

ATOMIC SCALE MODELING OF DEFECT PRODUCTION AND MICROSTRUCTURE EVOLUTION IN IRRADIATED METALS* — T. Diaz de la Rubia, N. Soneda[#], E Alonso, K. Morishita[§], and Y. Shimomura[♦] (Lawrence Livermore National Laboratory, L-268, P.O. Box 808, Livermore, CA 94550)

SUMMARY

Irradiation effects in materials depend in a complex way on the form of the as-produced primary damage state and its spatial and temporal evolution. Thus, while collision cascades produce defects on a time scale of tens of picoseconds, diffusion occurs over much longer time scales, of the order of seconds, and microstructure evolution over even longer time scales. In this report we present work aimed at describing damage production and evolution in metals across all the relevant time and length scales. We discuss results of molecular dynamics simulations of displacement cascades in Fe and V. We show that interstitial clusters are produced in cascades above 5 keV, but not vacancy clusters. Next, we discuss the development of a kinetic Monte Carlo model that enables calculations of damage evolution over much longer time scales (1000's of s) than the picosecond lifetime of the cascade. We demonstrate the applicability of the method by presenting predictions on the fraction of freely migrating defects in α -Fe during irradiation at 600 K [1].

INTRODUCTION

Ferritic steels and V-based alloys are candidate materials for the first wall of fusion reactors. Their behavior under irradiation is one of the critical issues that must be considered in the final decision making process. In particular, degradation of mechanical properties due to embrittlement and swelling is one of the major concerns. Our recent work is aimed at understanding the fundamental aspects of defect production and microstructure evolution in these materials, and at developing physically-based predictive models of radiation-induced changes in mechanical properties. As a first step, we report here on the development of a hybrid approach that combines molecular dynamics (MD) with kinetic Monte Carlo (KMC) simulations [1]. This enables not only calculations of the primary damage state due to displacement cascades, but also a description of the fate of the cascade-produced defects over time scales comparable to actual experimental conditions. Such simulations can be used to predict, at the atomic scale, the formation of microstructural features such as impurity precipitates and voids during prolonged irradiation. Precipitates of impurities can act as obstacles for dislocation motion and are thought to be responsible for the increase in yield stress associated with embrittlement in these materials. Voids can nucleate at the site of transmutation He atoms, and are responsible for swelling.

In the past, MD simulations have been used extensively to model the primary damage state induced by displacement cascades in metals (for a review, see e.g. [2]). Because of the length and time scales of MD (100's of Å and tens of picoseconds), this is an appropriate tool for these calculations, but cannot be used to investigate the fate of the resulting defects and their role in producing microstructural and compositional changes. To overcome these limitations, a way of connecting the MD results to other simulation methods

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such as rate theory [3] or Monte Carlo calculations [4,5] is required. In particular, Monte Carlo simulations appear very promising because they provide the ability to perform atomic level studies of the defect kinetics and microstructural evolution over relevant length and time scales.

In this report, we present MD simulations of displacement cascades in Fe and V at energies from 100 eV to 20 keV. We then connect these results to KMC calculations that describe the long time-scale migration and evolution of these defects for the particular case of α -Fe at 600 K. To carry out the KMC calculations, we also perform studies of the formation and migration energy of point defects and defect clusters using the MD technique, and these results then serve to define the interaction rules for the KMC simulator. Results on the escape probability of defects from their nascent cascade, and on the fraction of freely migrating defects under these irradiation conditions are presented and compared to experiments.

2. MOLECULAR DYNAMICS SIMULATIONS OF DISPLACEMENT CASCADES

Collision cascades due to neutron irradiation produce defects in the matrix of structural materials. Information on the number of point defects and defect clusters as well as their spatial configuration provides the starting conditions for all further considerations regarding microstructural evolution. Our simulations were carried out with the MDCASK code [6] which can be used on either RISC workstations or massively parallel computers. The performance on a DEC workstation with the 300MHz Alpha chip is 1.2×10^{-4} CPU sec/atom-MD step, equivalent to the performance on a vintage 1988 Cray 2 supercomputer. On a Cray T3D parallel machine, the performance is 3×10^{-6} CPU-sec/atom/step on 128 processors. The largest MD simulation boxes used were cubes 250 Å on a side containing 500,000 atoms.

