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Kinetic Monte Carlo studies of the reaction kinetics of crystal defects that diffuse one-dimensionally with occasional transverse migration

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Abstract

The reaction kinetics of the various species of mobile defects in irradiated materials are crucially dependent on the dimensionality of their migration. Sink strengths for one-dimensionally (1D) gliding interstitial loops undergoing occasional direction changes have been described analytically and confirmed by kinetic Monte Carlo (KMC) simulations. Here we report on KMC simulations investigating a different transition from 1D to 3D diffusion of 1D gliding loops for which their 1D migration is interrupted by occasional 2D migration due to conservative climb by dislocation core diffusion within a plane transverse to their 1D glide direction. Their transition from 1D to 3D kinetics is significantly different from that due to direction changes. The KMC results are compared to an analytical description of this diffusion mode in the form of a master curve relating the 1D normalized sink strength to the frequency of disturbance of 1D migration.

1. Introduction

In materials undergoing irradiation-induced displacement damage the evolution of the microstructure is essentially determined by the diffusion reaction kinetics of the various species of mobile defects that are produced or exist in the material. The reaction kinetics depends crucially on the dimensionality of the diffusion processes of the mobile defects involved. Indeed, there are experimental observations of features in irradiated materials that cannot be rationalized in terms of the 3D diffusion kinetics of vacancy and self interstitial atom (SIA) defects assumed in conventional rate theory. These involve highly heterogeneous and segregated defect accumulation, i.e., large (μm) scale separation of SIA clusters and voids, under cascade damage conditions. Such observed features include, for example, decoration of dislocations with SIA loops, rafts of loops, enhanced swelling near grain boundaries, saturation of void growth and void lattice formation [1,2]. Moreover, it has been recognized that some deviation of the reaction kinetics of SIA clusters from that for pure 1D diffusion are necessary for proper representation of phenomena described by the theory of radiation damage [3].

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Computer modeling has shown that energetic displacement cascades in metals produce clusters of self-interstitial crowdion defects (actually small glissile dislocation loops) that diffuse by random one-dimensional (1D) glide along close-packed directions with migration energies less than 0.1 eV [4,5]. Under certain conditions, the 1D migrating loops can individually undergo a thermally-activated change in the direction of their 1D migration (i.e., a change in the direction of the Burgers vector of the dislocation loop) to a different close-packed direction [6,7]. Direction changes can also be activated by interaction with other microstructural features [8]. In general, such defects diffuse through the material in all three dimensions, but their total migration path consists of a series of the different 1D migration paths they take during the time between direction changes. The frequency of their direction changes influences their probability of interacting with other features of the microstructure. As the frequency of their direction changes increases, the lengths of the one-dimensional path segments of these defects become smaller. In the absence of reaction partners, the limiting cases of 1D path segment length as a function of the frequency of defect direction changes are: (a) effectively 'infinite' for pure 1D migration, i.e. when all migration is along the same close-packed row of atoms, and (b) one atomic spacing for purely random three-dimensional (3D) migration, i.e., when the defect can 'hop' with equal probability in any direction. The dimensionality of migrating defects in real materials is most likely to lie somewhere between pure 3D and pure 1D. Thus, it is important to develop an analytical representation of defect reaction kinetics for defects migrating with some sort of '1D/3D' dimensionality within the range from pure 1D to pure 3D.

An analytical treatment of the reaction kinetics for 1D migrating defects disturbed by direction changes was developed for use in reaction rate theories of microstructure evolution under irradiation [9]. It describes the effects of the direction change of 1D migrating defects as a function of frequency, but it does not include the limiting case of pure 3D migration. Also, atomic-scale kinetic Monte Carlo (KMC) simulations were performed under a variety of specific conditions to investigate the 1D to 3D transition [10]. Subsequently, a 'master curve' was developed that describes the continuous transition of the sink strength as a function of the dimensionality of defect migration, from pure 1D to pure 3D as the frequency of Burgers vector direction change is increased [11]. As part of that work, KMC simulations of the 1D to 3D transition for defects interacting with a variety of sizes and densities of spherical sinks were also performed, and the results were perfectly described by the master curve [11].

