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Molecular Dynamics Simulations of the Radiation Damage in α-Iron

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Abstract

The method of molecular dynamics simulations has been used to investigate the development of displacement cascades in iron. Radiation damage caused by fast neutrons or ions results from the creation of energetic primary knock-on atom (PKA), which in turn produces displacement cascades. Cascades of 2 and 4 keV in energy have been simulated by molecular dynamics code, MOLDY, in α-iron at 290^{o}C. In order to investigate the damage evolution processes, the changes in cascade morphology during the collisional process are observed. And, we quantitatively analyzed the primary damage state, of which two parameters are taken into account: (1) the number of isolated point defects and (2) the distribution of point defect clusters formed following the collisional process. The simulation reveals that interstitials tend to form clusters whereas vacancies remain as isolated defects in α-iron.

1. Introduction

Many structural components in light water reactors (LWRs) are subject to neutron irradiation. It is well known that irradiation affects the microstructure and property of materials, which might degrade the integrity of structural components and cause safety problems. It is, therefore, a big concern to evaluate the end-of-life behavior of internal structures of currently operating LWRs in the nuclear industry.

Most problems of irradiated materials originate from the atomic collision between the energetic particles and lattice atoms. This collision leads to the atomic displacements through the energy transfer reaction and results in various types of point defects including vacancies, interstitials, and their clusters. The lattice atom first struck and displaced by the projectile is called the primary knock-on atom (PKA). This PKA possesses a certain amount of kinetic energy sufficient to bring about further atomic displacements, which is called a displacement cascade. The production and behavior of point defects created in the cascade are of importance because they play important role in the microstructural evolution and material property. Inherently, the effect of irradiation on materials is a multiscale process, spanning a wide range of time and dimension scales. A cascade generated by a PKA generally occupies a region about several nm in diameter and takes place in a time scale of the order of several ps. The microscopic changes in materials such as the number and configuration of the point

defects within several ps are termed as primary defects. The radiation damage is closely linked to the state of primary defects.

It is worth describing the evolution process of cascades with time from the initial collision between PKA and lattice atoms. Cascades can be largely divided into two stages as they develop with time [1]. The first is a ballistic phase lasting about a few tenths of a ps. During this period, the PKA energy is distributed to neighbor atoms by multiple collisions, which results in the creation of a disordered region. This region is composed of a central core surrounded by displaced atoms. The thermal spike phase follows in several ps, during which the disordered cascade region cools to the temperature of its environments. Most of the displaced atoms in the outer regions return to regular lattice sites by athermal relaxation. Other atoms that are displaced from the normal positions make a contribution to the radiation damage to materials.

It is impossible to investigate the primary damage through experimentation in a quantitative way. With the advance in computer capability and the development of many-body potentials, however, the use of molecular dynamics (MD) method enables us to simulate the displacement cascade on the atomic scale. In this work, we have used the MOLDY code[2] as a tool to study the structure and dynamics of displacement cascades in α-iron occurring under LWR environments (290^{o}C). Cascades of 2 and 4 keV in PKA energy are simulated by this code at 290^{o}C. These PKA energies are important to neutron spectra of concern. The emphasis is placed on the computation results obtained from a model of α-iron, which has the bcc crystal structure, because of the importance of ferritic steels to reactor technology. Using the graphic software, the morphology (atomic arrangements) of cascade evolution is visualized. This study has given us an opportunity to theoretically quantify the primary defects and to provide the basis of multiscale modeling for the radiation damage study.

2. Molecular Dynamics Simulations

Computer simulation by MD is a useful technique for investigating the defect production mechanism in displacement cascades. This method is relevant in that the time and dimension scales for a displacement cascade are too small to be open to experimentation. Development of mathematical modeling and rapid growth in computer capabilities make it possible to simulate the atomic behavior occurring within a picosecond. Of particular interest in this study is the demonstration of MD code, MOLDY. Particular features of the MOLDY code are described first. Then, the calculation results are presented concerning the evolution of defects in a displacement cascade as a function of PKA energy.

2.1 General Description of MOLDY code

The program used here was the MOLDY code of Finnis[2], which we modified to run on PC. This code is widely used for simulations of displacement cascades in metals. The code, written in FORTRAN, uses an atom block with periodic boundary conditions. The main function of MOLDY is to integrate the classical equations of motion (Newton's law) by a Gear 4-value predictor-corrector algorithm. The many-body interatomic potential for a-iron is embedded in the MOLDY code, which was derived by Finnis and Sinclair [3]. They have formulated an empirical many-body potential for transition metals for the purpose of atomistic simulation. The model energy consists of two terms; (1) a cohesive potential, which is the square-root of a site density summed over neighbor atoms and (2) a repulsive pairwise potential, which represents the repulsive interaction between ions in terms of interatomic distance. Both potentials are assumed to be short-ranged and are parameterized to fit such

material properties as the lattice constant, the elastic constants, and cohesive energy.

