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Letter to the Editor

Void concentration for heterogeneous microstructure development in annealed metals

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The development of spatially heterogeneous microstructure at elevated temperatures is a characteristic of low-dose neutron damage in fully annealed metals such as copper [1–5]. The nucleation and growth of voids take place exclusively in regions with relatively low interstitial loop and cluster density [3], with typical sizes of the order of several microns (about 10 average void spacings) [3]. At the same time the nucleation and growth of interstitial loops are found to concentrate in localized long narrow patches and blocks, with varying thicknesses from 0.05 to 0.5 μm [3], resulting in void and dislocation populations that are almost completely separated spatially [2–4].

Experimental results of electron and 3 MeV proton irradiations at the same dose rate showed a very different microstructure, one that is spatially more homogeneous and in which voids and dislocations are not segregated [2]. Moreover, compared to the results for the electron and proton irradiation, the microstructure of neutron irradiated specimens was found to contain high densities of small defect clusters [2,4]. In this regard it is worth noting that, unlike electron and 3 MeV proton irradiations, neutron irradiation produces a significant fraction of the damage in the form of small vacancy and interstitial clusters [6–10]. Molecular dynamic simulations also show that stable immobile interstitial clusters are directly created in collision cascades in fcc copper, unlike those, for example, in bcc iron [11].

In our previous work [12] we found that at elevated temperatures, continuous production of sessile interstitial clusters limits the homogeneous void concentration N_c to a temperature-dependent maximum value N_{max}. Moreover, if the void concentration approaches $0.4N_{max}$, further evolution of the homogeneous microstructure becomes absolutely unstable, and spatial heterogeneity starts to develop. The calculated values of the critical void concentration and the characteristic scale of the developing heterogeneity are in reasonable agreement with the experiments.

Void nucleation theory [13] has been applied to investigate whether a high enough void number density to satisfy the instability condition is feasible at the observed low dose. In the absence of continuously generated sessile in-cascade interstitial clusters, the calculated void number density is found to be consistent with the experimental observations [13]. However, under the continuous generation of sessile interstitial clusters, which act as recombination centers to suppress void growth [1,12], it is still a valid question to ask whether void nucleation can indeed proceed fast enough to produce the required void number densities at an irradiation dose as low as 10^{-4}–10^{-2} NRT dpa. In the present letter, following the void nucleation theory in our recent work [13], the condition of the development of spatial heterogeneity

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in the microstructure is examined, taking into account the in-cascade generation of sessile interstitial clusters.

According to [13], the void nucleation rate J_c under cascade damage irradiation is given by

$$J_c \cong \frac{G}{N_d} P_m, \tag{1}$$

where G is the actual production rate of point defects in both cluster and free form, and N_d is the average total number of point defects generated in a single cascade. The probability of void nucleation P_m is given by the approximation,

$$P_m \cong \sqrt{\frac{\beta}{6\pi r_{cr} n_{cr}} \frac{(D_v C_v - D_i C_i)}{D_i C_i (1 + d n_{cr}^{1/3})}} (n_0 - n_{min}) \times \exp\left[-\frac{\eta(\beta/r_{cr}) n_{cr}^{2/3} - n_0^{2/3}}{(1 + 1/(d n_{cr}^{1/3}))} \right]. \tag{2}$$

Here D_j and C_j ($j = i, v$) is the point defect diffusion coefficient and local concentration, respectively,

$$d = \frac{3(D_v C_v - D_i C_i)}{2 D_i C_i}, \tag{3}$$

$$\eta(x) = x(1 - x \exp(x) E_1(x)), \tag{4}$$

and $E_1(x)$ is the exponential integral function [14].

In Eq. (2), n_{cr} is the number of vacancies in the void of critical radius r_{cr}. According to [12], the spatially homogeneous evolution of the system of voids and sessile interstitial clusters becomes absolutely unstable at a void number density of $N_c = 0.4 N_{max}$. From Fig. 1 of [12], then, the critical void radius r_{cr} required to produce a void number density of $N_c = 0.4 N_{max}$ is equal to $\beta/6$, noting that $\eta(6) = 0.77$ in [14]. Here $\beta = 2\gamma_s \Omega/kT$, where γ_s is the surface tension, k is the Boltzmann's constant, T is the absolute temperature, Ω is the atomic volume. The unusual decrease with increasing temperature of the critical void radius ($r_{cr} = \gamma_s \Omega/3kT$) at constant surface tension has its origin in the temperature dependence of N_{max}. Indeed, N_{max}, and as a consequence, the total sink strength, decreases exponentially with increasing temperature [12], resulting in the growth of the net vacancy flux $D_v C_v - D_i C_i$ and the reduction of r_{cr}. On the contrary, in [13], where the total sink density is assumed to be constant and temperature independent, r_{cr} increases with increasing temperature, leading to a large reduction of the void nucleating probability with increasing temperature. In this case, the hard-to-justify [4] assumption of a decreasing surface tension with increasing temperature is needed to explain the experimental results [13].

