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Defect accumulation in fcc and bcc metals and alloys under cascade damage conditions – Towards a generalisation of the production bias model

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Abstract

In recent years, it has been shown that the problem of defect accumulation under cascade damage conditions can be properly addressed within the framework of production bias model (PBM) based on intracascade clustering of point defects, differences in the thermal stability of the resulting clusters and one-dimensional (1-D) diffusion of interstitial clusters. Within this framework, different aspects of defect accumulation such as the high swelling rate at low dislocation densities, enhanced swelling near grain boundaries, effects of grain size, irradiation dose and recoil energy on void swelling have been treated quantitatively. In the present work we have attempted to address the problem of differences in the defect accumulation behaviour between fcc and bcc metals under cascade damage conditions. In this analysis we have chosen copper and molybdenum to represent fcc and bcc metals, respectively. This choice was suggested by the fact (a) that a large amount of experimental information exists on these metals and (b) that the damage accumulation behaviour in copper can be fully accounted for in terms of the PBM. An analysis of the existing experimental observations in terms of the PBM raises the question about differences between the reaction kinetics of the one-dimensionally diffusing interstitial clusters with sinks in fcc and bcc metals. In the present paper, the impact of different frequencies of changes in the 1-D diffusion direction of such clusters on their reaction kinetics is discussed. The present considerations provide an appropriate framework for generalising the PBM in order to describe damage accumulation behaviour in metals and alloys in general, including the formation of void superlattices.

1. Introduction

During the last three decades, a considerable amount of experimental results have been accumulated on defect production and accumulation behaviour in a variety of metals and alloys irradiated under cascade damage conditions. Some of the salient features of these results have been compiled and compared recently by Singh and Evans [1]. The most interesting conclusion emerging from this work was that there are substantial and systematic differences in the defect accumulation both in the form of clusters (loops and stacking fault tetrahedra) and voids between fcc and bcc metals and alloys. These differences are observed in a wide range of irradiation temperatures starting from the recovery stage III and continuing up to temperatures well beyond stage V.

The recognition of these differences makes a very compelling case for a serious investigation into their origin. This, in our opinion, is essential from the point of view of establishing a clear and universal understanding of the processes involved in the evolution of irradiationinduced microstructures under cascade damage conditions. In addition, a proper understanding of these differences may help understand the response of complicated commercial alloys to irradiation in the environment of fission, fusion and spallation neutrons. Since

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no such investigation has been, to our knowledge, reported in the literature, in the following we describe these differences in the swelling behaviour between fcc and bcc metal and alloys and discuss their possible origin.

In order to pursue this objective, we first establish experimental and theoretical basis for the present investigation (in Section 2.1). This is followed by a brief outline of the PBM and its main features in Section 2.2. The saturation of void growth is analysed as an intrinsic property of the PBM in Section 2.3 and possible differences between fcc and bcc crystals are discussed in the light of experimental results in Section 2.4. Section 3 considers the impact of changes in the direction (Burgers vector changes) of one-dimensional (1-D) diffusion of interstitial clusters on their reaction kinetics with sinks, particularly on cluster absorption by dislocations (Section 3.1), on the global cluster reaction kinetics (Section 3.2) and on void lattice formation (Section 3.3). In this context, the question is addressed as to how the PBM could be generalised. A brief summary and main conclusions are presented in Section 4.

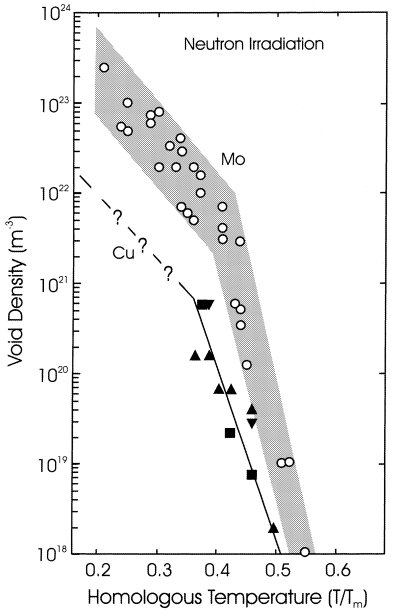

Fig. 1. Temperature dependence of void density in neutron irradiated copper [2–4] and molybdenum [5,6].

2. Damage accumulation in fcc and bcc crystals

In this section, we shall first present experimental results on void density and swelling revealing large differences in the behaviour between fcc and bcc metals irradiated with fission neutrons. The experimental results are then analysed within the framework of the PBM in its present form.

2.1. Experimental and theoretical basis for the present investigation

Since we wish to investigate the differences in swelling behaviour between fcc and bcc metals and alloys, let us first examine the nature and the magnitude of experimentally observed differences. Fig. 1 shows the void density in Cu [2–4] and Mo [5,6] irradiated with fission neutrons as a function of irradiation temperature (expressed by the homologous temperature, T_{irr}/T_m where T_{irr} and T_m are irradiation and melting temperatures, respectively). The temperature dependence of void density in other bcc metals such as Nb and V is found to be similar [7] to that reported in Fig. 1 for Mo. Fig. 1 demonstrates that the void density at any given homologous irradiation temperature is considerably higher in Mo than that in Cu. Equally significant information emerging from these results is that, while in Mo voids are nucleated already at a relatively low temperature of $\sim 0.2T_m$, void nucleation in Cu does not occur until the irradiation temperature reaches a value of $\sim 0.36T_m$ [3]. A similar difference has been reported for other fcc and bcc metals (see Table 2 in Ref. [1]). It is worth noting in this context, on the other hand, that the density of clusters (loops and stacking fault tetrahedra) is drastically lower in bcc than that in fcc metals in the whole temperature range [1].