Prior to conducting the cascade simulations, a block of atoms is equilibrated at a given temperature for about 10 ps to obtain a phonon equilibrium mode. Then, the cascade simulations are initiated by imparting a kinetic energy to an atom in a particular direction. The atom corresponds to the PKAs following the initial collision with fast neutrons. The size of an atomic block, which is the total number of atoms to be simulated, is determined by the PKA energy. Since the computing time with MOLDY is almost proportional to the number of atoms in the simulation, higher energy cases require a larger atom block [4]. In the present study, 128000 atoms are used as an atom block for the PKA energy of 2 and 4 keV cascades. The atom block size is $40a_o \times 40a_o \times 40a_o$, where a_o is the lattice constant of α-iron. The cascade simulations last until intra-cascade recombination of point defects is completed. Due to the high temperature in this simulation, the residual number of point defects fluctuates to a small extent after 10 ps simulation. All simulations have been carried up to about 10 ps under the assumption that the cascades reach the final stage after 10 ps. Although we extended one cascade simulation up to 20 ps, no differences could be seen. It is, generally, a rule of thumb to calculate the representative parameters from the repeated MD simulations in the given conditions by averaging output results.

2.2 Displacement Cascade Simulations

Eight cascade simulations were performed to observe the atomic behavior in α-iron at 290^{o}C up to about 10 ps. Two values of PKA energy, 2 and 4 keV, are chosen for comparing the calculation results. In fact, the average PKA energy for the commercial pressurized water reactor is in the range of several keV around the reactor pressure vessel inner surface. For the given PKA energy, two identical computations were performed with the specified initial PKA direction. Two initial PKA directions are considered, which are high-index direction [135] and low-index one [111]. In order to avoid channeling, referring to the long-distance displacement of an energetic atom down the open direction in the lattice, the use of high-index direction is recommended in the simulation [5].

The parameters of primary interest in this study are: (1) the number of isolated point defects and (2) the distribution of point defect clusters (PDC) formed following the collisional phase. The former defects are also termed as the freely-migrating defects (FMDs). FMDs are free to migrate over substantial distances that are large compared to nascent cascade size. Important property changes that happen to materials in reactor environments are driven by FMDs[6]. The microstructural changes related to property changes include void swelling, radiation-induced segregation, radiation-enhanced diffusion, and radiation embrittlement. The formation of PDC is also significant in that these small clusters provide embryos for the growth of larger defects, which affects mechanical property changes. The classical nucleation of defect clusters through the diffusive reactions is a time-consuming and slow process. This implies that the direct formation of small PDCs can be developed into the extended defects rapidly.

It is known that during the collisional phase of cascade, many atoms are displaced from the lattice sites temporarily and most of them disappear by reordering and/or recombination. This is illustrated in Figures 1, which show six stages in the formation and relaxation of 4 keV cascade in α-iron lattice at 290^{o}C. The initial direction of PKA is [135]. These plots were produced using PvMOL^{TM} program. Seen from Figures 1, the number of point defect reaches peak at time t = 0.25 ps. Thereafter, recombination of interstitials and vacancies starts to take place, which is a rather gradual process relative to a ballistic phase. Due to the high temperature, the number of point defects dose not reach constant values until 10 ps and

