

**KINETIC MONTE CARLO SIMULATIONS OF THE EFFECTS OF 1-D DEFECT TRANSPORT ON DEFECT REACTION KINETICS AND VOID LATTICE FORMATION DURING IRRADIATION** - H. L. Heinisch (Pacific Northwest National Laboratory)\* and B. N. Singh (Risø National Laboratory, Denmark)

**PURPOSE**

The purpose of this computational study is to study the defect reaction kinetics of one-dimensionally migrating crowdion clusters as a function of the frequency of their direction changes, specifically to determine the sink strengths for such one-dimensionally migrating defects, as well as to demonstrate the significant role crowdion clusters may have in the formation and stability of void lattices.

**SUMMARY**

Within the last decade molecular dynamics simulations of displacement cascades have revealed that glissile clusters of self-interstitial crowdions are formed directly in cascades. Also, under various conditions, a crowdion cluster can change its Burgers vector and glide along a different close-packed direction. In order to incorporate the migration properties of crowdion clusters into analytical rate theory models, it is necessary to describe the reaction kinetics of defects that migrate one-dimensionally with occasional changes in their Burgers vector. To meet this requirement, atomic-scale kinetic Monte Carlo (KMC) simulations have been used to study the defect reaction kinetics of one-dimensionally migrating crowdion clusters as a function of the frequency of direction changes, specifically to determine the sink strengths for such one-dimensionally migrating defects. The KMC experiments are used to guide the development of analytical expressions for use in reaction rate theories and especially to test their validity. Excellent agreement is found between the results of KMC experiments and the analytical expressions derived for the transition from one-dimensional to three-dimensional reaction kinetics. Furthermore, KMC simulations have been performed to investigate the significant role of crowdion clusters in the formation and stability of void lattices. The necessity for both one-dimensional migration and Burgers vectors changes for achieving a stable void lattice is demonstrated.

**PROGRESS AND STATUS**

This is an extended abstract of a paper submitted for publication in Philosophical Magazine as part of the proceedings of the First International Conference on Multiscale Materials Modeling, Queen Mary University of London, June 17-20, 2002.

We have used KMC simulations to study the effects of Burgers vector direction changes on the reaction kinetics of crowdion clusters, making many simplifying assumptions in order to isolate the relationships among the variables of interest. In these KMC defect reaction kinetics "thought experiments," sink strengths are determined as a function of the average length,  $L$ , of the 1-D path segments traversed by migrating crowdion clusters before they change their Burgers vector. The crowdion cluster does a 1-D random walk along a particular close-packed direction, then changes its Burgers vector and performs a random walk along a different close-packed direction. In this way it migrates in all three dimensions, but on a path made up of segments of 1-D walks. In our simulations we characterize such migration in terms of the average of the total lengths, minimum to maximum, of the paths traversed by the defect during  $n_{dc}$  hops in the absence of any defect interactions, where  $n_{dc}$  is the number of hops made between direction changes. Thus, in our simulations we define  $L$  as the average 1-D path length for defects that change direction regularly after every  $n_{dc}$  hops.

The usual test cell is a cube containing fcc lattice points with a volume of about  $1\mu\text{m}^3$ , assuming the lattice parameter of Cu. (The phenomena being investigated are not expected to differ qualitatively for fcc and bcc lattices.) The lattice points represent positions in the cell, and all defects, fixed or mobile, are

---

\* Pacific Northwest National Laboratory (PNNL) is operated for the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO-1830.

“associated” with a lattice site, even defects that are not localized at a lattice point in the real material, such as SIA dumbbells and crowdions. In the KMC all mobile defects are assumed to migrate by hopping from their present lattice site to a nearest neighbor lattice site. Defects are the interacting entities in the model; individual atoms are not specifically dealt with.

### Sink Strengths

Initial KMC studies of sink strength as a function of  $L$  were performed with  $10^4$  immobile spherical absorbers of capture radius  $R$  and number density  $N = 10^{22} \text{ m}^{-3}$  randomly arranged on lattice sites within the volume. A mobile defect representing a crowdion cluster is introduced into the central volume of the cell at a random position and is followed until it is absorbed by an absorber. The mobile defect performs a 1-D random walk along a randomly chosen close-packed direction (  $[110]$  fcc), changing to another randomly chosen close-packed direction each time it has completed a fixed number of hops,  $n_{dc}$ . When the mobile defect comes within the capture radius  $R$  of an absorber, it is absorbed without changing the size or position of the absorber, and the defect lifetime  $j$ , the total number of hops before absorption, is recorded. Each simulation is performed for a set of  $10^3$  mobile defects. The sink strength  $k^2$  is defined as the inverse of the square of the mean free path, and it is determined in the simulations for 1-D migrating defects in a field of spherical absorbers from the expression

$$k^2 = 2/a^2 \langle j \rangle , \quad (1)$$

where  $a$  is the hop distance along  $[110]$  and  $\langle j \rangle$  is the average defect lifetime in hops for the set of migrating defects.

Sink strengths  $k^2$  were determined for migrating defects in a field of spherical absorbers as a function of  $L$ . The individual data points in Figure 1 indicate the sink strengths determined in the simulations as a function of  $L$  for various values of  $R$  and  $N=10^{22} \text{ m}^{-3}$ . Since  $N$  is constant, the volume fraction of absorbers decreases with decreasing  $R$ . The sink strengths for pure 3-D ( $L = a$ ) are at the far left, and the sink strengths approach pure 1-D values on the right (large  $L$ ). The region between 1-D and 3-D, and especially the region of transition to 3-D, are the most important to understand, because that is where the behavior in the real case is expected to lie. For each value of  $R$  the sink strengths remain almost constant at the “3-D value” until about  $L=R$ . Thus, the transition of sink strengths from 1-D to 3-D depends on  $R$  as well as  $L$ .