Fig. 2 shows the temperature dependence of void swelling in neutron irradiated Cu and Mo (see [1] for details). All swelling results quoted in Fig. 2 refer to doses between 0.1 and 1.0 dpa. It should be mentioned here that the change in swelling in neutron irradiated fcc 316 stainless steel to doses in the range of 71–88 dpa

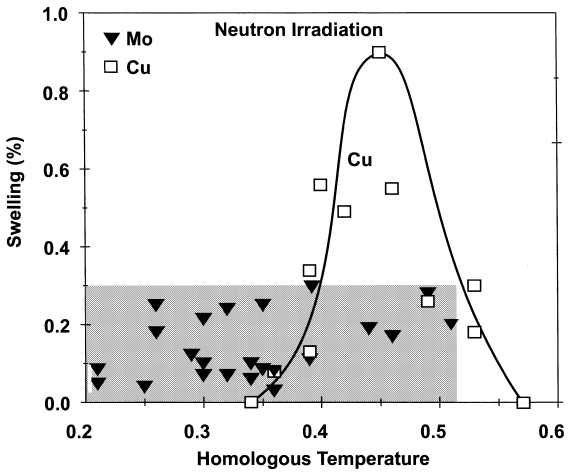

Fig. 2. Temperature dependence of void swelling in neutron irradiated copper and molybdenum (see Ref. [1] for details).

shows a temperature dependence quite similar to the one observed in Cu (Fig. 2) irradiated to rather low doses (0.3–1.0 dpa) [8]. These results demonstrate that the temperature dependence of swelling in fcc crystals is strikingly different from that in bcc crystals. Clearly, the swelling in bcc metals seems to depend only weakly on irradiation temperature and takes place in a wide range of temperatures from $\sim 0.2T_m$ to $\sim 0.5T_m$. In contrast, the swelling rate in fcc crystals appears to increase by a factor of almost 10 within a rather narrow temperature range of $\sim 0.35T_m$ to $\sim 0.45T_m$. It is interesting to note here that in the peak swelling temperature regime the swelling level in bcc metals remains considerably lower than that in the fcc metals even though the void nucleation is significantly more efficient in bcc than that in fcc metals (see Fig. 1). Furthermore, the void swelling in bcc metals seems to saturate already at low doses and at rather low swelling levels (see for example Table 1 in Ref. [5]).

Thus, the results presented in Figs. 1 and 2 raise the following fundamental issues deserving serious theoretical considerations:

- Why does void nucleation in bcc metals occur already in beginning of the recovery stage III whereas in fcc metals void nucleation does not start until around the recovery stage V?
- Even though the void nucleation in bcc metals is very efficient in the whole temperature range ($\sim 0.2T_m$ to $\sim 0.5T_m$) and is strongly temperature dependent, why is void swelling restricted to rather low level and only weakly temperature dependent?

It is worth adding here that the tendency for void lattice formation is clearly stronger in bcc than in fcc metals (see Table 1 in Ref. [9]). This difference, too, calls for an explanation.

By now the phenomenon of intracascade clustering of self-interstitial atoms (SIA) and vacancies both in fcc and bcc metals have been established both by experiments (e.g. [10]) and molecular dynamic (MD) simulations of cascades (see e.g. [11] for a review). Both MD simulations and diffuse X-ray scattering measurements (e.g. [10]) have shown that the clustering of vacancies in bcc iron is considerably less efficient than that in fcc copper. Singh and Evans [1] have argued that this difference may provide at least a qualitative explanation for the nucleation of voids in bcc metals already in the recovery stage III as well as for the fact that the void swelling in bcc metals does not respond to the recovery stage V (see Fig. 2). Recently, MD simulations have shown that all SIA clusters produced in cascades in bcc Fe are likely to be glissile [12]. Removal of these SIA clusters to sinks such as dislocations and grain boundaries would generate a high level of vacancy supersaturation during early stages of irradiation which, in turn, may be responsible for the efficient nucleation of voids in bcc metals in stage III (e.g. Fig. 1). Positron annihilation experiments have demonstrated recently that indeed the nucleation of voids is very efficient in bcc iron neutron irradiated at $\sim 0.21T_m$ (i.e. stage III) already at a dose of ~ 0.22 dpa [13]. In the case of fcc metals, the production of sessile clusters of SIAs and a high efficiency of vacancy clustering in cascades lead to a high density of clusters of SIAs and vacancies. Under these conditions, the level of vacancy supersaturation necessary for efficient void nucleation and growth is likely to occur around the recovery stage V when vacancies are released from the vacancy clusters (loops and tetrahedra). In other words, the differences in the nucleation behaviour between fcc and bcc metals can be rationalised at least qualitatively in terms of differences in the intracascade clustering behaviour of SIAs and vacancies and the properties of interstitial and vacancy clusters. For the time being, we will confine ourselves to this qualitative explanation for differences in void nucleation between fcc and bcc, and will focus in the following on discussing differences in void growth and swelling.

Let us now consider modelling of void swelling in fcc and bcc metals. In recent years, it has been shown that the problem of void swelling under cascade damage conditions can be appropriately treated within the framework of production bias model (PBM) [14–16] including 1-D diffusional transport of SIA clusters [17–19] formed in the cascades. Using this model, the experimental observations of near grain boundary effects [18], effects of grain size [20], irradiation dose [20], and recoil energy [21] on void swelling in fcc metals (e.g. Cu) have been successfully explained in terms of the PMB. In these calculations the values of parameters such as damage production efficiency and fractions of SIAs in sessile and glissile clusters correspond to the values generally obtained in MD simulations of cascades in copper.

In the calculations of void swelling within the framework of PBM, the most important damage parameters are the fractions of SIAs produced in the cascades in the form sessile and glissile clusters. The results of MD simulations of cascades have shown that the values of these fractions of SIAs produced in cascades in iron are not substantially different from that in copper [11,22]. Thus the observed large differences in the swelling behaviour between fcc and bcc metals must be due to other differences, for instance, in the ability of glissile SIA clusters/loops to change their Burgers vectors before getting absorbed by a sink. In fact, it has been emphasised earlier [18,19] that generally, the absorption of a glissile loop by a dislocation requires changes of the Burgers vector of the loop. However, the details of the consequences of such Burgers vector changes for the global reaction kinetics of the loops and the resulting damage accumulation behaviour have not been included in the PBM so far. A detailed discussion

of these consequences is the central objective of the present paper.

The possibility of changes in Burgers vector is supported by a number of MD simulation studies which have shown that at least small clusters in iron containing three crowdions do change their Burgers vector [23–25]. Recently, it has been also shown that the interaction of a glissile cluster with a sessile one in iron made the sessile cluster to change its Burgers vector and become glissile [26].

2.2. PBM and defect accumulation

Since 1990, the problem of defect accumulation has been treated in terms of PBM in a number of publications (e.g. [14–20]). This series of publications illustrates that a quantitative description of the damage accumulation under cascade damage conditions required a number of modifications of the original formulation of the PBM [14,15], particularly in the treatment of the properties and temporal evolution of SIA clusters (see [21] for review). The most comprehensive and quantitative version of the treatment has been recently described by Singh et al. [20]. For pure copper, this treatment yielded results fully consistent with the experimental observations. However, as pointed out in the previous section, even this treatment needs further improvement in order that the PBM can describe the defect accumulation in metals and alloys in general. Before addressing the question of generalising the PBM, it is necessary, in our view, to outline the synthesis and main features of the existing treatment [14–20].