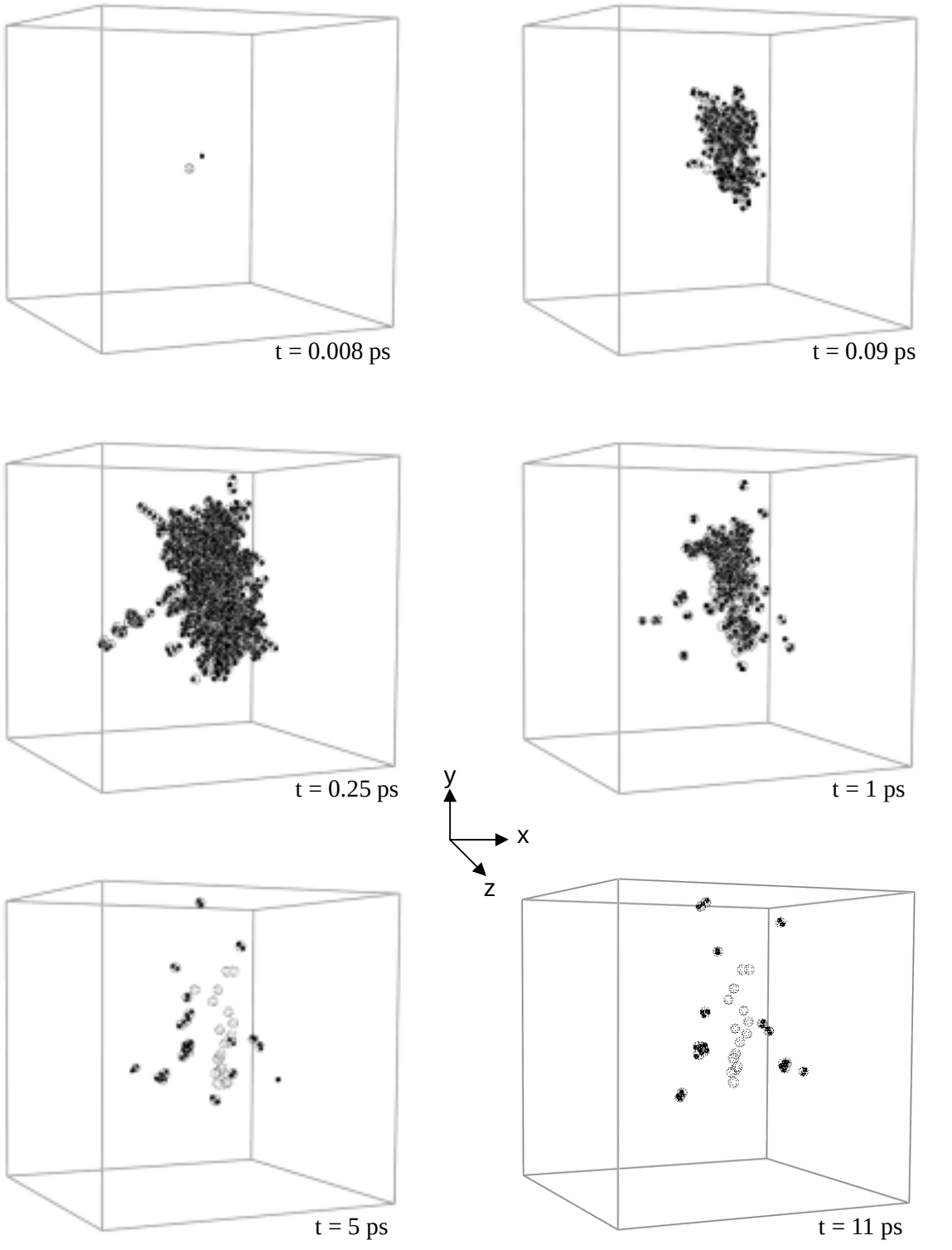

Figure 1. Illustration of displacement cascade evolution from a 4 keV PKA simulation in α-iron at 290°C. The size of the atomic block is $40a_o \times 40a_o \times 40a_o$ (a_o: lattice constant) and the initial PKA direction [135]. Filled squares represent interstitial atoms and empty ones vacancies

the survived defects, which take the form of either isolated or clustered defects, move through a diffusive process. However, since the number of point defects remain almost constant after t = 10 ps with a small amount of scatter, the number of point defects calculated at the final step of calculation was regarded as the final defects, in which the time elapsed is well over 10 ps. Only 32 interstitials remain at the end, of about 1200 displaced atoms at the peak of the cascade, which is the same case with vacancies. It can also be seen that the interstitials at the end are rather dispersed in the form of small clusters around the cascade core region while the vacancies tend to be located in the cascade core as an isolated type.

It is instructive to investigate the variation of number of point defects with time elapsed as a function of the initial PKA direction and its energy, which is plotted in Figure 2. It can be seen that beyond the peak, the number of point defects declines rapidly through recombination. Not significant differences in the final number of defects were found depending on the initial PKA direction. The PKA energy is the most influential factor to determine the number of point defects at the peak and at the end, as well as the cascade morphology. The high PKA energy brings about the wide range of displacement cascade events and the high fraction of defect clustering. The details of the analysis of final defects will be described in the next section.

