KINETIC MONTE CARLO SIMULATIONS OF DISLOCATION DECORATION AND RAFT FORMATION IN BCC-IRON UNDER CASCADE IRRADIATION— M. Wen, N. M. Ghoniem (Department of Mechanical and Aerospace Engineering, University of California, Los Angeles), and B. N. Singh (Risø National Laboratory, Denmark)

OBJECTIVE

Radiation-induced microstructural and compositional changes in metals are governed by the interaction between point and clustered defects and the internal microstructure. The objective of this computational study is to investigate the inhomogeneity and segregation of point defects, damage accumulation by diffusion and interaction, and the resulting dislocation decoration and formation of rafts of interstitial clusters. A new Kinetic Monte Carlo (KMC) computer code has been developed, with explicit representation of the influence of elastic interaction between microstructure features. Results of computer simulations are utilized to discuss the general conditions for experimentally observed dislocation decoration and formation of rafts.

SUMMARY

Under neutron irradiation, primary defect clusters of both self-interstitial atom (SIA) and vacancy type are directly produced in displacement cascades, which have been confirmed by experiments as well as molecular dynamics simulations. The highly mobile SIA clusters play a crucial role in the development of characteristic microstructures, such as rafts of SIA clusters and dislocation decoration, and the corresponding radiation hardening behavior [1]. We have developed a new approach to KMC simulations to investigate the segregation and accumulation of point defects at the atomic scale with incorporating the elastic interaction between defect clusters and microstructures by using the elastic representation of point defects due to Kröner [2]. The decoration of dislocations by SIA clusters and the formation of rafts in bcc-iron are modeled in detail, and the general conditions for the occurrence and development of both features have are discussed. We also present results of SIA cluster density as a function of irradiation dose, using cluster size distributions, cascade frequency, and individual cluster dynamic parameters obtained from molecular dynamics simulations. Good agreement is found between the results of our present KMC simulations and experimental observations. The one-dimensional motion of glissile SIA clusters and the interaction between defects and microstructures are shown to be the main cause for appearance and development of characteristic microstructures. It is demonstrated that KMC computer simulation is a valuable tool in studying defect kinetics and microstructure evolution, taking into account many different microscopic mechanisms and covering very different time and length scales.

PROGRESS AND STATUS

Introduction

Under neutron irradiation, primary defect clusters, which are directly produced in displacement cascades, play an important role in microstructural evolution and property changes in irradiated materials. Small interstitial loops can further re-organize to make up patches (or rafts) at elevated temperatures [3], and dislocations are often observed being heavily decorated by SIA clusters in the form of small interstitial loops [1]. Rafts of SIA loops and decoration of dislocations with loops have become a crucial issue for understanding radiation hardening under cascade damage conditions. It has been experimentally observed that in the deformation process of metals and alloys under cascade damage conditions, an increase in the upper yield stress without dislocation generation occurs, and is followed by an immediate yield drop and plastic instability. A ‘cascade induced source hardening’ (CISH) model has been proposed by Singh et al. [1] and applied to explain the experimental observation of yield drop. Huang and Ghoniem [4] investigated the interaction dynamics between sessile SIA clusters and dislocations in terms of elastic interactions with various SIA cluster densities, the spatial and size distributions of clusters, and the

orientation distribution of individual Burgers vectors by using the method of parametric dislocation dynamics (PDD) simulations.

In the study of radiation damage MD simulations using semi-empirical embedded-atom method (EAM) interatomic potentials have played a very important role in understanding the details of defect production in displacement cascades, and in understanding the dynamics of point defect and defect cluster diffusion. Radiation damage, however, includes a vast range of irradiation effects, such as production and diffusion of point defects, and their interaction with microstructures, which take place over time and length scales that span many orders of magnitude. MD techniques are inapplicable to deal with microstructure evolution in irradiated materials due to their limitation of time and length scales. Kinetic Monte Carlo (KMC) simulations, on the other hand, provide another option to perform atomic-level modeling of defect kinetics and microstructure evolution over relevant length and time scales. KMC has been used to investigate the local short-term intra-cascade annealing of individual cascades [5], as well as low dose defect accumulation [6]. In the present work, KMC techniques are used to study key aspects of microstructure evolution under irradiation: the inhomogeneity and segregation of point defects, and damage accumulation by diffusion and interaction, and the resulting dislocation decoration and formation of rafts of interstitial clusters.