To recapitulate the treatment of the defect accumulation under cascade damage conditions [14–20], let us first consider the balance equations for the concentrations, C_i, C_v, C_g of SIAs, of vacancies and glissile SIA clusters, diffusing with diffusivities D_i, D_v, D_g, respectively, in a crystal containing a (time independent) random distribution of sinks (see Eqs. (20) and (29) in Ref. [20]):

$$(1-\varepsilon_i^{g,\mathrm{eff}})G = D_iC_i(Z_i^vk_v^2 + Z_i^d\rho) + \mu_RD_iC_iC_v + Z_v^{ic}D_vC_vk_{ni}^2 + Z_i^{vc}D_iC_ik_{nv}^2, \quad (1a)$$

$$G = D_vC_v(Z_v^vk_v^2 + Z_v^d\rho) + \mu_RD_iC_iC_v + Z_v^{ic}D_vC_vk_{ni}^2 + Z_i^{vc}D_iC_ik_{nv}^2, \quad (1b)$$

$$\varepsilon_i^{g,\mathrm{eff}}(1-\varepsilon_r)G_{\mathrm{NRT}}/x_g = D_gC_gk_g^2. \quad (1c)$$

Here, $G = (1-\varepsilon_r)G_{NRT}$ is the effective Frenkel pair generation rate obtained from the NRT generation rate by accounting for the fraction of Frenkel pairs, ε_r, recombining during the cooling stage of the cascades. The quantity $\varepsilon_i^{g,\mathrm{eff}} = \varepsilon_i^g + \varepsilon_i^s x_g/\prec x_i^s \succ$ is an effective fraction of the glissile cluster component which consists of a partial fraction produced directly in cascades, ε_i^g, and a partial fraction arising as a result of a transformation of sessile clusters, produced in cascades with fraction ε_i^s, into glissile ones in the presence of a vacancy supersaturation, where x_g and $\prec x_s \succ$ are the sizes of glissile and the mean size of sessile SIA clusters generated in cascades, respectively. $Z_{i,v}^vk_v^2$with $k_v^2 = 4\pi R_vN_v$, and $Z_{i,v}^d\rho$ are the sink strengths of voids and dislocations for point defect absorption, respectively, where N_v is the number density of voids, R_v is their the mean radius and ρ is the dislocation density. (To show the symmetric structure of (Eqs. (1a) and (1b)) for later purposes we have kept here the efficiencies of voids for capturing single SIAs and vacancies, $Z_{i,v}^v \approx 1$.) Further, $Z_v^{ic}k_{ni}^2, Z_i^{vc}k_{nv}^2$ are the sink strengths of sessile SIA clusters (ic) and vacancy clusters (vc) for the absorption of vacancies (v) and SIAs (i), respectively, and k_g^2 is the total sink strength for the one-dimensionally diffusing SIA clusters [20] and is given by

$$k_g = \frac{\pi\rho d_{\mathrm{abs}}}{4} + \sqrt{\frac{2}{l(2R_g - l)}} + \sigma_vN_v, \quad (2)$$

where d_{abs} is the effective diameter of dislocations for absorbing glissile SIA clusters; R_g and l are grain radius and distance from the grain boundary, respectively, and $\sigma_v = \pi R_v^2$.

It should be emphasised that under cascade damage conditions where intracascade clustering ensures a continuous production of sessile SIA clusters (i.e. when $\varepsilon_i^s \neq 0$), the steady state equations (1a)–(1c) are valid only if the fraction of glissile clusters, ε_i^g, is finite. In the case of $\varepsilon_i^g = 0$, $\varepsilon_i^s \neq 0$, the sink strength of the sessile SIA clusters would increase indefinitely. If continued, this process would unavoidably lead to a completely unrealistic high density of SIA clusters (see Fig. 3 in Ref. [16] and Eq. (25a) in Ref. [20]). Note that an analogous unlimited cluster accumulation can take place due to the intracascade clustering of vacancies as well at the irradiation temperature below recovery stage V (BEK model, [27]). In the case of cascade clustering of SIAs, this unlimited cluster accumulation would occur in the whole range of irradiation temperatures of our interest because of their high thermal stability. This means that any model for damage accumulation under cascade damage conditions not only has to consider the intracascade clustering of SIAs but also a mechanism of cluster removal such as the 1-D diffusion of glissile SIA clusters.

Under these conditions, the void swelling rate is determined by the fluxes of point defects and glissile SIA clusters to the voids (the number density of which is assumed to be given) [18,20] and can be described as

$$\frac{dS}{dt} = (D_vC_vZ_v^v - D_iC_iZ_i^v)k_v^2 - D_gC_gx_gk_g\sigma_vN_v. \quad (3)$$

The second term on the right-hand side of Eq. (3) can be taken directly from Eq. (1c) whereas the first one determined by Eqs. (1a) and (1b) is somewhat more complicated. Under pronounced cascade damage conditions, the difference in the point defect fluxes to dislocations (dislocation bias) as well as the recombination and defect cluster terms on the right-hand side of Eqs. (1a) and (1b) represent only small perturbations in the swelling rate described by Eq. (3) when steady state is reached (particularly when cluster production in cascades is restricted to glissile SIA clusters, $\varepsilon_i^s = \varepsilon_v = 0$, $\varepsilon_i^{g,eff} = \varepsilon_i^g$). Neglecting recombination and defect cluster terms in Eqs. (1a) and (1b) we may write Eq. (3) in the simple form

$$\frac{dS}{dt} = G\left[\varepsilon_i^g\left(\frac{Z_v^v k_v^2}{Z_v^v k_v^2 + Z_v^d \rho} - \frac{\sigma_v N_v}{k_g}\right) + (1-\varepsilon_i^g) p_1 \frac{Z_v^v k_v^2 Z_v^d \rho}{(Z_i^v k_v^2 + Z_i^d \rho)(Z_v^v k_v^2 + Z_v^d \rho)}\right], \quad (4a)$$

where

$$p_1 = (Z_i^d/Z_v^d - Z_i^v/Z_v^v) \approx (Z_i^d/Z_v^d - 1) \quad (4b)$$

is the conventional single point defect dislocation bias. In the following, voids will be assumed to represent neutral sinks, $Z_v^v = Z_i^v = 1$.