In conjunction with these KMC simulations, an analytical expression for sink strengths for 1-D migrating defects as a function of  $L$  interacting with spherical absorbers was developed by Trinkaus et al. (2002) strictly from theoretical considerations. It has the form

$$k_m^2 = 0.5 k_1^2 \{ 1 + [ 1 + 4/(L^2 k_1^2/4 + k_1^4/k_3^4) ]^{1/2} \}, \quad (2)$$

where the sink strengths  $k_1^2$  and  $k_3^2$  are for pure 1-D and pure 3-D defect migration, respectively, and they can be expressed analytically in terms of continuum models. However, to compare the analytical expression eq. 2 to the simulation results, it is necessary to account for the effects of discreteness of the crystal lattice. Hopping on a discrete lattice at the “3-D limit,”  $n_{dc} = 1$  ( $L=a$ ), the defect makes random hops to nearest neighbor sites. The nature of the defect’s migration path is still a series of 1-D segments, but the segments are only one hop in length in this limiting case. Thus, the values for  $k_3^2$  used in eq. 2 were taken to be the sink strength values extracted from the simulations for  $L=a$ . The expression in eq. 2, modified for the discrete simulations, is used to plot the solid line curves in Figure 1. The agreement between the analytical expression and the simulation results is excellent.

### Void Lattices

Simulations of void lattice formation were performed using the same basic KMC model, starting from a random array of small voids and introducing randomly positioned crowdion and vacancy clusters that

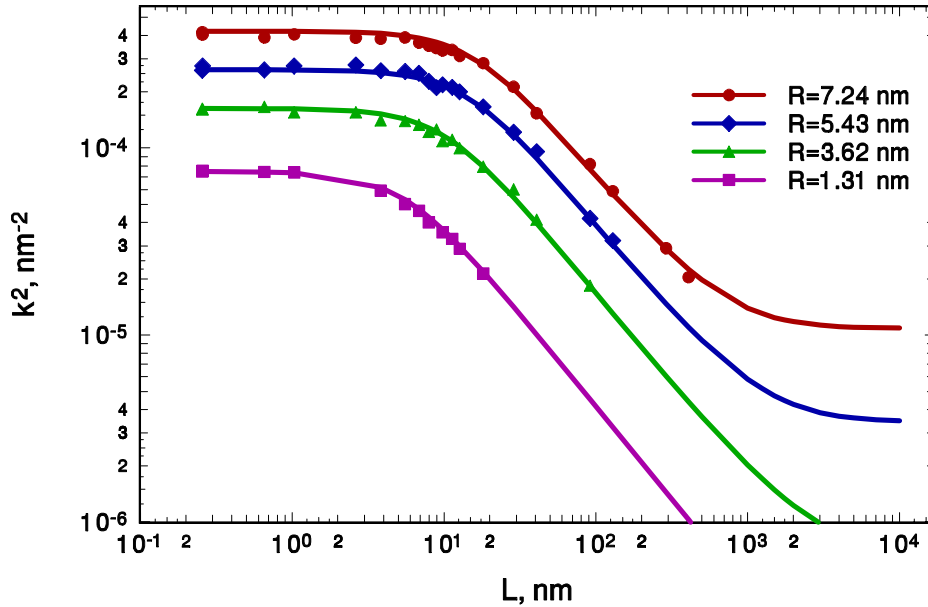


Figure 1. Sink strengths as a function of  $L$ , the average 1-D path length of 1-D/3-D migrating defects, for spherical absorbers of various radii. The points are results of the KMC simulations and the lines are calculated from Equation 2.

interact with the voids. Periodic boundaries are imposed, and the mobile defects interact only with the voids.

Many initial scenarios for void lattice formation from a set of randomly distributed voids were explored before success – quantifiable void ordering – was achieved. An initial minimum density of small, randomly-positioned voids seems to be a prerequisite for forming a void lattice. Starting with a low density of random large voids will lead to a void lattice only if the voids can be efficiently moved into position by collisions or destroyed and regrown, which are highly unlikely processes in such a situation where the shadow effect of already aligned voids is weak. The most likely scenario starts with a high enough concentration of small voids that there is a small void very near every position a lattice void will occupy. Then, by the shadow effect, the crowdion clusters will select the lattice, destroying the voids not in lattice positions and allowing the others to grow.

In radiation experiments void lattices are observed to form in small local regions of ordering which grow in extent with increasing dose. This implies that initial ordering takes place locally, perhaps as a result of the shadow effect caused by one significantly larger void (or perhaps two or more large voids correlated along a close-packed direction) occurring as a result of statistical fluctuations. To test this, simulations were performed in a fcc cell with a cube edge of 120 atomic lattice parameters, starting with a 1000 small voids and a “seed” consisting of 14 much larger voids arranged in a fcc super-lattice. Simulations were also done with the same initial conditions but with a seed consisting of a single large void in the center of the cell, and also with no seed. After irradiating with  $6 \times 10^5$  crowdion clusters with  $n_{dc} = 2 \times 10^4$  hops ( $L \approx$  cell cube edge) and  $5 \times 10^5$  vacancy clusters, very similar ordered lattices of voids were produced in all three cases, but with a void lattice parameter somewhat smaller than that of the 14 void seed. Figure 2 shows the initial configuration and the ordered configuration after irradiation in the simulation with no



information to experimental measurements of the spacing of void lattices and the void size. In this way, void lattice experiments may provide a direct link, through the KMC modeling, for investigating characteristics of the primary defect production.

#### **ACKNOWLEDGEMENTS**

This work was supported by the Office of Fusion Energy Sciences of the U.S. Department of Energy and the European Fusion Programme.