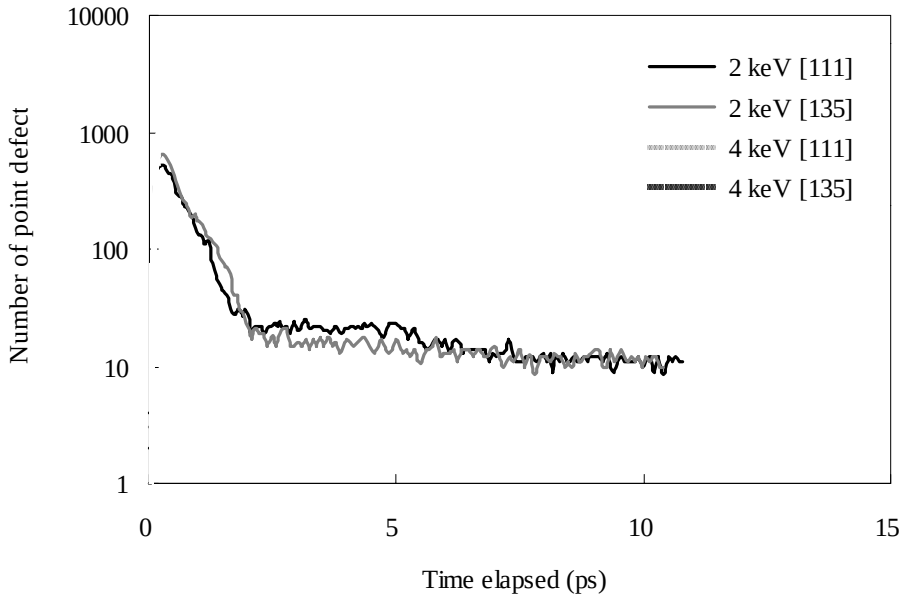

Figure 2. Variation of the number of point defects with time elapsed for four cascades at 290°C. The simulation PKA energies are 2 and 4 keV with different initial PKA directions of [111] and [135].

3. Analysis of Final Defect State

Various parameters were produced throughout the simulation and the results examined using the graphic program. The ability to analyze the final defect state for examination and to obtain data for model development is essential for this type of research. For the purpose of this, we will analyze the final defect state. Emphasis is place on identifying the type of defects and quantifying them.

The simulation results are summarized in Table 1, which includes the time to reach

Table 1. Summary of simulation results in α-iron at 290°C.

PKA energy	Initial PKA direction	Time to reach peak in defects (ps)	Peak number of defects	Number of surviving defects	Time at the end of simulation (ps)
2 keV	[111]	0.25	536	11	10.8
		0.23	586	21	10.5
	[135]	0.30	644	10	10.4
		0.24	500	20	10.3
4 keV	[111]	0.36	1484	32	12.2
		0.27	1145	34	11.9
	[135]	0.32	1249	34	11.6
		0.25	1170	32	11.4

the peak number of point defects, the maximum number of point defects, the number of surviving defects, and the time at which surviving defects were measured. It is seen that there is a big scatter of number of surviving point defects for 2 keV cascades, ranging from 10 to 20 defects in number. Averaging over the four results of 2 keV cascades and comparing with 4 keV cascades, the number of surviving defects generated for 4 keV cascades is as twice as that for 2 keV.

It is also of our interest to investigate the distribution of point defect clusters at the end of simulations. The interstitial cluster is defined such that within it, every self-interstitial atom (SIA) has at least one nearest-neighbor SIA. The vacancy cluster is also defined in a similar way. The distribution of interstitial clusters produced by eight cascades is shown in Figures 3. The MD results from 2 keV cascades are shown in Figure 3 (a) and those from 4 keV cascades in Figure 3 (b). Various size of interstitial cluster is distributed, ranging from single to octa-interstitial. For higher energy cascades, however, larger interstitial clusters could be found. Generally, interstitials tend to be existent in the clustering form rather than the isolated defects. And, the ratio of single defects to total surviving defects is less than 50%. It is probable that these clusters will be developed into dislocation loops. In case of vacancy defects, higher proportion of surviving defects remain as single vacancies and a smaller fraction is taken up in clusters. And, most of clusters are composed of small number of defects, which is dominant in a small PKA energy cascade. These distributions are shown in Figures 4. Lower energy PKA cascades tend to preferentially generate single vacancies and to create smaller size of clusters. We could see outlier deviated from the general behavior in the cluster formation. For example, hepta-interstitials are created from 2 keV cascade and deca-vacancies from 4 keV cascade. Although its probability is low, such an event might happen at high temperature in the displacement cascade, which is one of the random processes. The possibility of producing large size of PDCs directly from cascades makes it difficult to predict the microstructural behavior under irradiation. However, at least six different cascades are required to obtain statistically representative parameters for the given PKA energy and temperature [4].