According to Eq. (4a), the swelling rate is, as expected, proportional to the effective Frenkel pair generation rate $G = (1-\varepsilon_r)G_{NRT}$ and depends via the damage efficiency, $(1-\varepsilon_r)$, on cascade parameters such as recoil energy, atomic mass (density), crystal structure and irradiation temperature. The first term in the square brackets on the right-hand side of Eq. (4a) represents the contribution of the production bias to swelling and the second term that of the conventional single defect dislocation bias. Eq. (4a) shows explicitly that the contribution of the production bias to swelling is determined by the one-dimensionally diffusing SIA clusters ($\propto \varepsilon_i^g$) whereas the one of the dislocation bias ($\propto (1-\varepsilon_i^g)$) is controlled exclusively by the three-dimensional diffusing single point defects. Note that in the limiting case of single Frenkel pair production, where no cascades are available for generating SIA clusters, i.e., $\varepsilon_i = \varepsilon_i^g + \varepsilon_i^s = 0$, the first term in Eq. (4a) vanishes and the PBM transforms into the conventional dislocation bias (SRT) model. For pronounced cascade damage conditions where ε_i is significant and even can reach values close to 1, on the other hand, the conventional dislocation bias contribution is small compared to that of the production bias. It should be noted here, that Eq. (4a) is similar to the corresponding one obtained by Woo and Frank [28–30] while treating the problem of void lattice formation in terms of the crowdion model.

Assuming that the void density N_v remains constant after the void nucleation stage and that the second term in the square brackets of Eq. (4a) representing the conventional dislocation bias may be neglected under cascade damage conditions, an implicit analytical expression describing the dose dependence of swelling can be derived by integrating Eq. (4a) (Eq. (34) in Ref. [20] for $Z_v^d = 1$). For $Z_v^d \neq 1$, the solution may be written as

$$\varepsilon_i^g G t = \left\{\frac{3}{2\alpha}S^{2/3} - \left(\frac{\alpha}{\gamma}+1\right)S - \frac{3\alpha}{4}S^{4/3} - 3\left(\frac{1}{\alpha}+\frac{2}{\gamma}+\frac{\alpha}{\gamma^2}\right) \times \left(\frac{S^{2/3}}{2} + \frac{S^{1/3}}{\gamma} + \frac{\ln(1-\gamma S^{1/3})}{\gamma^2}\right)\right\}, \quad (5a)$$

where

$$\alpha = \frac{(48)^{1/3}(\pi N_v)^{2/3}}{Z_v^d \rho}, \quad \gamma = \left(\frac{3}{4\pi N_v}\right)^{1/3}\frac{Z_v^d \rho}{4k_g^0},$$

$$k_g^0 = \sqrt{\frac{2}{l(2R_g - l)}} + \frac{\pi d_{abs}\rho}{4}. \quad (5b)$$

It has been shown earlier [17–20] that different aspects of defect accumulation in the form of voids such as the high swelling rate at low doses, even with low dislocation densities and particularly near grain boundaries, as well as the saturation of void growth and swelling at higher doses can be treated quantitatively on the basis of Eqs. (4a)–(5b).

2.3. Saturation of void growth

In the following, the saturation of void growth and swelling will be discussed in greater detail. According to Eq. (4a), the swelling rate decreases with increasing size of the voids because of their increasing efficiency for capturing glissile SIA clusters, and eventually vanishes when a maximum void size, R_v^∞, is reached. Also the swelling would saturate at an upper limit, S^∞, if the void density remained constant at the high doses. Neglecting, as in Eqs. (5a) and (5b), the conventional dislocation bias ($p_1 \to 0$), we find from Eqs. (4a) and (4b)

$$R_v^\infty = 4k_g^0/Z_v^d\rho, \quad S^\infty = \frac{4\pi}{3}(R_v^\infty)^3 N_v. \quad (6a)$$

From Eq. (5a), the limitation of swelling follows from the condition that the argument of the logarithm has to be positive. For the interior of large grains, $l \gg \left(k_g^0\right)^{-1}$, Eq. (6a) simplifies to the expression derived in Ref. [17]:

$$R_v^\infty = \pi d_{abs}/Z_v^d, \quad S^\infty = \frac{4\pi}{3}(R_v^\infty)^3 N_v. \quad (6b)$$

According to (Eqs. (6a) and (6b)), R_v^∞ and S^∞ do not depend on the cascade parameters, ε_r, ε_i^g, and R_v^∞ is even independent of the void number density, N_v.

It should be emphasised here that the swelling saturation for random distributions of voids in metals under cascade damage conditions is an intrinsic property of the PBM. Thus, the defect accumulation under these conditions is fundamentally different from that for single

Frenkel pair production (SRT) where the swelling rate does not vanish (see second term in Eq. (4a)). This prediction of the PMB is confirmed by experimental observations of swelling saturation in fcc and bcc metals and alloys under cascade damage conditions (see below).

2.4. Application to swelling saturation in fcc and bcc metals

Swelling saturation is clearly realised in the case of nickel. Fig. 3 shows the dose dependence of swelling of Ni bombarded with self-ions as well as selenium ions at 798 K ($0.41T_m$) [31,32]. Swelling saturates at about 50 dpa where it reaches about 4% (arrows in Fig. 3 mark the doses for swelling saturation and void lattice formation). This value of S^∞ corresponds to $R_v^\infty \approx 13$ nm [31,32]. From the latter value, the effective diameter d_{abs} of dislocations for the absorption of glissile loops may be estimated according to Eq. (6b). Using $Z_v^d \approx 1$, we find $d_{abs} \approx 4$ nm. This value of the absorption diameter is, as expected, clearly smaller than the values of trapping diameters, d_{tr}, which have been estimated to reach several tens of nm [17–19]. For other fcc metals, saturation is not as clearly established as in Ni. In Al, large void radii up to 30 nm and swelling of several % have been found at a few tens of dpa for random void distributions [33,34], and even radii up to 90 nm and swelling up to about 20% in partially ordered void distributions [35,36]. To our knowledge, Cu has been irradiated only up to medium doses somewhat above 1 dpa [2–4] where, as expected for such low doses, neither saturation of void sizes and swelling nor void lattice formation have been observed. Maximum void radii and swelling reached under these conditions are $R_v \approx 20$ nm and $S \approx 8\%$, respectively, which would yield a lower limit for d_{abs} according to Eq. (6b), i.e. $d_{abs} > 6$ nm for $Z_v^d = 1$. It should be noted here that $d_{abs} = 7$ nm was used in Ref. [20].