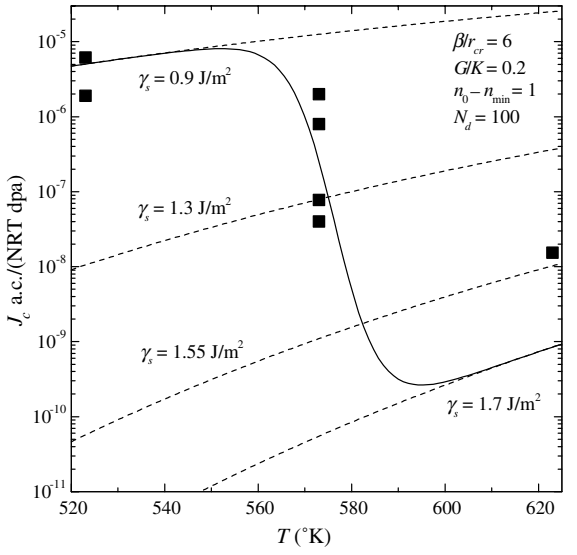

Fig. 1. Rate of void nucleation in annealed copper at different values of void surface energies (dashed lines). Solid line is for the temperature dependent surface tension coefficient $\gamma_s(T)$ obtained in Ref. [4]. Experimental points are obtained from Refs. [4] by dividing the experimental values of void concentration by the corresponding irradiation doses.

In Eq. (2), we denote by n_0 the average initial size of the void embryo on creation in a collision cascade, and by n_{min} the size of the embryo, below which vacancy cluster becomes mobile, i.e., ceases to be an embryo. Since the initial void embryo contains only a few vacancies more than the corresponding minimum value [15], we assume, for simplicity, $n_0 - n_{min} \cong 1$.

The nucleation of voids in a material under irradiation occurs via a stochastic process that can be described in terms of diffusion in the size space of small void nuclei. While shrinking on the average, there is a finite probability for some of the void embryos to reach the critical size due to stochastic fluctuations. The average time t_{nucl} required for a small embryo to reach the critical size by diffusion in the size space can be estimated as $n_{cr}^2/2D(n_{cr})$, where $D(n)$ is the size-dependent diffusion coefficient describing the random walk along the n-axis of void embryo sizes, with $n = 4\pi r_c^3/3\Omega$ being the number of vacancies in an embryo. Using the expression $D(n) = 9 D_v C_s^e(r_c) n^{4/3}/2r_c^2$ for the diffusion coefficient given in [13], we have

$$t_{nucl} \cong \frac{(\beta/6)^4 e^{-6}}{9(3\Omega/4\pi)^{2/3} D_v C_\infty}, \tag{5}$$

where $C_s^e(r_c) = C_\infty \exp(\beta/r_c)$ is the equilibrium concentration of vacancies in the neighborhood of the void of radius r_c. C_∞ is the equilibrium vacancy concentration. Substituting $\beta = 2.5$ nm (at $\gamma_s = 0.9$ J/m^2, $T = 523$ K) and the corresponding values of vacancy formation and migration energies in copper [12] into Eq. (5), $t_{nucl} \cong 9 \times 10^2$ s. Thus, the void nucleation probability calculated from Eq. (2) is indeed applicable to the nominal irradiation doses Kt above 10^{-4} NRT dpa ($K < 10^{-7}$ NRT dpa/s [3–5]).

In the system of voids and sessile interstitial clusters the law of matter conservation requires the number of vacancies in voids to be equal to the number of interstitials in clusters [1,12]. Since the average size of primary clusters is much smaller than that of the growing voids, the sink strength of clusters is much larger than the void sink strength [1,12]. Thus, from the homogeneous balance Eqs. (8) and (9) in [12], the ratio $(D_vC_v - D_iC_i)/D_iC_i$ is, approximately,

$$\frac{D_vC_v - D_iC_i}{D_iC_i} \cong \frac{\varepsilon_i}{(1+\alpha-\varepsilon_i)}, \tag{6}$$

where ε_i is the fraction of clustered interstitials produced in a cascade, $\alpha = n_{i0}/(n_{ig} - n_{i0})$, and n_{ig}, n_{i0} have the same meaning in the case of clusters as n_0 and $n_{\min}$, respectively, in the case of small void embryos. The parameter α takes into account the mobility of the clusters smaller than n_{i0} [12]. Since the majority of interstitial clusters at the moment of creation have sizes of $n_{ig} \approx 5-10$ atoms [8], we have $\alpha \approx 1$.

According to [4], an oxygen concentration on the order of 1 ppm is sufficient to reduce the void surface tension γ_s from a value of 1.7 J/m^2 to below 1.0 J/m^2. The calculations in [4] suggest that at the highest irradiation temperature studied, 623 K, the equilibrium oxygen coverage of void surfaces is too low to significantly reduce the surface energy from its value of 1.7 J/m^2 in the pure metal. However, the value of γ_s decreases rapidly over a narrow range of temperatures between 570 and 580 K [4]. At temperatures below this transition range, the value of γ_s is estimated to be in the range of 0.8–1.0 J/m^2 [4]. Thus, 573 K is just at the low end of the transition, and 523 K is well below the range over which γ_s decreases rapidly.