In the present work, we examine a different type of 1D/3D reaction kinetics in which the random 1D migration of gliding dislocation loops is occasionally interrupted by excursions due to conservative

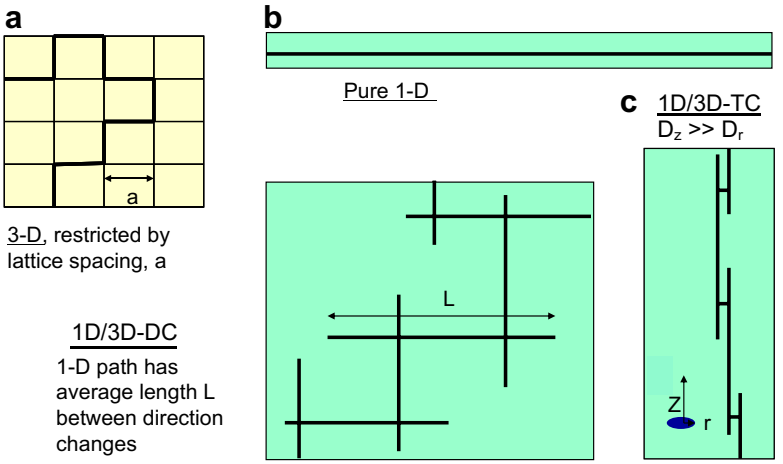

Fig. 1. Schematic illustration of defect migration as random walks on a 3D lattice: (a) 'Pure 3D' consisting of a 3D random walk of equally probable hops to any nearest neighbor site. (b) 'Pure 1D' consisting of a 1D random walk of equally probable hops along the same close-packed direction, as by gliding crowdion defects. (c) '1D/3D-DC' consisting of a series of segments of 1D random walks in different random directions, as by crowdion clusters gliding along close-packed directions that occasionally change their glide direction. (d) '1D/3D-TC' consisting of a series of segments of 1D random walks, all along the same direction, occasionally shifted transversely in two dimensions such as gliding interstitial loops that occasionally migrate by conservative climb.

climb by dislocation core diffusion. Each 2D climb excursion is in a plane normal to the direction of glide, after which glide is resumed along the original glide direction. This mode of defect migration was considered some time ago, especially in the case where the climb excursions are small and infrequent, where it was referred to as 'preferential 1D migration', [12]. Fig. 1 illustrates schematically the nature of 1D migration interrupted both by direction changes (1D/3D-DC) and by transverse climb excursions (1D/3D-TC). An analytic theory similar to that for 1D/3D-DC reaction kinetics has been developed for 1D/3D-TC that describes the effects of the frequency of climb excursions on sink strengths throughout the range from pure 1D to pure 3D. The development of this theory will be described in detail elsewhere [13]. In the present paper, we report on KMC simulations performed in support and corroboration of the analytical theory.

2. Sink strength

In reaction rate theories for microstructure evolution under irradiation, the interactions of migrating defects with the various microstructural features are represented by the 'sink strength' k^2 of each type of sink. The contribution of a certain sink type to the total sink strength is a function of the sink type, size, shape, and concentration (sink density). However, the sink strength is proportional to the inverse of the migrating defect's mean free path, so the dimensionality of the defect migration (1D, 2D, 3D) also has a major influence on the effectiveness of sinks, since it strongly affects the probability of a migrating defect encountering a sink.

The effect of dimensionality of defect migration on the sink strengths is dramatically manifested in the dependence of the sink strengths on the size and number of sinks. Consider the simple example of small migrating interstitial clusters interacting with spherical voids. For voids with radius R and number density N the sink strength k_3^2 for interactions with interstitial clusters diffusing by pure 3D migration is proportional to $4\pi RN$ (for relatively small R and N). On the other hand, if the clusters in the same system are migrating by pure 1D, the sink strength k_1^2 is proportional to $6\pi^2R^4N^2$ [2]. Thus, the dependence of the sink strength on void size R and number density N is qualitatively different in the two cases, and the difference in sink strengths results solely from the difference in dimensionality of the migrating interstitial loops. The nature of the transition from pure 1D to pure 3D kinetics depends on the type of disruption of 1D motion that is involved, i.e., whether pure 1D gliding of the defect is interrupted by a change of glide direction (1D/3D-DC) or by a transverse climb excursion (1D/3D-TC).