2.1. α -Fe

For the MD simulations presented here, the analytic EAM interatomic potential for α -Fe developed by Johnson and Oh [7] and modified by Guellil and Adams [8] was used after modifications to account properly for high energy scattering during the early stages of the cascade.

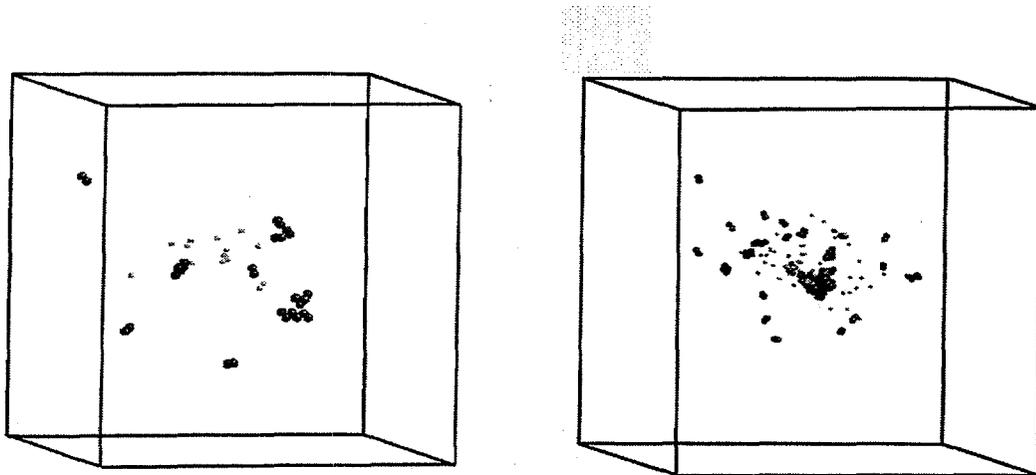


Figure 1. 5 and 20 keV displacement cascades in α -Fe. The blue spheres are interstitials and the small dots represent vacancies. A fraction of the interstitials is produced in clusters, but the vacancies are not.

Figure 1 shows a comparison of typical defect configurations produced by 5 and 20 keV displacement cascades 10 ps after the initiation of the recoil. Pairs of open circles are interstitial dumbbells, and small dots are vacancies. Fifteen Frenkel pairs are generated in this 5keV cascade simulation. Vacancies are located in the center region of the cascade, while isolated $\langle 110 \rangle$ interstitial dumbbells produced by replacement collision sequences (RCSs) can be found in the periphery. Two di-interstitials were also formed. On the other hand, the 20keV cascade shown in Fig 1b generated fifty five Frenkel pairs. Many mono-interstitial dumbbells are located at the edge of the cascade region, but large interstitial clusters are also present.

Detailed analysis of the cascade evolution process shows that these large interstitial clusters are formed at the interface of local subcascades. For the 20 keV case, the large interstitial cluster results from agglomeration of three smaller sub-clusters driven by the very high local stresses in the cascade periphery. No vacancy clusters were formed in any of these events. This is in sharp contrast to the case of Au where large vacancy loops are observed for 10 and 30 keV cascades [9], but agrees well with results in V [10].

The number of Frenkel pairs normalized by the number calculated by the modified Kinchin-Pease (or NRT) model [11] is plotted in Fig. 2 as a function of PKA energy. For very low PKA energies such as 100 and 200 eV, the results agree well with the NRT model. For higher PKA energies, however, the NRT model clearly overestimates the number of Frenkel pairs. In the 20 keV recoil events, the defect production efficiency is only 30% of the NRT model, which is consistent with experimental studies in Cu and Ag [12] and with previous MD simulations in many metals for PKA energies up to 25 keV [2]. Note that these results are based on the number of defects at 10 ps after event initiation. In order to calculate the fraction of freely migrating defects at elevated temperature, we need to consider how many of these defects undergo recombination within their nascent cascade and how many are able to escape. This issue is addressed section 3.