The void nucleation rate calculated using Eqs. (1), (2), and (6) is shown in Fig. 1 as a function of temperature at different values of the surface tension γ_s. It is easy to see that, taking into account temperature dependence of the surface tension, there is good agreement between the present calculations and the experimental data. The only exception is at 623 K, where a value of γ_s for the pure-metal gives a void nucleation rate about an order of magnitude lower than the experimental one. In this regard, we note the experimental data refer to the average value of void number density in void growth regions in a heterogeneous distribution. This density is obviously higher than the global average, the correct quantity that should be used for comparison, when we are dealing with calculations involving the homogeneous distribution.

It is even more important that the maximum spatially homogeneous void concentration $N_{\max}$ at 623 K is significantly lower than the experimental one in the void growth regions [12]. This means that, unlike at 523 K and 573 K, the instability of the spatially homogeneous solution at 623 K has occurred already before the spatially homogeneous void concentration approaches values comparable to those experimentally observed. Thus, the discrepancy at 623 K may also be accounted for by the significant void nucleation that takes place in the void growth regions after the instability set in, i.e., when the microstructure is already heterogeneous, and the present analysis based on the homogeneous assumption ceases to apply. Indeed, at 623 K a significant void number density (6.7×10^{-11} in atomic units) was experimentally observed only at the largest irradiation dose of 4.4×10^{-3} NRT dpa, when the spatial heterogeneity is already in full development [4].

At 623 K the calculated void concentration $N_{\max}$ is 2.4×10^{-12} (Fig. 2 in [12]). Eqs. (1) and (2) yield a void nucleation rate of 8.5×10^{-10}/(NRT dpa) ($\gamma_s = 1.7$ J/m^2, $G/K = 0.2$), from which the dose at which the void concentration reaches the value of $0.4N_{\max}$, can be calculated, and is given by $\sim 1.1 \times 10^{-3}$ NRT dpa. Similar calculations of the corresponding irradiation dose at 523 and 573 K result in 10^{-3} ($\gamma_s = 0.9$ J/m^2) and 5.5×10^{-4} NRT dpa ($\gamma_s = 1.3$ J/m^2), respectively. Thus, at all three temperatures the irradiation dose for the formation of spatially heterogeneous microstructure, is estimated to be around 10^{-3} NRT dpa. This value is consistent with experimental observations [2–4].

Based on the foregoing, we may conclude that taking into account of the continuous production of immobile primary interstitial clusters achieves better explanation of the experimental observation. Although the effect is significant, it does not generally alter our basic understanding of the development of spatial heterogeneity of microstructure at low irradiation doses (10^{-4}–10^{-2} NRT dpa), as due to the development of instability in the kinetics during the microstructure evolution.

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References

[1] B.N. Singh, A.J.E. Foreman, Philos. Mag. A 66 (1992) 975.
[2] B.N. Singh, M. Eldrup, A. Horsewell, P. Ehrhart, F. Dworchak, Philos. Mag. A 80 (2000) 2629.
[3] B.N. Singh, T. Leffers, A. Horsewell, Philos. Mag. A 53 (1986) 233.
[4] C.A. English, B.L. Eyre, J.W. Muncie, Philos. Mag. A 56 (1987) 453.
[5] B.N. Singh, A. Horsewell, P. Toft, D.J. Edwards, J. Nucl. Mater. 224 (1995) 131.
[6] T. Diaz de la Rubia, M.W. Guinan, J. Nucl. Mater. 174 (1990) 151.

[7] T. Diaz de la Rubia, M.W. Guinan, Phys. Rev. Lett. 66 (1991) 2766.
[8] D.J. Bacon, F. Gao, Yu.N. Osetsky, J. Nucl. Mater. 276 (2000) 1.
[9] C.A. English, W.J. Phythian, A.J.E. Foreman, J. Nucl. Mater. 174 (1990) 135.
[10] A.J.E. Foreman, C.A. English, W.J. Phythian, Philos. Mag. A 66 (1992) 655.
[11] Yu.N. Osetsky, D.J. Bacon, A. Serra, B.N. Singh, S.I. Golubov, J. Nucl. Mater. 276 (2000) 65.
[12] A.A. Semenov, C.H. Woo, Appl. Phys. A 74 (2002) 639.
[13] A.A. Semenov, C.H. Woo, Phys. Rev. B 66 (2002) 024118.
[14] M. Abramowitz, I.A. Stegan (Eds.), Handbook of Mathematical Functions, Dover, New York, 1965, p. 231.
[15] Yu.N. Osetsky, D.J. Bacon, B.N. Singh, J. Nucl. Mater. 307–311 (2002) 866.