Model

The basic ideas behind KMC simulations have been comprehensively described in the literature, e.g. [7, 8]. In our work, microstructure evolution of the cascade-induced defect clusters is accomplished by a KMC procedure in which one reaction is executed at one site during each time step. The jump frequency for a possible jump of a defect cluster, i, to take place is given by: $r_i = \omega_0 \exp(-E_i / k_B T)$, where ω_0 is the pre-exponential factor of the defect cluster, k_B the Boltzmann constant, E_i the 'effective' activation energy for jumps of the cluster, and T is the absolute temperature. The probability of selecting an event is simply equal to the rate at which the event occurs relative to the sum of all possible event rates. At each KMC step, one event denoted by m is randomly selected from all possible M events, as follows:

$$\frac{\sum_{i=0}^{m-1} r_i}{\sum_{i=0}^{M} r_i} < \xi_1 < \frac{\sum_{i=0}^{m} r_i}{\sum_{i=0}^{M} r_i} \tag{1}$$

where r_i is the rate at which event i occurs ($r_0 = 0$) and ξ_1 is a random number uniformly distributed in the range [0, 1]. Once an event is chosen, the system is changed correspondingly, and the list of events that can occur at the next KMC step is updated.

The reciprocal of an atomic jump probability per unit time is the residence time for a defect cluster that moves by that specific type of jump. Since the jump probabilities of all the different types of jumps are independent, the overall probability per unit time for the system to change its state by any type of jump step is just the sum of all the possible specific jump type probabilities, and so the residence time that would have elapsed for the system in a specific configuration is the reciprocal of this overall jump probability: $\Delta t = 1 / \sum_{i=0}^{M} r_i$. For random uncorrelated processes, this is a Poisson distribution. If ξ_2 is a random number from 0 to 1, the elapsed time for a particular transition is given by: $\Delta t = -\ln \xi_2 / \sum_{i=0}^{M} r_i$.

The system is then advanced to the final state of the chosen transition and the process is repeated. The expression for Δt is rigorous, and a derivation is also provided by [8]. In our KMC simulations, the elastic interaction is incorporated. The influence of other defects and the external stress on one SIA or vacancy cluster is given by the stress field σ_{ij}. By applying the infinitesimal dislocation loop approximation for SIA clusters, the work necessary to form the loop characterized by normal $\mathbf{n}$, Burgers vector $\mathbf{b}$ and area δA in

the stress field σ_{ij} is SIA cluster interaction energy E_{int}, and is given by: $E_{\text{int}} = \int_V \sigma_{ij}^{(1)} \varepsilon_{ij}^{(2)} dV$, in which $\sigma_{ij}^{(1)}$ is the stress arising from the first dislocation and $\varepsilon_{ij}^{(2)}$ the strain originating in the other. For the present study, the second loop (defect cluster) is assumed to be infinitesimal, the interaction energy can be simplified to [9]: $E_{\text{int}} = \delta A^{(2)} n_i^{(2)} \sigma_{ij}^{(1)} b_j^{(2)}$, where $n_i^{(2)}$ is the unit normal vector to the defect cluster habit plane of area $\delta A^{(2)}$. The total cluster activation energy is then given by: $\widetilde{E}_m = E_m + \Delta E_{\text{int}}$, where E_m is the activation energy in a perfect crystal structure and can be obtained by either experiments or MD simulations, and ΔE_{int} the difference in the interaction energy of an defect cluster placed at two neighboring equivalent position in the crystal. The numerical method developed by Ghoniem [10], and Ghoniem and Sun [11] was employed in our simulation to evaluate the interaction between small defect clusters and slip dislocation loops. A computation cell of 100 × 100 × 100 nm^3 is taken with periodic boundary conditions. For the purpose of studying dislocation-cluster interaction, a slip dislocation loop lying on the [1 0 1] plane, with Burgers vector **b** = [-1 1 1] / 2 is introduced into the simulation box.

Results

Dose dependence of defect density

Doses up to 5.21 × 10^{-3} dpa were simulated at a damage rate of 5 × 10^{-8} dpa/s, and damage accumulation was observed as a function of dose. Fig. 1 shows the SIA cluster density as a function of dose up to 5.21 × 10^{-3} dpa with considering recombination, as well as of dose up to 1.48 × 10^{-3} dpa without recombination centers. In addition, SIA clusters containing more than 100 interstitial atoms (diameter > 3 nm) are counted as visible clusters, and the corresponding cluster density is calculated and compared with the experimental results for bcc Fe irradiated at ~70°C in the HFIR rector at Oak Ridge National Laboratory to displacement dose levels in the range of 10^{-4} to 0.72 dpa [12].

