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A molecular dynamics study for ultrafast process of radiation damage in materials

K. Morishita [a,*], N. Sekimura [b], T. Diaz de la Rubia [c]

[a] *Nuclear Engineering Research Laboratory, The University of Tokyo, Tokai-mura, Naka-gun, Ibaraki 319-11, Japan*
[b] *Department of Quantum Engineering and Systems Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113, Japan*
[c] *Lawrence Livermore National Laboratory, The University of California, PO Box 808, Livermore, CA 94550, USA*

Abstract

Molecular dynamics calculations are performed to simulate displacement damage processes in irradiated metals. The interaction of atoms is described using the embedded atom method (EAM) potential that is modified at close atomic separation to merge smoothly with the universal ZBL-potential for description of high energy atoms. The displacement events of high energy atoms simulated here are initiated both from a single recoil atom with the kinetic energy of 20 keV in vanadium and from dimer recoil atoms of 2 keV in copper. The initial evolution of the displacement events is represented by replacement collision sequences (RCSs) and subsequent molten-structure at the center of the events. The monomer recoil event in vanadium shows a higher probability of defect production and lower probability of defect clustering, compared to the MD cascades in other metals. The dimer-recoil events in copper show a higher probability of defect clustering compared to a monomer-recoil event. © 1997 Elsevier Science B.V.

1. Introduction

One of the most important features in materials interacting with quantum beam is a successive atomic displacement event which is called 'a cascade' [1]. In the event, a large number of athermal lattice defects are created. The produced defects would play a very important role on microstructural changes in materials during the subsequent thermal interaction stage. However, the number of the athermal defects really produced during the displacement event is actually far from the standard estimation model established by Norgett, Robinson and Torrens (NRT) [2,3]. The NRT model just considers a simple extrapolation of the production mechanism of a single defect pair into higher energy regime; the number of the NRT-displacements is described as $\kappa E_D/2E_d$, where E_D and E_d are deposited damage energy and displacement threshold energy, respectively. The nascent recombination of defects with the first 10 ps of the cascades is ignored in the model. The difference in numbers between the model estimation and the real number of produced defects could become a critical problem in materials irradiated by high energy ions and neutrons, where the nascent recombination could be significant.

The evolution of the displacement cascades has recently become a focus of the research of materials science and technology. In the field of manufacturing of semiconductor device where the precise control of dopant distribution after ion implantation is required, all processes of the implantation of dopant atoms and the smoothing of surfaces can involve the development of cascades. In the field of materials design for fusion reactor components that could be irradiated by 14 MeV DT neutrons, high energy cascades are greatly expected to occur in the materials. In these research fields, it is very much important to establish the model for precise description of the number, size and morphology of defects produced due to the displacement cascades.

* Corresponding author. Present address: Research Institute for Applied Mechanics, Kyushu University, Kasuga, Fukuoka 816, Japan. Tel.: +81-92 583 7718; fax: +81-92 583 7690; e-mail: mori@riam.kyushu-u.ac.jp.

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2. Molecular dynamics model

A molecular dynamics (MD) method is a well-defined microscopic description of a many-body systems, computing the phase space trajectories of a collection of particles that individually obey classical laws of motion. By using the MD method, one obtains the dynamic properties of a system as well as static one. Due to recent advances in computational facilities and development of realistic potentials, this technique has become a practical tool for the many body problems in question. We applied this technique to the simulation of cascade events that take place in materials interacting with high energy particles. The cascade event takes place within 10 ps at nanometer length-scale in irradiated materials, so that it is very difficult to observe it directly by any experimental techniques at present. As a usual procedure of this kind of simulation, an atom in a well-equilibrated crystal is chosen to have some kinetic energies that are greater than the threshold value of atomic displacement. The atom is called a primary knock-on atom (PKA), initial kinetic energy of which is used as one of characteristic parameters for the events.

All the MD simulations shown in the present study are performed using the molecular dynamics code, MDCASK [4], the parallel version of which runs efficiently on a CRAY T3D MPP parallel computer at Lawrence Livermore National Laboratory's National Energy Research Supercomputing Center. The serial version of the code also efficiently runs on a HITAC S-3800 supercomputer at the University of Tokyo at a rate of about 1×10^{-5} s/atom/step. The serial version of the code is fully-vectorized at the supercomputer and the parallel version employs PVM message passing routines for communication between processors.

