\mathrm{i}}^{\mathrm{g}}$=1. As can be seen, with decreasing $\varepsilon_{\mathrm{i}}^{\mathrm{g}}$, the SDF becomes wider and shifts towards bigger void size due to increase of x_{m} according to Eq. (2). Additional data shown on the same graph by full symbols are the result of a full-scale calculations of the temporal evolution of SDF, performed using a computer code described in ref. [4,21], and compare perfectly well with the analytical results. We note that the case of electron irradiation is obtained in the limit $\varepsilon_{\mathrm{i}}^{\mathrm{g}} \to 0$. Eqs. (2) and (7) are not supposed to be valid in this case, but, show qualitatively correct behaviour, namely, that there is no saturation of swelling.

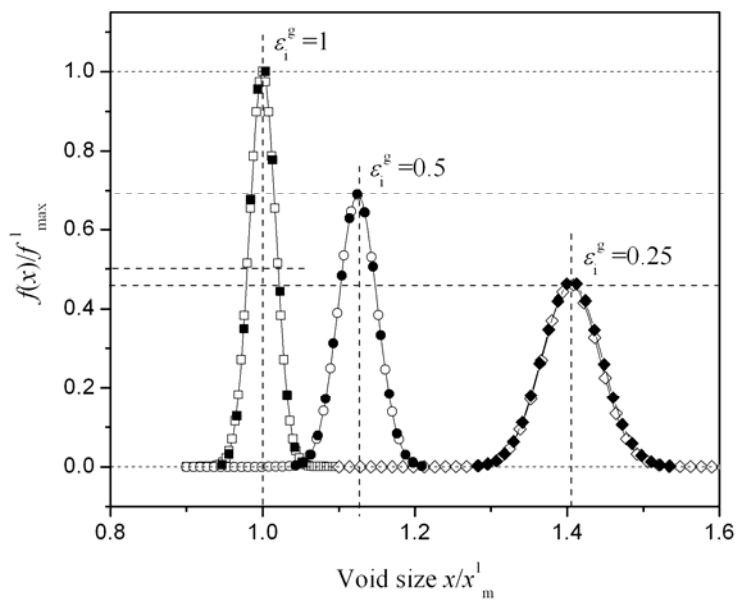

Figure 1. The SDF of voids calculated for B =0.04, α =1 and ε_i^g =0.25, 0.5 and 1. x_m^1 =10^4 is the most probable void size for ε_i^g =1. The SDFs are normalised by the maximum value, f_{max}^1, calculated for ε_i^g =1, assuming C_0 to be the same.

The analysis presented is limited to random distribution of voids and any correlation in void positions, *e.g.* due to formation of ordered structures [15,16], would unavoidably change the kinetics [23]. The most significant result is the contradiction of a very narrow width of the theoretical SDF with much bigger spreads of the void sizes observed. This discrepancy seems important for uncovering fundamental mechanisms of damage accumulation, and is discussed further below.

III. Approach of the steady state

To analyse how the steady state is established, we simplify Eq. (1) by taking $Z_i = Z_v$:

$$\frac{dS}{d\phi} \approx \frac{\varepsilon_i^g \alpha r r_m (r_m - r)}{(r_m + \alpha r)(r_m^2 + \alpha r^2)}. \tag{8}$$

In the limiting case, when $r << r_m$ and voids are dominant sinks, the dose dependence of the mean void radius from its initial value r_0, formed during nucleation stage not considered here, is readily obtained

$$r = \left[r_0^5 + \frac{5\varepsilon_i^g Z_v \rho r_m}{(4\pi N)^2} \phi \right]^{1/5}, \quad \alpha \left(\frac{r}{r_m} \right)^2 >> 1, \tag{9}$$

which is similar to that obtained in [4,24].

Eq. (8) predicts a big difference in doses required to reach the equilibrium at low and high void density. This effect is demonstrated by the full-scale calculations of the dose dependence of the mean void radius on the irradiation dose performed using a computer code described in ref. [4,21]. The results are shown in Fig. 2. The value of ε_S is assumed to be equal to 0.1 (*i.e.* ~ half that given by MD simulation of cascades for E_{PKA} > 5keV [22]; the factor of ½ is an assumed fraction of defects that recombine during the cascade annealing). The value of π corresponds to the void saturation radius at $\varepsilon_i^g = 1$ or B =0. As

can be seen, when the void size is small enough, the curve calculated for α =100 is described satisfactorily by Eq. (9). Also, the doses required for reaching the steady state in this case is higher compared to that for α =0.01 by more than three orders of magnitude (a precise value is unknown since the calculations for α =100 were terminated at 100dpa). There are two important consequences of this effect.

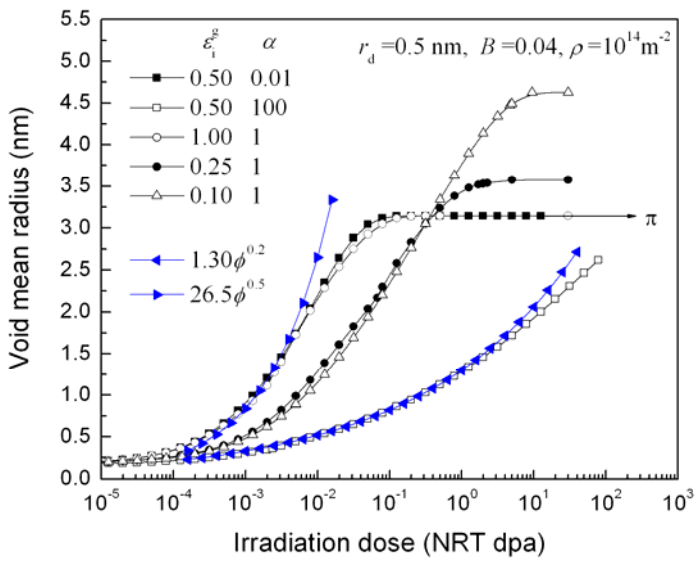

Figure 2. The dependence of the mean void radius on the irradiation dose, ϕ, calculated for different values of ε_i^g and α .