3. Kinetic Monte Carlo simulations

We have used KMC modeling to explore the phenomena associated with the variation in dimensionality of migrating defects as they interact with sinks, and to corroborate predictions of the analytical theory. These KMC studies are not intended to mimic reality, but rather to isolate and study the effects of the dimensionality of defect migration in idealized situations.

The KMC model consists of immobile spherical 'absorbers' that are unsaturable, maintaining a constant radius R and number density N throughout the simulations. The absorbers are randomly arranged in a large test volume, and migrating 'defects' are introduced into the volume one at a time, starting at random positions near the center of the volume and hopping on a face-centered cubic lattice until being absorbed by an absorber. All absorbers and migrating defects are centered on lattice sites. During the migration of each defect (a one-dimensional random walk along a close-packed direction) its deviations from pure 1D hopping occur regularly according to the conditions under investigation, e.g., it makes one 2D transverse climb excursion after every fifty 1D hops. All hops of a defect are chosen randomly from the possible hops permitted by the restrictions on its motion. The 'lifetime' of each defect is defined as the total number of hops it makes before being absorbed by one of the absorbers. The value of the sink strength for defects that migrate under the same specific hopping conditions is determined from the average lifetime of many such defects. Each data point in the KMC results reported in Figs. 2 and 3 represent the average behavior of at least 1000 migrating defects.

In all of the simulations the spherical absorbers each have a radius of 5.4 nm, and they are distributed randomly in a KMC test cell of volume $V = 10^{-18}\,\mathrm{m}^3$, with a number density of $N = 10^{22}\,\mathrm{m}^{-3}$. The test cell is a cube for the 1D/3D-DC simulations and a rectangular parallelepiped along the 1D diffusion direction for the 1D/3D-TC simulations. The sink strength is plotted on a log–log graph as a function of the average distance between

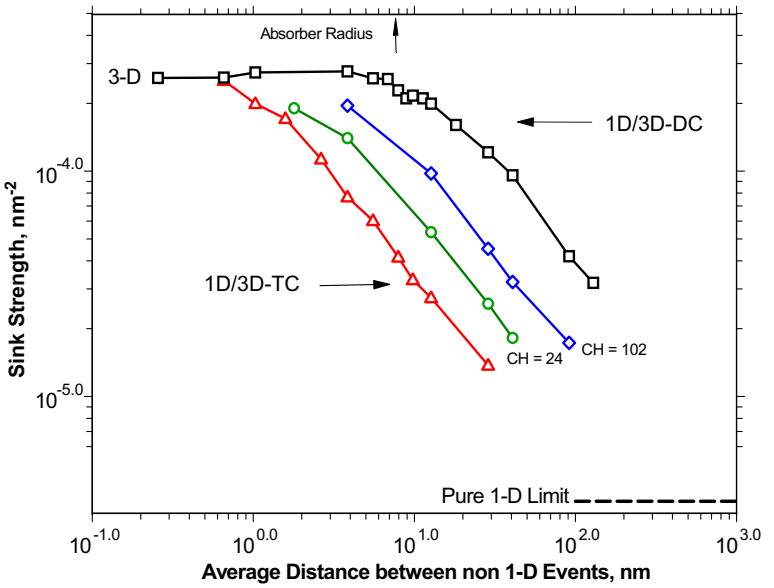

Fig. 2. Sink strength as a function of the average length of 1D migration path between disturbances of the 1D migration by climb events (1D/3D-TC) compared to effects of the disturbance by change of glide direction (1D/3D-DC). Data lying between the '1D/3D-DC' and '1D/3D-TC' curves are for cases of 1D/3D-TC in which more 'climb hops' (CH) are performed during each climb excursion from 1D migration, for CH = 24 and 102. Distances in nm represent values for a face-centered cubic Cu lattice.