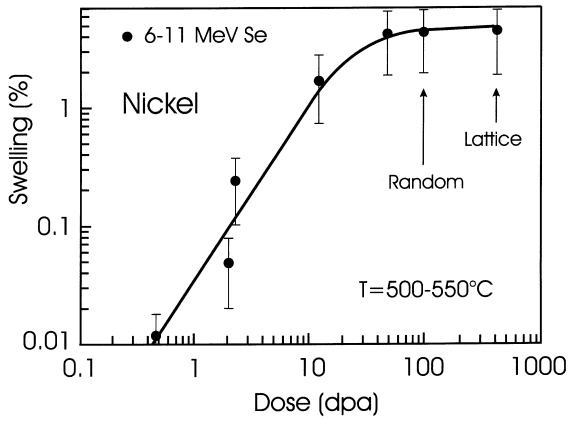

Fig. 3. Dose dependence of void swelling in ion irradiated nickel [31].

In the bcc metal Mo, much lower doses than that in Ni, namely only a few dpa, are sufficient to reach maximum values of void radii and swelling of only about 4 nm and less than 1%, respectively, at temperature of maximum swelling [5,32]. According to Eq. (6b), the maximum void sizes observed would correspond to a value of d_{abs} of only about 1.3 nm. According to this value, small glissile SIA loops would have to come rather close to a dislocation to get absorbed into it. Such small values for d_{abs} have, however, to be considered with care and probably as lower limits since Eq. (6b) is only valid for random void distributions for which saturation of void growth and swelling is not clearly established experimentally. It is interesting that values of d_{abs} obtained by estimating the conditions for Burgers vector changes of small loops in the elastic field of a dislocation are of similar magnitude with the same difference between fcc and bcc as given above, namely 6 nm for fcc copper and 3 nm for bcc iron [37].

It is worth noting here that in the values for swelling, the much larger void densities in bcc as compared to fcc metals are not sufficient to compensate the effect of the smaller void sizes which enter swelling in the third power. The relatively weak temperature dependence of swelling, $S(T)$, is due to a partial compensation of the strong temperature dependence of the void density, $N_v(T)$, by that of the void volume, $\propto R_v(T)^3$. This would imply that the absorption diameter, d_{abs}, increases with temperature.

There are perhaps other reasons for differences between fcc and bcc, but the present interpretation of swelling saturation raises the question about the mechanisms controlling the absorption of glissile SIA loops by dislocations, the value of the corresponding absorption diameter, d_{abs}, and its possible dependencies on crystal structure and temperature. It has been already recognised in Refs. [18,19] that changes in the 1-D diffusion direction of glissile SIA clusters (Burgers vector changes) are generally required for their absorption by dislocations. On the other hand, it has been tacitly assumed in this earlier work that such direction changes would not affect the global reaction kinetics of SIA clusters with sinks.

For small glissile SIA clusters, Burgers vector changes may be assumed to occur by thermal activation. For larger clusters changes in Burgers vectors may happen as a result of the elastic interaction with another defect [26], particularly with a dislocation. In the following we will consider both the role of (thermally activated) Burgers vector changes of SIA clusters in their absorption by dislocations and their global reaction kinetics, including its modifications in void lattice formation.

3. Impact of changes in the direction of 1-D diffusing SIA clusters

3.1. Impact on cluster absorption

As has been argued earlier [17–19], an SIA cluster may be considered to be trapped by a dislocation when the magnitude of their mutual attractive elastic interaction energy becomes larger than the thermal energy kT. This condition may be used to define a trapping diameter $d_{tr} \propto x_g/T$ where x_g is the number of SIAs in the cluster. Differences in x_g for fcc and bcc would be associated with differences in d_{tr} which are, however, not relevant for the reaction kinetics as formulated in the PBM.

The fate of a glissile SIA loop once trapped in the stress field of a dislocation has been discussed in connection with the problem of dislocation decoration with loops [38]. A trapped loop would be absorbed by the dislocation only after performing Burgers vector changes (or substantial conservative climb) before it would be detrapped by thermal activation. Thus, the likelihood for the absorption of a trapped loop is determined by the competition between Burgers vector change and detrapping. Accordingly, the absorption diameter may be defined as the maximum distance where a Burgers vector change of a loop is at least as likely to occur as its detrapping (implying $d_{abs} \leqslant d_{tr}$). On this basis, it can be expected that d_{abs} decreases, relative to d_{tr}, with increasing ability of loops to change their Burgers vectors which decreases with increasing loop size. The latter dependence is most likely to be stronger than the dependence of d_{tr} on loop size. Considering these trends and our above interpretation of void growth saturation (with due reservations) to be relevant for differences between fcc and bcc, the lower values of d_{abs} for bcc than for fcc, as deduced from maximum void sizes according to Eq. (6b), would indicate that, for given loop sizes, Burgers vector changes would be more difficult in bcc than in fcc.

The increase of d_{abs} with temperature may be qualitatively explained in the following way: Because of the correlation between the probabilities for reactions with two partners on the track of a 1-D diffusing defect, the likelihood for the detrapping of a glissile loop from the stress field of a dislocation decreases with increasing distance from other traps [38] and, correspondingly, with temperature. In turn, the chance for a Burgers vector change at a given distance from the dislocation, and correspondingly the absorption diameter increase with temperature. A quantitative treatment of these relationships is beyond the scope of the present paper.

3.2. Impact on the global cluster reaction kinetics

It is useful in the present context to briefly review the main ideas in earlier work on 1-D diffusion-reaction kinetics. The concept of 1-D atomic transport in three-dimensional crystals was introduced already in 1950 [39] by Paneth who has shown that SIAs in the 'crowdion' configuration can migrate only one-dimensionally along close packed rows of atoms. A first attempt to account for the consequences of the 1-D transport on defect recovery kinetics was made by Lomer and Cottrell [40] but a detailed treatment of the problem of 1-D diffusion and reaction in the three-dimensional space of a real crystal was formulated by Seeger and co-workers [41,42]. They found that under defect annealing conditions, the so-called 'pure 1-D diffusion' of crowdions would lead to a reaction constant decreasing with time and to a varying reaction order. Furthermore, it was shown that crowdion–crowdion reactions have to be treated in terms of 2-D rather than 1-D diffusion. In addition, it was demonstrated that the reaction kinetics for preferentially 1-D diffusion, when the tensor of the diffusion coefficient of diffusing species has at least two non-zero components with a small ratio of them, reflects features of 1-D diffusion as well as of 3-D diffusion. It was shown (see [43]) that even when the diffusion component perpendicular to the principal diffusion direction is by orders of magnitude smaller, a change in the reaction kinetics is likely to occur. This particular conclusion is of crucial importance in the present context since in the treatment of SIA clusters in the PBM (Section 2.2) it has been tacitly assumed that direction changes in the 1-D diffusion of SIA clusters do not seriously affect their reaction kinetics. This assumption may be correct in some cases but cannot be expected to be justified in general.