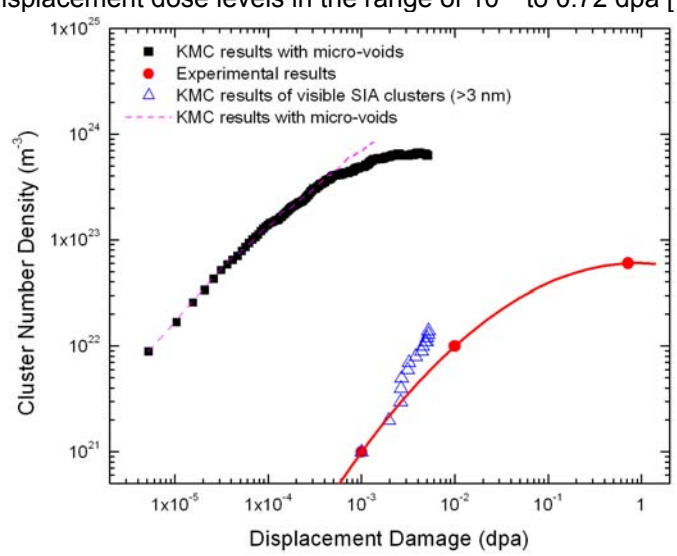

Fig. 1. SIA cluster densities as a function of accumulated dose for bcc iron irradiated at 300 K under cascade irradiation at 5 × 10^{-8} dpa/s. Simulation results are compared to experimental data from TEM observation of iron irradiated in HFIR [12].

At the initial stages of irradiation (dpa < 10^{-4} dpa), the cluster density increases almost linearly with damage dose. The increase in cluster density is then slowed down, when the dose level is higher than 10^{-4} dpa and lower than about 3 × 10^{-3} dpa. Cluster density approaches a saturated value and does not change much beyond a dose level of 3.5 × 10^{-3} dpa. At very low dose, cluster densities of both interstitials and vacancies are rather low, and the chance that one interstitial cluster could get close

enough to another interstitial or vacancy cluster so that they can feel each other in terms of elastic interaction is rather small. Because of the 1D motion of SIA clusters, the recombination cross-section with vacancy clusters produced as well as the agglomeration cross section with other interstitial clusters are small. Theoretically, there are only two mechanisms by which a SIA cluster could change its diffusion direction: either by thermal activation or by the interaction with other defects or microstructures. The effect of thermal activation can be rule out here because our simulation is carried out at constant room temperature. At low damage dose level, the big distance between clusters makes the interaction between them weak. This hardly affects the migration of SIA clusters, resulting in a linear increase of cluster density with dose.

When the damage builds up to a certain level, say 10^{-4} dpa in our simulations, the simulation box becomes crowded. Newly introduced interstitial clusters as well as existing mobile clusters have higher probability of encountering one another and interacting with each other. The events of recombination and agglomeration would occur more frequently than at the early stage of irradiation. Consequently, these reactions slow down the increase in the number of SIA clusters within the simulation box. Once the damage accumulates to some extent, 3.5×10^{-3} dpa in our simulation, the number of interstitial clusters that annihilate or form larger clusters reaches dynamic equilibrium with clusters produced by new cascades. Thus, the density of clusters in the simulation box reaches its saturation level. Vacancy clusters, i.e. micro-voids in our simulation, have significant influence on the density of interstitial clusters. As we can see in Fig. 1, at a dose level lower than 5×10^{-4} dpa, the difference between the cases of including micro-voids and no micro-voids was is distinct. But the difference increases as the dose increases, and at a dose of 1.5×10^{-3} dpa, the cluster density of the micro-voids free case is about 50% higher than that of the case with micro-voids. This indicates that the existence of vacancy clusters considerably reduces the density of mobile interstitial clusters, and that recombination plays an important role in microstructural evolution.