First, the steady state of void population distributed randomly is likely to be unrelated to the void lattices formation and possible saturation of swelling in void lattices. Indeed, the formation of void lattices in metals and alloys under cascade irradiation is observed at high void densities and in the dose range from several to several tens of dpa [16]. As can be seen from Fig. 2, at a high void density, α =100 (for $\varepsilon_i^g = 0.5$), the irradiation dose required to reach the steady state is much higher than 100 dpa, hence much higher than in experiments. In addition, it has been shown previously [23] that the formation of free channels between voids in void lattices provides escape roots for the SIA clusters to dislocations and leads to a significant increase of the void saturation radius. Thus, there must be some other reasons behind the saturation observed.

Second, it is usually observed that, at relatively low void densities, when $\alpha \leq 1$, voids start to grow after some incubation period and the growth is unlimited [25]. Our calculations presented on Fig. 2 show that the dose required to reach the steady state in these conditions is small, ~1 dpa for α =1 and ε_i^g =0.25, and generally in the range from 10^{-2} to 10 dpa depending on ε_i^g and α . It is tempting to think that the incubation period of swelling could be because voids reached their critical radius. It is worth mentioning that, despite many successes of the PBM in explaining features of microstructure evolution of metallic materials under neutron irradiation at low irradiation doses (<1dpa), an unlimited void growth observed at higher doses after the incubation period of swelling cannot be explained in the framework of the model as formulated. Possible ways of resolving this contradiction are proposed in a separate paper [18].

The c-type voids were the smallest and made the smallest contribution to swelling, while the a-type voids were the largest.

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REFERENCES

[1] G.W. Greenwood, A.J.E. Foreman, D.E. Rimmer, J. Nucl. Mater. 4 (1959) 305.
[2] C. Cawthorne, E.J. Fulton, Nature 216 (1967) 575.
[3] H. Trinkaus, B.N. Singh, A.J.E. Foreman, J. Nucl. Mater. 199 (1992) 1.
[4] B.N. Singh, S.I. Golubov, H. Trinkaus, A. Serra, Yu.N. Osetsky, A.V. Barashev, J. Nucl. Mater. 251 (1997) 107.
[5] S.I. Golubov, B.N. Singh, H. Trinkaus, Philos. Mag. A 81 (2001) 2533.
[6] B.N. Singh, M. Eldrup, S.J. Zinkle, S.I. Golubov, Philos. Mag. A 82 (2002) 1137.
[7] B.V. Guerard, D. Grasse, J. Peisl, Phys. Rev. Lett. 44 (1980) 262.
[8] T. Diaz de la Rubia, M.W. Guinan, Phys. Rev. Lett. 66 (1991) 2766.
[9] A.J.E. Foreman, W.J. Phythian, C.A. English, Philos. Mag. A 66 (1992) 671.
[10] M. Kiritani, J. Nucl. Mater. 251 (1997) 237.
[11] T. Hayashi, K. Fukmuto, H. Matsui, J. Nucl. Mater. 307-311 (2002) 993.
[12] K. Arakawa, K. Ono, M. Isshiki, K. Mimura, M. Uchikoshi, H. Mori, Science 318 (2007) 956.
[13] Yu.N. Osetsky, D.J. Bacon, A. Serra, B.N. Singh, S.I. Golubov, Philos. Mag. A 83 (2003) 61.
[14] A.J.E. Foreman, UKAE Authority Harwell Report No. AERE-R-7135 (1972).
[15] J.H. Evans, Nature 229 (1971) 403.
[16] W. Jäger, H. Trinkaus, J. Nucl. Mater. 205 (1993) 394.
[17] M.J. Norgett, M.T. Robinson, I.M. Torrens, Nucl. Eng. and Design 33 (1975) 50.
[18] A.V. Barashev and S.I. Golubov, ORNL Report ORNL/TM-2008/141 (2008), available online via the US DOE information bridge http://www.osti.gov/bridge.
[19] A.V. Barashev, S.I. Golubov, submitted to Philos. Mag. (2008).
[20] F.C. Goodrich, Proc. R. Soc. London A 277 (1964) 167.
[21] S.I. Golubov, A.M. Ovcharenko, A.V. Barashev, B.N. Singh, Philos. Mag. A 81 (2001) 643.
[22] D.J. Bacon, Yu.N. Osetsky, R. Stoller, R.E. Voskoboinikov, J. Nucl. Mater. 323 (2003) 152.
[23] A.V. Barashev, S.I. Golubov, H. Trinkaus, Philos. Mag. A 81 (2001) 2515.
[24] B.N. Singh, A.J.E. Foreman, Philos. Mag. A 66 (1992) 975.
[25] F.A. Garner, M.B. Toloczko, B.H. Sencer, J. Nucl. Mater. 276 (2000) 123.
[26] A.V. Kozlov, I.A. Portnykh, L.A. Skryabin, E. A. Kinev, J. Nucl. Mater. 307–311 (2002) 956.
[27] I.A. Portnykh, A.V. Kozlov, V.L. Panchenko, V.M. Chernov, F.A. Garner, J. Nucl. Mater. 367–370 (2007) 925.