A general and rigorous treatment of the reaction kinetics of SIA clusters changing their 1-D diffusion direction is a complicated problem. Formally, this is due to the fact that a finite mean lifetime of a specific defect configuration associated with a certain 1-D diffusion direction, τ_1, introduces, in addition to the static length scales of the microstructure, a dynamical length scale into the reaction kinetics: the mean 1-D diffusion length, l_1, limited by Burgers vector changes

$$l_1 = \sqrt{2D_g\tau_1}. \tag{7}$$

In the case of crowdions, which are considered to be thermally unstable, the lifetime of a configuration is limited by its thermally activated conversion into the more stable dumb-bell configuration. On the other hand, even small SIA clusters have been shown by MD simulations (see, e.g. [11]), to be thermally stable. In this case, the lifetime of a specific configuration is limited by a change into another equivalent configuration with another Burgers vector.

We first consider the role of Burgers vector changes for the cluster reaction kinetics in a random distribution of sinks. The relation of the mean 1-D diffusion length of glissile SIA clusters to the length scales of the micro-

structure determines the character of the reaction kinetics. The latter are the sizes of the microstructural components, i.e. the sizes of grains and voids, R_g and R_v, respectively, together with the absorption diameter of dislocations, d_{abs}, on one hand, and the mean free diffusion ranges in the microstructure, on the other hand. Without Burgers vector changes, the mean free paths of glissile SIA clusters performing 1-D diffusion through a microstructure containing voids and dislocations, respectively, are given by Refs. [17,18]

$$\lambda_{1v} = 1/\pi R_v^2 N_v, \quad \lambda_{1d} = 4/\pi d_{abs}\rho,$$
$$\lambda_1 = (\lambda_{1v}^{-1} + \lambda_{1d}^{-1})^{-1} = k_g^{-1}. \tag{8}$$

The mean ranges for SIA clusters diffusing in 3-D may be expressed as

$$\lambda_{3v} = (Z_{cl}^v k_v^2)^{-1/2}, \quad \lambda_{3d} = (Z_{cl}^d \rho)^{-1/2},$$
$$\lambda_3 = (\lambda_{3v}^{-2} + \lambda_{3d}^{-2})^{-1/2}, \tag{9}$$

where Z_{cl}^v and Z_{cl}^d are the efficiencies of voids and dislocations for capturing SIA clusters, respectively.

In order to illustrate the differences between these parameters, Eqs. (8) and (9), the data of Ref. [5] together with Eq. (6b) for d_{abs}, have been used to calculate their values for neutron irradiated Mo as a function of irradiation temperature as shown in Fig. 4. The striking feature revealed by Fig. 4 is that the mean free paths for 1-D diffusion are significantly larger than the ranges for 3-D diffusion, and that the partial ranges for the dislocation component is larger than those for the void component.

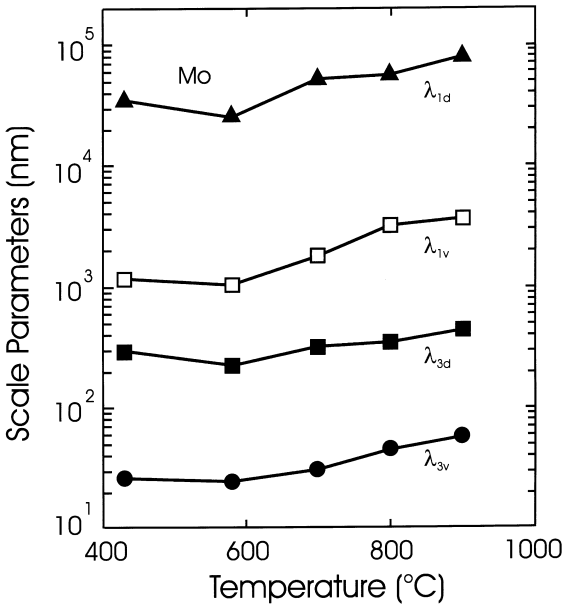

Fig. 4. Temperature dependence of the average mean free paths, λ, for glissile SIA clusters to reach dislocations (d) and voids (v) for 1-D and 3-D diffusion calculated (Eqs. (8) and (9)) using the measured dislocation and void densities in neutron-irradiated molybdenum [5] and the $d_{abs} = 3$ nm [37].

The characteristic types of the reaction kinetics may be classified by setting the mean 1-D diffusion length, l_1, in relation to the microstructural length scales as done in Table 1. We first consider random distributions of sinks where three different cases may be distinguished. The first is characterised by $l_1 \gg \lambda_1$. In this case, the chance of a cluster for changing its Burgers vector before getting absorbed by one of the two sinks on its track is negligibly small. Thus, Burgers vector changes do not play any role in the cluster reaction kinetics which may therefore be considered to be of 1-D type and accordingly of third order because of the diffusional correlation between sinks [18]. The PBM as formulated in Ref. [20] is strictly valid only for this case.

The second case, $\lambda_1 \gg l_1 \gg R_v$, d_{abs}, is less clear. In this case, the 1-D diffusional correlation between sinks is obviously broken. Due to its Burgers vector changes, a glissile loop is now inspecting a larger space for getting absorbed by a sink than without Burgers vector changes, meaning that the sink strength for its absorption is increased. However, the cluster reaction kinetics still keeps features of the 1-D kinetics. Thus, simple scaling arguments show that the partitioning of clusters over fixed sinks (where common factors describing an increase of the sink strength cancel) must still be proportional to their relative cross sections weighted by their number densities as in the second term in the large curved brackets of Eq. (4a). This would suggest that, in the present case, Eq. (4a) need not be modified substantially. It should be noted, however, that in swelling of Ni and Mo considered above, both terms in the large curved brackets of Eq. (4a) are very close to unit when swelling achieves saturation since the sink strength of dislocations is much smaller than that for the voids, for point defects as well as for SIA clusters. This means that even small variations in the relative sink strengths of voids and dislocations could change the magnitude of saturated swelling which calls for being cautious in drawing conclusions from simple arguments. Rigorous results for this case are presently not available.

The third case in its extreme form, R_v, $d_{abs} \gg l_1$, is again clear. In this case, a mobile cluster in the vicinity of a sink (void or dislocation) is able to inspect many sites in all possible directions of Burgers vectors before it escapes or gets absorbed by the sink. The diffusion and reaction kinetics of such clusters are effectively of 3-D type. It should be noted here that an intermediate case $R_v \gg l_1 \gg d_{abs}$, is conceivable where the cluster reactions with voids are clearly of 3-D type while those with dislocations are still between 1-D and 3-D type (mixed 1-D and 3-D).