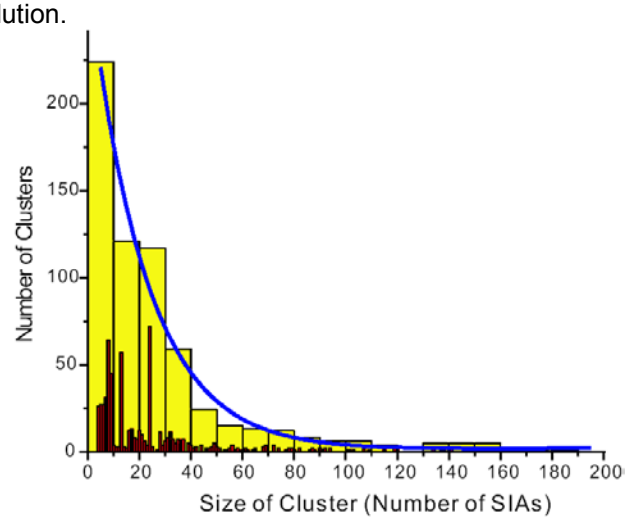

Fig. 2. Size distribution of SIA clusters at a dose level of 5.21×10^{-3} dpa. The red slim bar is the number of clusters of a certain size (fine resolution), and the yellow thick bar are the total number of clusters within a certain range of sizes (coarse resolution).

Although the number density of SIA clusters reaches steady state after 3.5×10^{-3} dpa, the size of clusters continues to increase with increasing dose. In order to compare the results from our simulations with experimentally measured cluster densities, it is necessary to assume a minimum size that can be resolved in the experiments. A value between 1.5 and 2 nm in diameter is quoted in the literature as the minimum size resolved by TEM [13]. For the infinitesimal dislocation loop approximation adopted here, the relationship between radius of the loop (R) and the number of defects (N) is $N = \sqrt{2}\pi R^2 / a_0^2$ for a bcc material, where a_0 is the lattice parameter. A radius of 1.4 nm corresponds to approximately 100 defects in bcc iron. Considering visible interstitial clusters as those with more than 100 interstitial atoms, we

extracted the total 'visible' cluster density from our simulations, as we show in Fig. 1. We can see that the fraction of 'visible' interstitial clusters obtained in the simulations presented here is larger than experimental measurements at high doses, but is still in good agreement. The number of SIA clusters as a function of cluster size at dose of 5.2×10^{-3} dpa is demonstrated by the histogram in Fig. 2. More than half of the total interstitial clusters consist of more than 30 defects though small clusters consisting of less than 10 SIAs still have the highest value for one single section. It can be expected that distribution of Fig. 2 will shift towards the bigger cluster size as damage accumulates.

Characteristics of decoration and raft formation

The main feature of the present KMC simulations that is different from previous work is that the elastic interaction between defects and microstructure is included. Distinct spatial heterogeneity features of damage accumulation under cascade irradiation, such as dislocation decoration and raft formation, can actually be studied. With regard to dislocation decoration, except for a direct encounter of a mobile SIA cluster with a dislocation, clusters will generally be trapped in a region of strong attractive elastic interaction. As expected, at very low dose levels ($< 10^{-4}$ dpa), the probability that a dislocation finds a cluster in the attractive region, and can thus affect the motion of the cluster is rare. Thus, the decoration phenomenon does not occur in the early stage of irradiation. The decoration process builds up quickly with increasing dose, and at 3×10^{-4} dpa, the dislocation has attracted a group of interstitial clusters trapped in its neighboring area. The microstructure configuration of interstitial clusters at 1.3×10^{-3} dpa is shown in Fig. 3(a) where the decoration by interstitial clusters along the dislocation line has become very significant. The accumulation of glissile interstitial clusters near the dislocation is further intensified at dose of 5.2×10^{-3} dpa as shown in Fig. 3(b). When an extremely mobile one-dimensionally migrating interstitial cluster passes through the neighborhood of a pre-existing dislocation, it will feel the influence of the strain field of the dislocation. As long as the defect/dislocation interaction is attractive and the distance is small enough, the interaction of the defect with the stress field of the dislocation could be so strong that the cluster cannot escape from within the attractive zone by thermally activated random walk. Once an interstitial cluster is trapped into the strain field of a dislocation, it rotates its Burgers vector to accommodate to the strain field of the dislocation, and migrates along the direction of lowest energy barrier.