The simulations employ the isotropic many-body potentials of the EAM type developed by Johnson and Oh [5] for vanadium simulation and by Daw and Baskes [6] for copper simulation. This type of potentials are known to provide a realistic description of the energetic features of many metals. The potentials are modified to be splined to the universal potentials [7] of Biersack and Ziegler (ZBL) for description of high energy recoil atoms. Morishita and Diaz de la Rubia [8] show that the modified potential for vanadium can reproduce defect properties, melting temperature and displacement characteristics very well. The computational cells employed here have periodic boundary conditions and are well equilibrated at 10 K to establish an appropriate phonon mode prior to an initiation of displacement events.

3. Evolution of cascade damage process

Evolution of radiation damage processes in materials is defined depending on the time-constant of interaction of defects; the collisional, cooling, thermal and diffusional phases. The collisional phase is defined as a stage of atomic collision sequences, which begins with the first collision of an irradiating particle with target atoms, ending when no atoms have enough energy to create another stable displaced atom. During the collisional phase, the PKA energy is distributed by multiple collisions among many atoms, with the result that disordered regions are created. The cooling phase is defined as a stage of the dissipation of energies deposited locally during the collisional phase, ending when the cascade region becomes thermal equilibrium with its surroundings. After the cool-

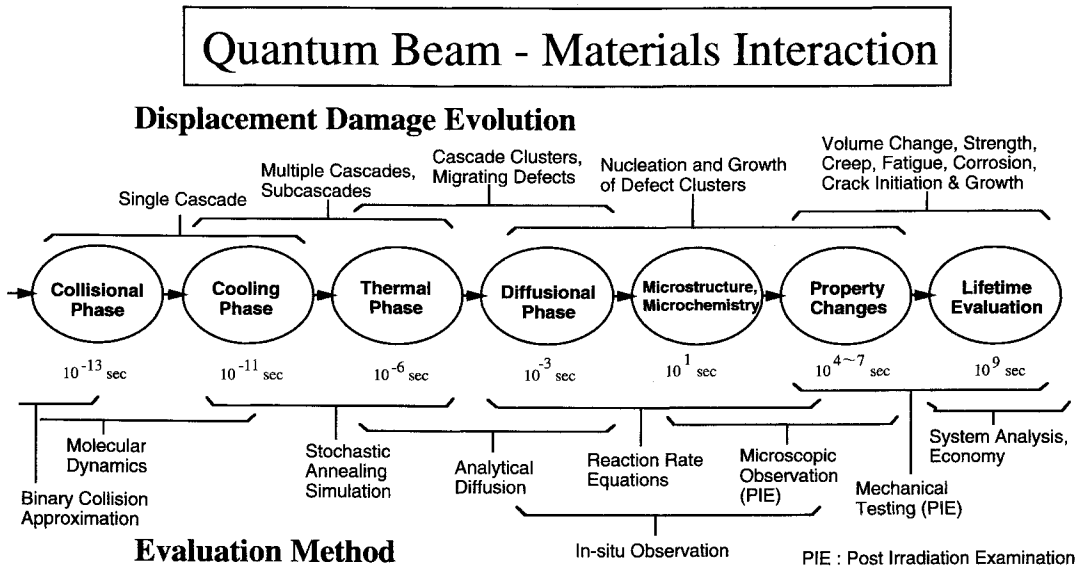

Fig. 1. Schematic representation of time evolution of interaction between high energy particles and materials [9,10].

ing phase, normal, thermally activated diffusion process of defects takes place. The thermal and diffusional phases are the stages of the thermal activation process of defects. The radiation damage processes [9,10] are schematically illustrated in Fig. 1.

Fig. 2 shows the time evolution of cascade damage processes in vanadium after the impact of a 20 keV recoil atom. The open and closed circles in the figure show a vacant lattice site and a displaced atom, respectively. The calculations are carried out at constant MD volume with 549250 atoms, which is large enough to simulate all the displacement effects initiated from a PKA of this energy range. The detail of the MD simulations, including the PKA energy dependence of the evolution, are shown elsewhere [11]. Some key features of cascade evolution, such as replacement collision sequences (RCSs) and local melting at the center of cascades, are observed in the figure. Fig. 2(c) shows defect configuration at the end of the collisional phase, during which replacement collision sequences (RCSs) mainly take place. The RCSs are first identified by Vineyard and co-workers [12] as the mechanism of the efficient separation of vacancy–interstitial pairs in irradiated crystals. During the subsequent cooling phase which continues for several picoseconds, the center of the cascades has a liquid-like disordered structure, as shown in Fig. 2. The possibility of local melting behavior