Table1
Length scale relations and types of reaction kinetics

Type of void distribution	Length scale relation	Type of reaction kinetics
Random	(1) $l_1 \gg \lambda_1$	1-D, correlation between sinks, sink strength according to Eqs. (4a) and (4b)
	(2) $\lambda_1 \gg l_1 \gg R_v$ & d_{abs}	Mixed 1-D and 3-D, loss of correlation between sinks, sink strength larger than in (1) partitioning similar as in (1)
	(3) R_v & $d_{abs} \gg l_1$	3-D, sink strength larger than in (2) according to Eqs. (10a) and (10b)
Ordered	(4) $l_1 \gg R_g$ or λ_d	1-D, periodic sink strength, continued swelling
	(5) R_g & $\lambda_d \gg l_1 > N_v^{-1/3}$	Mixed 1-D and 3-D, periodic sink strength, saturated swelling

When both single SIAs and SIA clusters perform 3-D diffusion, the swelling rate follows an expression analogous to the conventional single defect dislocation bias, including now, however, an SIA cluster term analogous to that for single SIAs and can be written as

$$\frac{dS}{dt} = (1-\varepsilon_r) G_{NRT} \frac{Z_v^v k_v^2 Z_v^d \rho}{(Z_v^v k_v^2 + Z_v^d \rho)} \times \left[\frac{\varepsilon_i^g p_{cl}}{(Z_{cl}^v k_v^2 + Z_{cl}^d \rho)} + \frac{(1-\varepsilon_i^g) p_1}{(Z_i^v k_v^2 + Z_i^d \rho)} \right], \quad (10a)$$

where

$$p_{cl} = (Z_{cl}^d / Z_v^d - Z_{cl}^v / Z_v^v) \quad (10b)$$

is the bias factor for SIA clusters. Now, it is no longer obvious that (small) voids may be considered as neutral sinks for capturing SIA clusters, $Z_{cl}^v > Z_{i,v}^v \approx 1$.

According to Eq. (10a), the swelling rate for 3-D cluster reaction kinetics is positive for any microstructural situation, as in the conventional single defect dislocation bias, meaning that saturation in swelling cannot occur in this case. But now the magnitude of the swelling rate is clearly higher than in the latter case since the elastic interaction of clusters with dislocations is stronger (depending on cluster size) than that of single SIA, i.e. $p_{cl} > p_1$. The quantity responsible for the large magnitude of p_{cl} is the efficiency of dislocations for absorbing clusters, Z_{cl}^d. Values of this quantity may be directly deduced from estimated values of the trapping diameter, d_{tr} [17–19] since $d_{tr} \approx d_{abs}$ for 3-D diffusion of clusters. Because of the dependence of the cluster bias on cluster size the magnitude of the swelling rate is expected to depend on cascade parameters such as recoil energy and atomic mass (density). Large values of the cluster bias are probably responsible for the high swelling rates observed in some alloys such as stainless steels (e.g. [44]).

Before concluding this subsection, we emphasise that the type of cluster reaction kinetics for a random distribution of both dislocations and voids does not depend on the partial 1-D mean free paths, l_{1v} and l_{1d}, separately but only on the global 1-D mean free path, l_1, and not at all on the ranges for 3-D diffusion, l_3. Between the three characteristic regions and cases listed in Table 1 for random sink distributions, there are, of course, transitional regions which are even less clear than the former. In spite of arguments against serious changes in the partitioning of SIA clusters over sinks as long as the diffusion of the clusters does not change to 3-D behaviour, there remains an uncertainty whether these arguments are really valid and future work is therefore needed for definitive conclusions.

3.3. Impact on void lattice formation

Since the discovery of void lattice formation in metals under irradiation [45,46], the origin of this phenomenon has attracted the interest of theoreticians [9,28,47–50]. The most spectacular feature of void lattice formation is the isomorphy of the void lattice with the underlying crystal structure (each of both of fcc , bcc or hcp). Independent of differences in the description of void lattice formation, there seems to be agreement now [28,47,49,50] that this close structural relationship originates in the one-dimensional motion of some interstitial type defect (single or clustered) along close packed directions and the associated mutual screening of correspondingly arranged voids in absorbing such defects, as originally suggested by Foreman [47]. There are other interesting observations which need to be rationalised, too. Why are void lattices easily formed under neutron and ion irradiation but hardly under electron irradiation? Why is void lattice formation apparently easier in bcc than in fcc ? What are the conditions for saturation of void growth and swelling of a once formed void lattice, or, in turn, under which conditions is void growth and swelling possible in a void lattice?

In attributing void lattice formation to the 1-D motion of SIA type defect, Foreman [47] and the theoreticians working out this idea in detail [28,50] have considered single SIAs in the crowdion configuration to move along close packed directions. For this type of defect, the (thermal) instability, i.e. its conversion into the more stable dumbbell configuration is a limiting factor which has to be taken into account in the theory. Glissile clusters, on the other hand, are thermally stable and can only change their direction of motion by changing their Burgers vector. The occurrence of void lattice formation under cascade damage conditions and

its absence under single Frenkel pair production as in electron irradiation provide strong evidence for the key role of the 1-D motion of thermally stable SIA clusters directly produced in cascades.

In the following, we continue our discussion of the role of Burgers vector changes of glissile SIA clusters for the cluster reaction kinetics, but now under the changed conditions of void lattice formation. For clusters gliding in a certain direction, the sink strength is characterised by a 2-D periodicity corresponding to the projection of the void lattice on a plane normal to the glide direction. The sink strength is high within the close packed rows of voids, where it is essentially controlled by the voids, and it is low in the void depleted 'channels' in between the rows of voids where it is mainly controlled by dislocations and grain boundaries crossing the channels. The relation of the mean 1-D free path in these channels to the mean diffusion length between Burgers vector changes is crucial for the occurrence or absence of void growth and swelling within a void lattice (see Table 1). In discussing this, we assume that the distribution of dislocations remains random during void lattice formation.

If at least one of the two length scales, R_g or λ_d, is much smaller than l_1, the cluster reaction kinetics is effectively of 1-D type. In this case, an SIA cluster gliding in a void depleted channel is most likely to get absorbed by a sink interrupting the channel (dislocation or grain boundary) before having got a chance for changing its Burgers vector. The vacancies corresponding to the SIAs in the cluster will be absorbed in the rows of voids adjacent to the channel. This would result in continuous void growth and swelling. Thus, the apparently continuous void growth in (partially ordered) void 'hyperlattices' formed in Al under neutron irradiation [35,36] is most probably due to the absorption of glissile clusters by dislocations crossing void depleted channels.