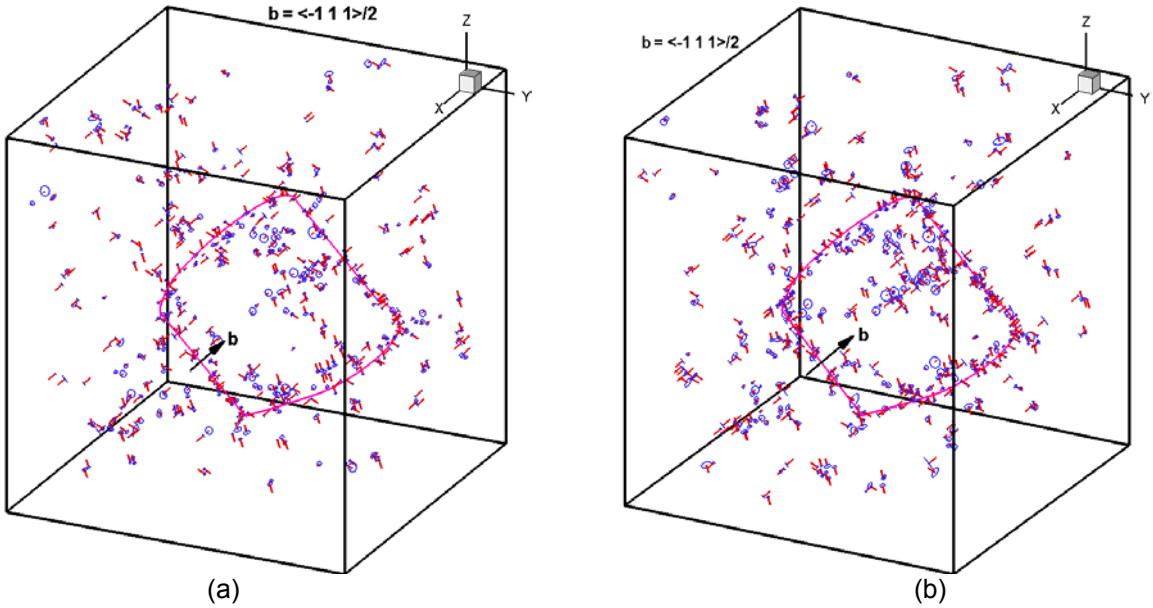

Fig. 3. Snap-shots of the microstructure of interstitial clusters at: (a) 1.3×10^{-3} dpa, where the decoration by interstitial clusters along the dislocation line can be clearly seen; (b) 5.2×10^{-3} dpa, where the accumulation of glissile interstitial clusters near the dislocation is further intensified.

Using dislocation dynamics simulation, Ghoniem et al. [14] investigated the elastic interaction between nano-size prismatic loops and grown-in dislocation loops in bcc Fe and fcc Cu. Their calculation shows that the iso-energy surface for bcc iron has a maximum width around the edge component of the dislocation. Our simulations show a good agreement with their results. It can be clearly seen in Fig. 3 that the pure edge components of the slip dislocation gets more SIA clusters trapped in its vicinity than the mixed character segments. Due to the influence of the strain field of the dislocation, the majority of the decorating clusters have changed their Burgers vector to parallel to the Burgers vector of the dislocation. Trapped clusters can still serve as sinks for glissile clusters, and increase their size by agglomeration before they make a Burgers vector rotation and finally get absorbed by the dislocation. With the accumulation of clusters along the dislocation line, a repulsive force field is gradually built up against further cluster trapping.

Another major striking feature of microstructure evolution under cascade induced irradiation damage is the formation of SIA cluster rafts. This type of frequent experimental observation is also demonstrated here. Fig. 4 shows a configuration of rafts of interstitial clusters forming at a dose level of 1.8×10^{-3} dpa. The Burgers vectors of the clusters making up the raft are parallel to each other, which is in agreement with the experimental observation. When an interstitial cluster approaches another one, the two are subject to the influence of the strain field of each other. If the interaction between them is attractive and strong enough to overcome the energy barrier for directional change, clusters will comply with the interaction between them in terms of adjusting the relative positions and orientations. This scenario is similar to the pinning of clusters, which has been demonstrated in our earlier work [15]. The nature of the configuration of pinning clusters is still glissile, which has been observed in our simulations. The development of this pinning process eventually leads to formation of an extended stable complex that consists of a group of SIA clusters lying on parallel planes. As the number of clusters within a complex increases the mobility of the complex as a whole decreases. The decrease in the mobility of individual clusters can be attributed to the elastic interaction between member clusters within one complex. Such strong interaction prevents clusters from breaking the strain field of others and jumping away. The complex of clusters can grow further, attracting more glissile cluster with the same Burgers vector, thereby creating a raft of SIA clusters.