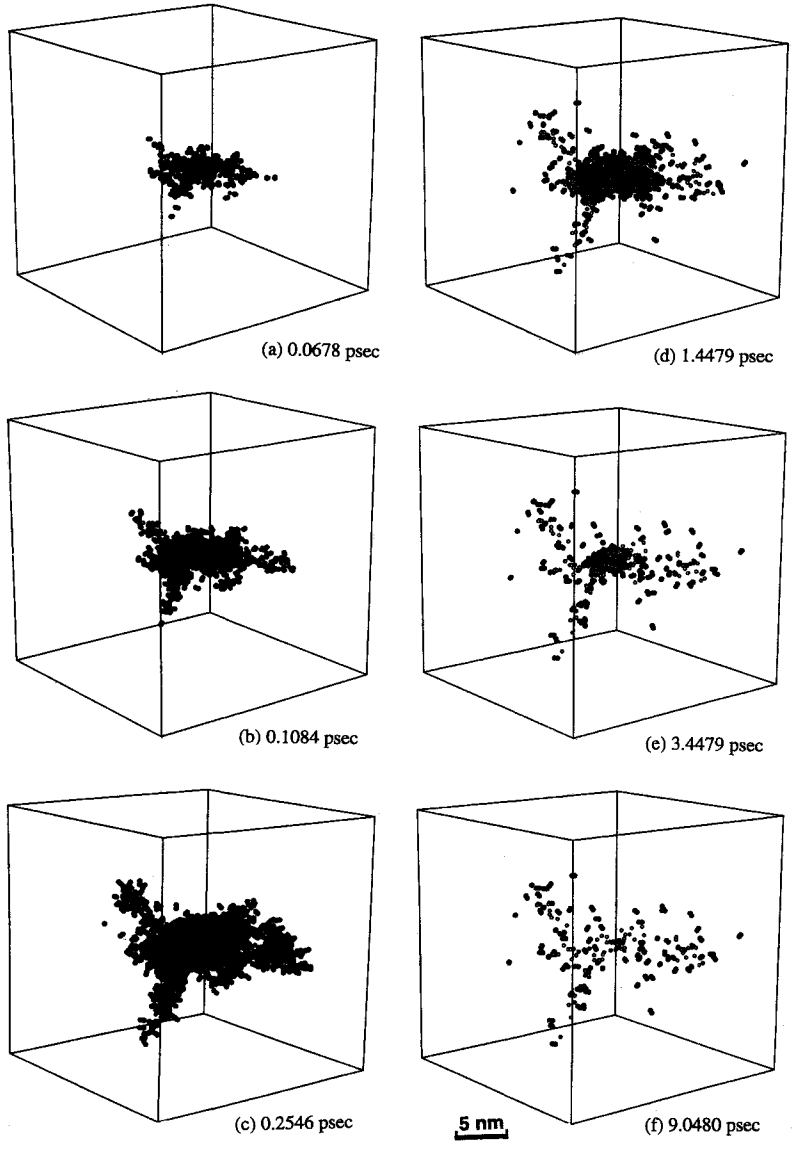

Fig. 2. Time development of ultrafast process of radiation damage in vanadium, that is initiated from a 20 keV recoil atom.

in cascades has been considered first by Seitz and Koehler [13], and was confirmed by MD simulation works of Averback and co-workers [14]. As shown in the figure, the majority of stable self-interstitial atoms (SIAs) are associated with the RCS that is the mechanism [15] of the efficient separation of an SIA from the melting zone at the center of the cascade. As the melt zone resolidifies, only SIAs outside the melt zone and an equal number of vacancies within the melt zone, can survive after the cascade cooling. During the cooling phase, the significant rearrangement of defects takes place including recombination and clustering. The number of defects survived after the cooling phase at the present 20 keV event in vanadium is only 35% of the NRT model estimation. The fraction is higher than those of MD cascades in other metals [16,17].

The direct formation of defect clusters in cascades is another interesting aspect of cascade evolution, since the clustering of defects in cascades could influence microstructural evolution during the subsequent thermal and diffusional phases, including a bias effect that is an difference in behavior between vacancies and interstitials [18,19]. Morishita and co-workers found [20] a criteria of direct formation of vacancy cluster in cascades in several pure metals from a correlation between ion-irradiation experiments and binary collision simulations. Foreman and coworkers found interstitial cluster formation as the result of ballistic ejection from the periphery of the cascade by their MD simulation work [21,22]. The present simulation results show that the production efficiencies of defect clusters (number of atoms ≥ 3) directly formed in cascades in vanadium are found to be about 10 and 4% of all the produced defects for vacancy- and interstitial-clusters, respectively. The efficiencies are much smaller than those of the MD results of several other metals [16,17]. The smaller efficiencies are consistent with experimental results [23] that show no vacancy cluster is observed by transmission electron microscopy (TEM) in self-ion irradiated vanadium.