If, on the other hand, both length scales, R_g and λ_d, are much larger than l_1, even a glissile cluster originally gliding in a void depleted channel will most probably leave this channel (and later possibly other channels) by a Burgers vector change and find a void in the void lattice where it will get absorbed. Consequently, void growth and swelling will be negligible. The alignment of randomly distributed voids by glissile clusters during void lattice formation requires, however, that the mean 1-D diffusion length between Burgers vector changes is at least of the order of the mean void distance. Accordingly, the relation $R_g \,\&\, \lambda_d \gg l_1 > N_v^{-1/3}$ represents the necessary and sufficient condition for the formation of a swelling saturated void lattice (to make the condition safely sufficient, the last inequality sign > may be substituted by $\gg$). The simplest explanation for the observation that void lattice formation is apparently easier in bcc than in fcc may be the experimental findings that void densities are larger and, correspondingly, void distances are smaller in bcc than in fcc. This, of course, shifts the problem only to the difference in void nucleation which is not yet fully understood. Accepting this difference, for the time being, as an experimental fact, we may say that both the conditions for the formation and the stability of void lattices can be rationalised in terms of the PBM provided Burgers vector changes are properly taken into account.

4. Summary and conclusions

In the present paper, the development of the PBM based on intracascade clustering of point defects and differences in the thermal stability of the resulting clusters is reviewed. An important step in this development was the inclusion of glissile SIA clusters diffusing, like crowdions, one-dimensionally along close packed directions of the crystal lattice. Key input parameters of the model are those which characterise the mode of defect production depending on the type of projectile particle, its energy, the mass of the target material and its crystal structure (fcc or bcc). The limiting cases are the production of single Frenkel pairs under electron irradiation and defect production in displacement cascades occurring under (heavy) ion and neutron irradiations. One of the differences between fcc and bcc metals is, for instance, the larger clustering efficiency in the former than in the latter case. Taking the defect production characteristics properly into account, the PBM in its present form incorporates previous models such as the standard rate theory model, particularly the conventional dislocation bias model, for the case of single Frenkel pair production, as well as models taking into account only vacancy clustering in cascades (BEK model). The present picture of the processes participating in the damage accumulation during irradiation is schematically illustrated in Fig. 5.

The production of one-dimensionally diffusing SIA clusters in cascades results in enhanced swelling near grain boundaries, grain size effects in swelling, saturation of void growth and swelling as well as void lattice formation. An analysis of the void growth limitation and its application to void swelling in fcc and bcc metals indicates that differences between fcc and bcc are most likely due to differences in the ability of glissile SIA loops to change their Burgers vector. This property of SIA cluster deserves, therefore, a careful examination. The following aspect have been discussed in the present paper:

1. The absorption of a glissile SIA loop by a dislocation requires Burgers vector changes of the loop. The effective diameter of a dislocation for absorbing such a cluster increases with the ability of the cluster to change its Burgers vector.

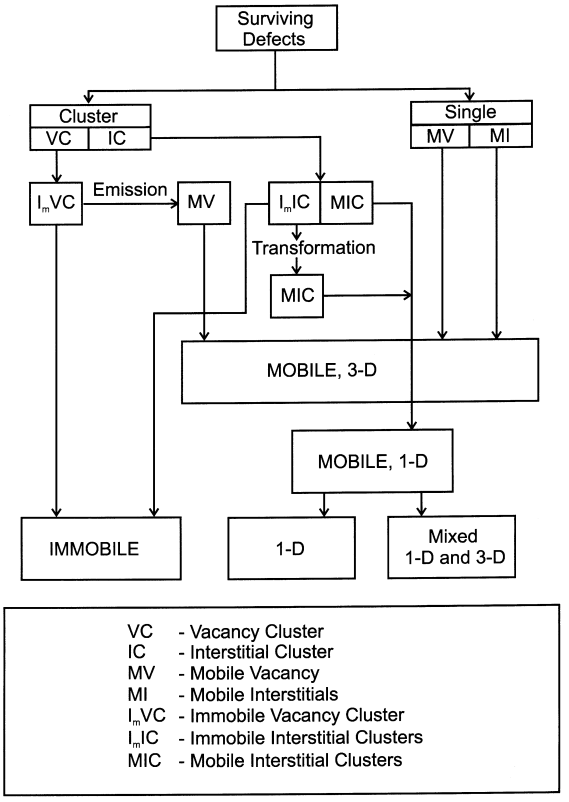

Fig. 5. Schematic illustration of various fractions of surviving defects under conditions of Frenkel pairs (i.e. single defects) production and displacement cascades production where both single defects and defect clusters are produced. Note that the one-dimensionally diffusing SIA clusters may maintain their direction (pure 1-D) or may change their Burgers vector during their diffusion motion (mixed 1-D and 3-D).

2. The relation of the mean 1-D diffusion length of SIA clusters between Burgers vector changes to the mean free path for 1-D cluster diffusion through the microstructure determines the character of the global cluster reaction kinetics. This ranges from effectively 1-D (for rare Burgers vector changes) over an intermediate kinetics between 1-D and 3-D (mixed kinetics) to effectively 3-D type (for very frequent Burgers vector changes). In the latter case, a cluster dislocation bias analogous to the conventional single defect dislocation bias may be defined for random distributions of voids.
3. Void ordering is due to the 1-D diffusion of SIA clusters. Burgers vector changes of these clusters are essential for maintaining a constant void size in a void lattice. The control parameter for this is the ratio of the mean 1-D diffusion length of SIA clusters between Burgers vector changes to their mean 1-D free path in void depleted channels. For low values of this ratio (mixed 1-D and 3-D kinetics) void growth ceases whereas it continues for large values (effectively 1-D kinetics).

Presently, it seems that, for given dislocation and void densities, the PBM is able to rationalise the whole variety of experimental observations in cascade damage accumulation if Burgers vector changes of one-dimensionally diffusing SIA clusters are properly taken into account, in local absorption cross sections as well as in the global reaction kinetics. In the case of random distributions of sinks (dislocations and voids), for which a rigorous treatment of the mixed 1-D and 3-D kinetics is in preparation, the treatment of defect accumulation by the recent version of the PBM does not require substantial revision and generalisation as long as the cluster reaction kinetics does not become effectively of 3-D type. It should be pointed out, however, that in order to treat the formation and evolution of void lattices, details of reaction kinetics used in the PBM would have to be modified. Since the frequency of Burgers vector changes of SIA clusters in the form of small dislocation loops plays a key role in the present context future MD studies should focus on this SIA cluster property.

Finally, we emphasise that a full understanding of the damage accumulation under cascade damage conditions would have to include proper treatments of void nucleation and dislocation evolution which are still not available.

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