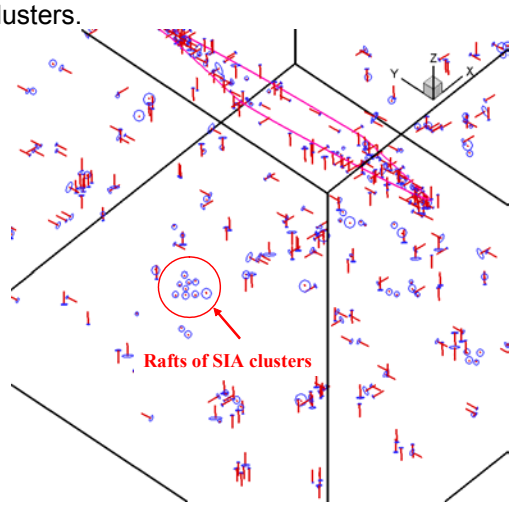

Fig. 4. A close view of the configuration of a raft of interstitial clusters formed at a dose level of 1.8×10^{-3} dpa.

There are two key factors that play an important role in the process of damage accumulation and resultant dislocation decoration and raft formation: (1) the one-dimensional motion of glissile SIA clusters created in displacement cascades, and (2) the interaction between glissile defects and the microstructure. The restriction of diffusion of SIA clusters to 1-D leads to a reduction in the reaction rate with other defects, because one dimensionally moving clusters are able to travel in the atmosphere of randomly

distributed lattice defects through larger distances than those 3-D moving ones. Consequently, the possibility that such SIA clusters annihilate at extended sinks such as pre-existing dislocations and grain boundaries increases. Our simulation demonstrates that even at a fairly low dose of 1.3×10^{-3} dpa a clear decoration phenomenon has already taken place in the vicinity of the pre-existing dislocation loop.

The energy barrier for directional change is a crucial quantity in controlling when, how and in what extent the dislocation decoration and raft formation occur. It determines the maximum range for the elastic interaction that could be strong enough to surpass the barrier and thereby leads to Burgers vector change, as well as the trapping region in the vicinity of dislocations and clusters. In other words, the interaction between defects and microstructure contributes to dislocation decoration and the formation of rafts in terms of changing the diffusivity and the characteristics of mixed 1D/3D migration. Our present simulations suggest that raft formation could be achieved just by prismatic glide of glissile interstitial clusters under the condition of one-dimensional migration combined with strain-field-induced Burgers vector changes. A necessary condition for pronounced formation of rafts of glissile clusters is that the group of clusters having the same Burgers vector is big enough to trap a single glissile cluster in the strain field formed by the group and prevent any trapped cluster from de-trapping or Burgers vector change before it is immobilized by other clusters in the same group. Our simulations indicate that small rafts containing two or three clusters are still mobile; more specifically, these small patches still perform one-dimensional migration. With the size of a patch increasing, the mobility of the whole patch decreases, and a raft consisting of more than five clusters is virtually immobile. Due to thermal activation or interaction with other defects, an interstitial cluster trapped in the outer region of a raft may break loose from the strain field of the raft, and thus gets de-trapped from the raft.

Conclusions

A new approach of KMC simulations, which incorporates elastic interaction between various microstructures, has been developed. The approach is used to investigate the spatial inhomogeneity and segregation of point defects, as well as damage accumulation under cascade irradiation in bcc iron. KMC results have been validated by comparison with experimental observations, including dislocation decoration and raft formation, and the dose dependence of defect density. KMC simulations proved to be a valuable tool in modeling defect kinetics and microstructure evolution at the atomic-level, taking into account many different microscopic mechanisms and covering very different time and length scales.

The importance of glissile SIA clusters produced in displacement cascades has been demonstrated to play a decisive role in irradiation-induced microstructural evolution. The decoration of dislocations by SIA clusters and the formation of rafts have been modeled in detail. The one-dimensional motion of glissile SIA clusters and the interaction between defects and microstructure are shown to be the main cause for the appearance and development of rafts and dislocation decorations. At a rather low dose around 1.0×10^{-3} dpa, a large portion of the initially glissile clusters is trapped around slip dislocation loops (the majority of them are distributed along edge components) and become nearly immobile. The high concentration of SIA loops around slip dislocations results in an extremely inhomogeneous spatial distribution. Our simulations show a clear picture of how SIA clusters are attracted to dislocations to decorate and lock them in place. The simulations show good agreement with experimental observations, at least qualitatively. In addition, for the first time our KMC simulations demonstrate the formation of rafts of glissile clusters with the same Burgers vector. It is suggested that raft formation could be achieved just by prismatic glide of glissile interstitial clusters under the condition of one-dimensional migration combined with strain-field-induced Burgers vector changes. Dose dependence of defect density was investigated quantitatively, using characteristic cluster size distributions, cascade frequency and individual cluster dynamic parameters. The results for visible clusters show fairly good agreement with experimental measurements.

Acknowledgements

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