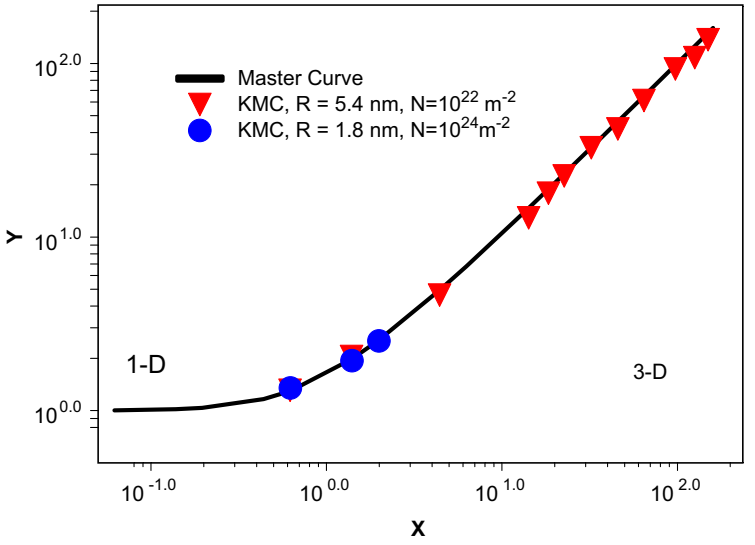

Fig. 3. Comparison of the analytical expression of the '1D/3D-TC master curve', Eqs. (2), (3) and (5), to the KMC results for two different conditions involving absorbers of different radius R and number density N.

non-1D events for both types of defect migration. For 1D/3D-DC migration the 1D glide migration of each defect along a close-packed row of atoms is regularly interrupted by a change in direction of the 1D migration after a given number of 1D hops. For 1D/3D-TC migration, the 1D glide migration of the defect is regularly interrupted by a 2D climb excursion transverse to the 1D glide migration direction to an adjacent close-packed row, at which point it continues 1D migration along the original 1D glide direction (see Fig. 1). In both cases less frequent interruption of the 1D glide results in greater distance traveled during each individual 1D glide migration path segment.

It is possible to determine the limiting cases of the interaction probabilities of migrating defects with

spherical voids for pure 1D and pure 3D defect migration using simple, geometrically-derived expressions. However, when using the KMC simulations to investigate the sink strengths near, but not at, these limiting cases, i.e., for 'almost pure 1D' and 'almost pure 3D', some difficulties are encountered. When modeling the almost pure 1D case for defects in a field of absorbers of realistic size and number density, an extremely large model volume is necessary to contain the 1D migrating defects within the model until absorption. To make the effective volume larger, periodic boundary conditions can be applied to the test cell, but only for cases where the 1D path lengths of migrating defects are significantly smaller than the size of the test volume. The lack of KMC data points near the pure 1D limit in Figs. 2 and 3 is a result of the difficulty in treating the near-1D condition. A different problem is encountered when approaching the analytical 'pure 3D' case in the KMC simulations. The geometrically-derived 3D sink strength, $k_3^2 = 4\pi RN$, described in Section 2, and used in reaction rate theories, assumes the material is a continuous medium, whereas in the KMC simulations the defects make discrete hops on a crystal lattice. Thus, in the 3D limit of KMC (defined as equal probability of hops to all nearest-neighboring lattice sites) the discrete hops in specific directions do not sufficiently approximate a random displacement in a continuous medium. The KMC-derived values of sink strengths for 'pure 3D migration' by hopping on a crystal lattice, where pure 3D migration is defined as equally probable single hops to all nearest neighbor lattice sites, are therefore operationally defined as

$$k_3^2 = 12/(a^2\langle n_3\rangle), \tag{1}$$

where a^2 is the square of the lattice parameter and $\langle n_3\rangle$ is the average number of '3D hops' until absorption. Since the numerical values of this quantity determined in KMC simulations are somewhat different from the corresponding continuum values, in all comparisons of the results of the analytical theory and the KMC simulations, the KMC-derived values for k_3^2 were used as the standard values for pure 3D defect migration.

In Fig. 2, the values of the sink strengths determined from the KMC simulations are plotted as a function of the average distance traveled by the defects in each 1D migration segment, i.e. the distance traveled between the occurrence of the non-1D events. In this paper, the non-1D events studied are either 1D/3D-DC or 1D/3D-TC. In both cases the average length of the 1D segments is proportional to the average of the square root of the number of 1D hops in each 1D segment. All simulations reported in Fig. 2 were done on a lattice representative of face-centered cubic Cu in a test cell containing the same set of absorbers, having the same size and number density. Thus, all differences in the sink strength values that represent the defect absorption are due solely to the differences in the migration characteristics of the defects.