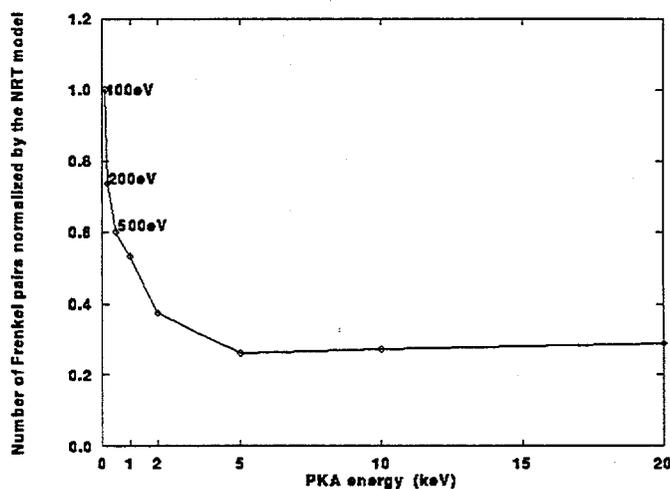


Figure 2. Defect production efficiency in α -Fe (Number of defects in MD cascades normalized by the NRT prediction) as a function of recoil energy

2.2 Vanadium

Vanadium-based alloys are strong candidate materials for the first wall of fusion reactors because of their low activation. It is therefore important to understand the form of the primary damage state in this materials in order to establish the conditions for further modeling of microstructure evolution under irradiation. We have carried out simulations of displacement cascades in V at energies from near threshold to 5 keV [10]. The simulations were carried out with the Johnson-Oh EAM-type potential [8] which was demonstrated to provide a good description of the formation and migration energies of vacancies, the melting point, and the fcc-bcc structural energy difference [10]. As for the case of Fe, the potential was modified at short range to account for scattering during the early collisional stages of the cascade.

The threshold energy for defect production was found to have a minimum value of 18 eV near the $\langle 100 \rangle$ direction and to increase to 80 eV 27% away from $\langle 111 \rangle$ towards $\langle 100 \rangle$ and 35% away from $\langle 100 \rangle$ toward $\langle 221 \rangle$. As in the case of α -Fe, very few interstitial and vacancy clusters were found for any of the recoil events simulated. The largest cluster contained four interstitials in one of the 5 keV recoil events. No vacancy clusters larger than two were found.

3. Kinetic Monte Carlo Studies

As discussed in the introduction, interstitials, vacancies and some of their clusters are mobile at elevated temperatures. Therefore, soon after production in the cascade these defects begin to migrate and interact. Some of them will escape their nascent cascade to migrate over long distances in the matrix, thus contributing to the formation and evolution of microstructures such as impurity precipitates and voids. The estimation of the production efficiency of such defects, commonly termed Freely Migrating Defects (FMD), is a key requirement for the development of a quantitative understanding of the effect of particle irradiation on materials properties. In this section, we report on recent calculations of the production efficiency of FMDs in α -Fe at 600 K using kinetic Monte Carlo simulations [1]. The input data necessary for the KMC simulations, such as defect diffusivity, defect cluster binding energies, and the initial defect configurations produced by displacement cascades, were obtained by MD. The displacement cascade results were discussed in section 2. The mobility of the various point defects and defect clusters were also calculated and will be reported in the literature. For completeness, suffice to mention here that we found that small interstitial clusters are highly mobile and migrate by executing a random walk in one dimension.

For a given PKA energy, the starting defect microstructure is introduced at the center of the KMC computation box according to the spatial configuration obtained by the MD simulation of the corresponding displacement cascade. Then, the system is annealed at 600K for 1000 s, and the number of defects that are able to reach the edges of the KMC computational box (which is a cube 0.1 μm on a side) are counted to determine the escape fraction. We consider clusters that contain between 3 and 15 interstitials to execute a one-dimensional random walk. This is consistent with the ideas of Trinkhaus et al [13,14], who have reported that small interstitial clusters can unfault to form glissile dislocation loops and therefore migrate in one dimension. We didn't consider the migration of small vacancy clusters because their binding energies are small compared with the migration energy of the mono-vacancy, and are therefore expected to dissolve quickly at 600 K.