4. Overlapping effect of cascades

As increasing recoil energies, the size of cascades becomes greater and it is finally broken up into some energy dense regions, each of which is called a 'subcascade'. The break-up energy is typically 30 keV in metals [24]. In the energy range above the break-up, interaction between the subcascades would be of interest for defect production.

We performed MD simulations to investigate cascade overlapping effects in copper [25,26]. Two PKAs are simultaneously introduced in parallel direction with kinetic energies of 1 keV each. The dimer-recoil dissociates on impact with enhanced energy density and vacancy supersaturations. In the present study, the number of defects produced in the dimer-recoil events in copper is evaluated as a function of distance between two PKAs, dx, ranging

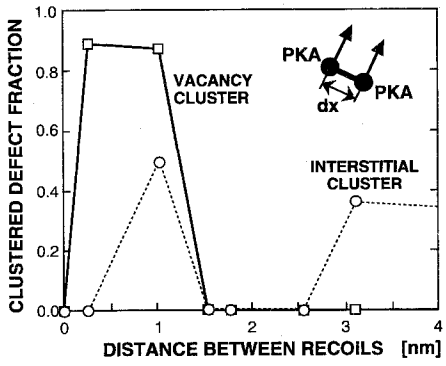

Fig. 3. The production efficiency of clustered defects compared to the NRT displacement at dimer recoil events in copper. Total kinetic energy of recoils is 2 keV.

from 0.25 nm to 3.10 nm. In a monomer-recoil event, SIAs lie outside the central core of cascade in general, while vacancies are distributed in a compact depleted zone at the center of the cascade. In a dimer-PKA event, however, overlapping of the effects of two cascades takes place; the regions of the vacancy–vacancy overlapping, interstitial–interstitial overlapping and vacancy–interstitial overlapping are created depending on the distance, dx. The range of the distance, dx, simulated here covers all the regions above. Fig. 3 shows the fraction of clustered defects to the number of the NRT displacement as a function of the distance, dx. The three kinds of overlapping effects are actually observed as follows. The fractions of clustered defects are exactly zero for both types of clusters at a monomer–recoil event. At dimer–recoil events, however, the fractions are varied depending on the distance, dx. In the event, $dx < 1.0$ nm, the vacancy–vacancy overlapping is dominated, where vacancy cluster is formed. In the event, $dx > 3.0$ nm, the interstitial–interstitial overlapping is dominated, where interstitial clustering occurs at the overlapping region. The intermediate event, $1.0\ \text{nm} < dx < 3.0$ nm, creates the vacancy–interstitial region, where different types of defects are recombined with each other with a result of smaller number of defects survived.

More interestingly, both types of clusters are produced at the event, $dx = 1.0$ nm. In this event, the displaced atoms produced in one cascade (named 'A-cascade') are recombined with the vacant sites produced in another cascade (named 'B-cascade'), with a result of an efficient separation of the vacant sites of the A-cascade and the displaced atoms of the B-cascade. This efficient separation mechanism in the dimer–recoil event would promote the clustering efficiencies of both types of defects, as shown in the figure.

5. Summary

We have reported the results of MD simulations for the atomistic behavior of materials interacting with high en-

ergy particles during the first 10 ps. The mechanism of defect production in cascades, as shown in the MD simulations, is not reflected within the framework of the NRT model estimation, where the number of produced defects is estimated based on the simple extrapolation of 'one defect production mechanism'. Actually, only 35% of the NRT displacements are produced as defects at the 20 keV event in vanadium. Moreover, the dimer–recoil event makes the estimation more complicated. Our dimer–recoil events in copper show a higher probability of defect clustering compared to that of monomer–recoil events. The MD simulation studies will enable us to establish a new estimation model beyond the NRT description for better estimation of produced defects in cascades.

In this kind of simulation technique, however, there still exists a problem of the reliability of interatomic potentials employed in calculations. We need to compare simulation results directly with experiments. At present, however, the experimental techniques may provide just a 'debris' of the cascade evolution. The complexities of observing cascade processes in real materials are due to the swiftness ($\sim$ 10 ps) and smallness ($\sim$ 10 nm) of the phenomena. A sub-picosecond X-ray analysis using a sub-picosecond electron pulse linear accelerator could conquer the difficulty in the swiftness, and would enable us to observe directly the ultrafast dynamic process of cascades in the near future [27].

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