In Fig. 2, the KMC simulation results for the sink strength of spherical absorbers interacting with defects migrating by 1D/3D-DC are compared to the results for defects migrating by 1D/3D-TC glide that is modified by a single climb excursion to a nearest neighbor or next-nearest neighbor lattice site before resuming 1D glide. Perhaps not surprisingly, Fig. 2 shows that the defects migrating by 1D/3D-TC exhibit more 1D-like behavior nearer the pure 3D limit than do the defects migrating by 1D/3D-DC. When the average distance between non-1D events is on the order of the absorber radius R or smaller, the 1D/3D-DC migration remains effectively pure 3D, whereas for 1D/3D-TC migration the near-3D sink strength appears to be unaffected by the absorber radius. The two sets of data points plotted between the 1D/3D-TC and 1D/3D-DC data sets are results for the 1D/3D-TC mode when the defects make multiple 2D climb hops (of 24 and 102 hops, respectively) during each excursion from 1D migration. The addition of larger climb excursions during each interruption of 1D pushes the sink strengths for 1D/3D-TC toward the 1D/3D-DC values.

4. Master curve

For 1D diffusion disturbed by direction changes, 1D/3D-DC, an analytical single value function for the sink strength ('master curve') interpolating between the 1D and 3D limiting cases was developed earlier by Trinkaus et al. [11], and it was shown to match very well with sink strength data generated in KMC simulations for 1D/3D-DC migration. This master curve concept has subsequently been applied to describe 1D/3D-TC migration as well. Surprisingly, the sink strength for the latter case can be described by a master curve of the same form as for 1D/3D-DC if the variable for the 'disturbance of 1D' is chosen properly – in spite of a completely different functional form of this variable. The new master curve is presented

here for comparison with results of the KMC simulations. Details of the derivation of the new master curve and discussion of its implications will be presented in another publication [13].

The master curve has the form

$$Y = 0.5\{1 + [1 + 4X^2]^{1/2}\}, \quad (2)$$

where Y is the sink strength normalized to its value for pure 1D,

$$Y = k^2/k_1^2 \quad (3)$$

and X is a measure of the 'disturbance of 1D diffusion'.

The 'disturbance of 1D diffusion' by Burgers vector changes (1D/3D-DC) is represented as

$$X = (l^2k_1^2/12 + k_1^4/k_3^4)^{-1/2}, \quad (4)$$

where l is the 1D path length between direction changes.

For 1D/3D-TC the disturbance of 1D diffusion due to conservative climb is given as

$$X = (\delta^{1/2} f(\delta) k_3^2/k_1^2), \quad (5)$$

where $\delta = D_{tr}/D_{lo}$, the ratio of the transverse and longitudinal diffusivities, and

$$f(\delta) = 3[1 - \delta]^{1/2}/\{(1 + 2\delta)\arccos(\delta^{1/2})\}. \quad (6)$$

According to the parameters contained in the variable X, the sink strength does not only depend on the parameters describing the intrinsic disturbance, l and δ, but also on parameters characterizing the sink structure (density and size expressed by the 1D and 3D limiting cases of the sink strength).

In Fig. 3, the KMC results for 1D/3D-TC migration are compared to the analytical master curve, Eq. (2). In addition to the KMC simulations described in Fig. 2, simulations were also performed for a system of absorbers having a smaller size, $R =$ 1.8 nm, and a higher number density, $N = 10^{24}\,m^{-3}$, especially to achieve results closer to the pure 1D limit, as well as to test the master curve over a range of conditions. The agreement of the master curve with the KMC results for both sets of absorbers is excellent.

5. Conclusions

The KMC simulations confirm the 1D to 3D behavior predicted by the analytical expressions for both 1D diffusion disturbed by direction changes (1D/3D-DC) and by transversal diffusion due to climb (1D/3D-TC). The master curve relating the normalized sink strength to the disturbance of 1D diffusion, corroborated by KMC modeling, illustrates the validity of the comprehensive analytical representation of these phenomena. Furthermore, the analytical representation of 1D to 3D diffusion reaction kinetics utilized to form the master curve is well-suited for modeling damage accumulation under the locally anisotropic diffusion conditions of defects in materials where such 1D migrating defects are formed.

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