A typical defect evolution time-history during a 600 K anneal of a 20keV cascade shows that free mono-interstitials begin to migrate at $t = 1 \times 10^{-10}$ s. This causes an increase in the number of interstitials in clusters and a decrease in the number of free vacancies through recombination. At 1×10^{-8} s, the number of

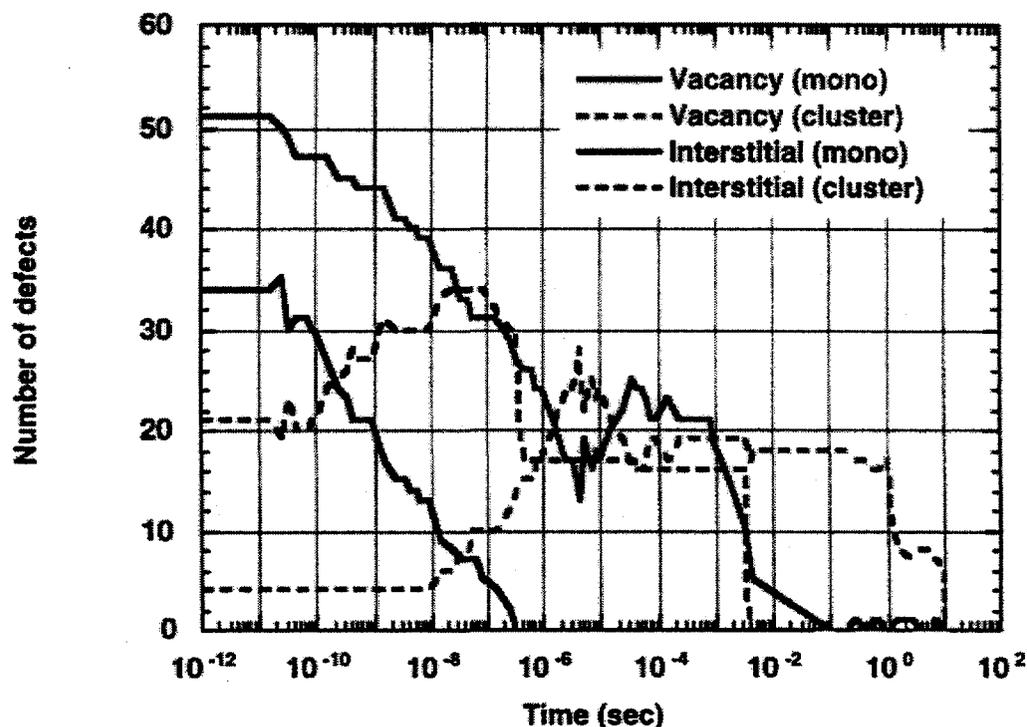


Figure 3. Time evolution of the defect microstructure produced by a 20 keV cascade in α -Fe during annealing at 600 K.

vacancies in clusters starts to increase, as a result of free vacancy migration. The number of free mono-interstitials becomes zero within 1 μ s due to either recombination, clustering, or migration out of the KMC box. A large decrease observed in the number of interstitials in clusters at times around 1 μ s and 10 ms arises from the escape of two large interstitial clusters from the box at these times. An increase in the number of free vacancies at 1×10^{-5} s is caused by dissociation of small vacancy clusters. Two large decreases in the number of vacancies in clusters at around 1 s and 10 s also arise from dissociation of vacancy clusters. The emitted mono vacancies from these clusters can migrate to the surface of the KMC computational box very quickly compared with the time scale of 1 s. All the defects annihilate by recombination or escape from the box within the first 10 s.

The results of these simulations show that a very small difference can be observed between the escape ratios of interstitials and vacancies for recoil energies greater than 10 keV. This is due to the formation of very large (>15) immobile interstitial clusters, an event which occurs with very small probability in all of the calculations performed here. For recoil energies less than 1 keV, the escape ratios of vacancies and interstitials are identical and exceed 80%. This value decreases to about 65% as the recoil energy increases to 20 keV. The sum of the escape ratios of mono- & di-interstitials, which can migrate three-dimensionally, and larger mobile interstitial clusters, which migrate one-dimensionally is the overall escape ratio of interstitials. For low recoil energies, the escape of mono- and di-interstitials is dominant. However, this tendency is reversed between 5 and 10 keV, and the escape ratio of mono- & di-interstitials becomes only

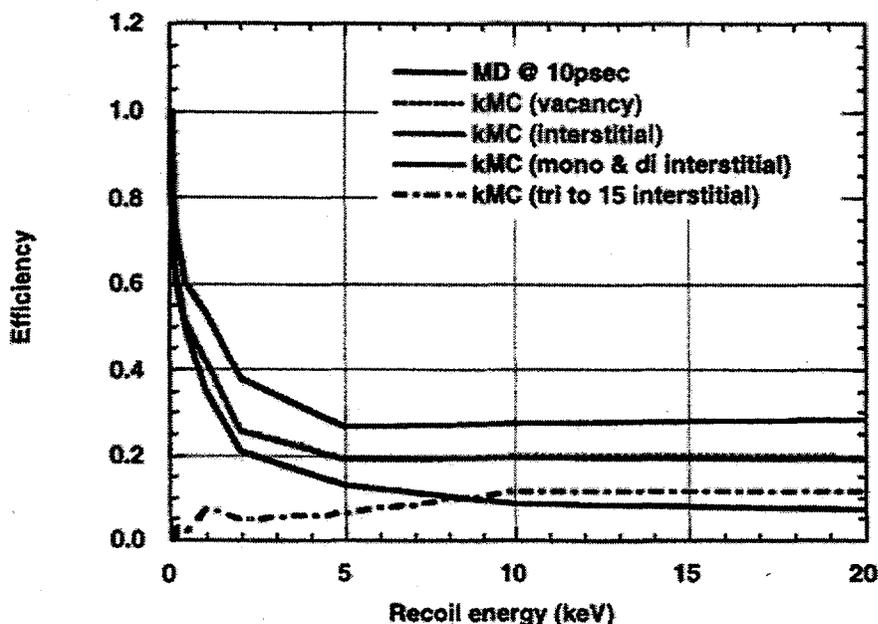


Figure 4. Freely Migrating Defect production efficiency in α -Fe during irradiation at 600 K as a function of recoil energy. At 20 keV, 60% of all defects produced escape their nascent cascade. The same number of vacancies and interstitials escape, but the 80% of the escaping interstitials do so as loops that glide in one dimension.

20% at 20 keV. On the other hand, all the vacancies escaping from the cascade are mono-vacancies, which migrate three-dimensionally. The FMD production efficiency is the product of the production efficiency in the cascade and the escape fraction, and is shown as a function of recoil energy at 600 K in Fig. 4. Total FMD production efficiencies for vacancies and interstitials decrease rapidly as the recoil energy increases, and saturate at about 20% of the NRT model predictions for the PKA energies larger than 5keV.

4. Summary

In summary, we have presented MD simulations of defect production in Fe and V at recoil energies typical from 100 eV to 20 keV. The simulations show that above 5 keV interstitials are produced in clusters with high probability, and that virtually no vacancy clusters result from these cascades. These results agree well with other MD simulations of cascades in α -Fe by Stoller et al. [15]. We have shown how the large local stresses within the cascade region can act to unfault small interstitial clusters resulting in the production of mostly glissile loops.

The MD results were used as input to KMC simulations of defect diffusion and interaction. The KMC simulations provide insight into the physical processes that take place within a few seconds following the displacement cascade. Our results show that for recoil energies less than 1 keV about 80% of the vacancies and interstitials produced in the cascade escape recombination. This value only decreases to 65% when the recoil energy increases to 20 keV. This high escape probability even in the case of dense displacement cascades is associated with the fact that in α -Fe many interstitials are produced in the form of small glissile loops, and the vacancies are all produced as isolated mono-vacancies and not as large collapsed Frank loops.

Two thirds of all escaping interstitials did so as gliding loops and only one third as three-dimensionally migrating self-interstitials. The production efficiency of freely migrating defects in Fe is 20% of the dpa standard for 20 keV recoils.

Work is under way to include the effect of sinks such as dislocations, grain boundaries, alloy components, and impurity atoms (e.g. He, C) in the KMC simulations of defect microstructure evolution. In addition, the role of inter-cascade annihilation (dose, dose rate) in the evolution of the system will be considered in the near future. Work is also under way to develop three-dimensional dislocation dynamics simulations to predict the changes in yield stress and strain hardening exponents as a function of irradiation time associated with the evolving microstructure as obtained by the MD/KMC simulations.

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