4. Discussion

The purpose of this paper is primarily to demonstrate that the MD cascade simulations

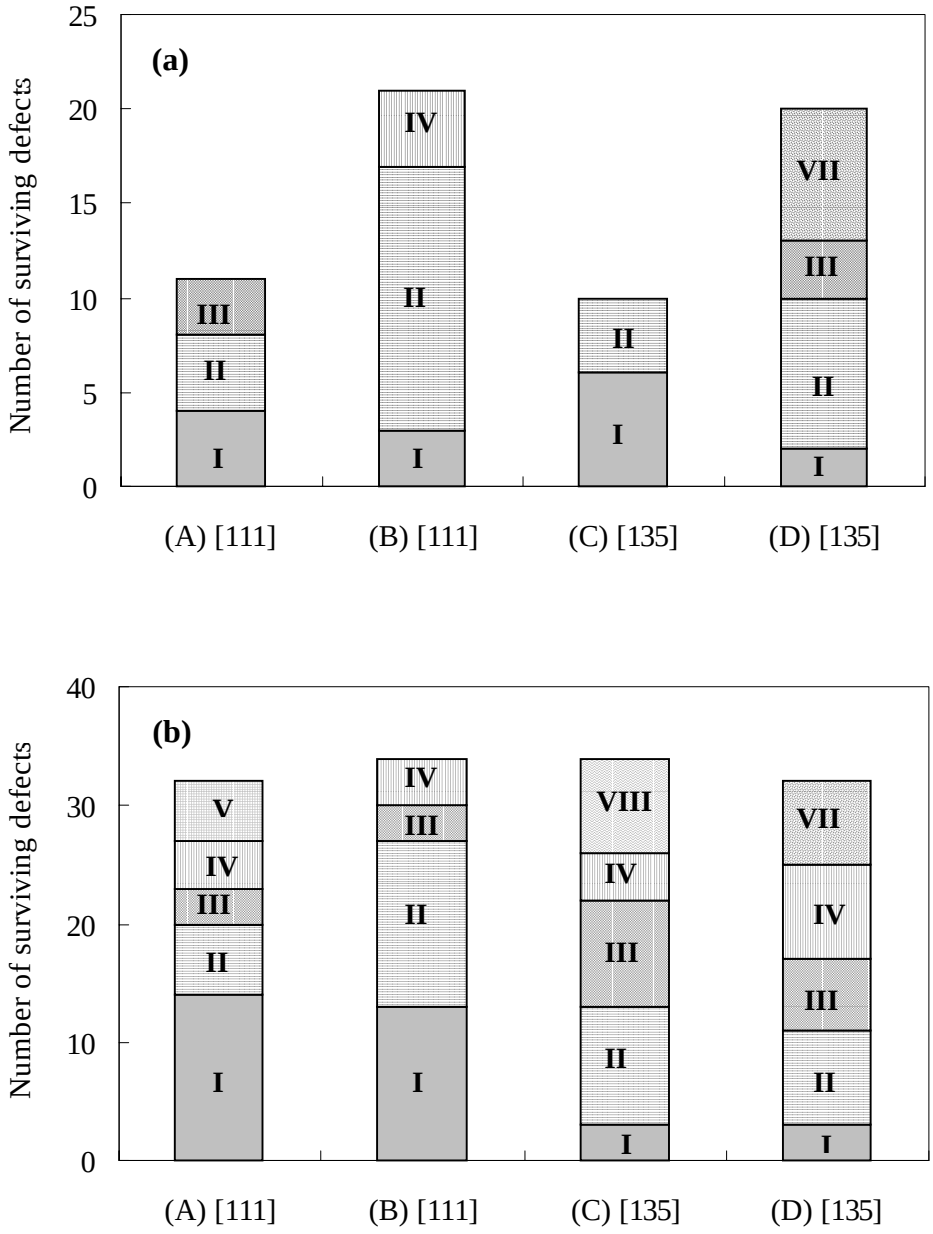

Figure 3. Interstitial cluster spectrum for α-iron at 290°C at the end of an MD simulation. Roman numerals inside the box stand for the size of interstitial clusters (**I** : single interstitial, **II** : di-interstitial, **III** : tri-interstitial, etc.). The number in the bracket on the abscissa is the initial direction of PKA. (a) 2 keV Iron cascade and (b) 4 keV Iron cascade.

provide a convenient way in the radiation damage study. A displacement cascade initiated by a PKA advances in a small region less than 10 nm across in a time scale of the order of ps. Such an event can cause changes in extensive atomic arrangements and subsequently result in the highly disordered structure. The MD-based primary damage evaluation can be applied to the quantitative prediction of radiation damage, which is relevant to LWR pressure vessel steels [7]. In predicting the amount of radiation hardening of ferritic steels, the results obtained

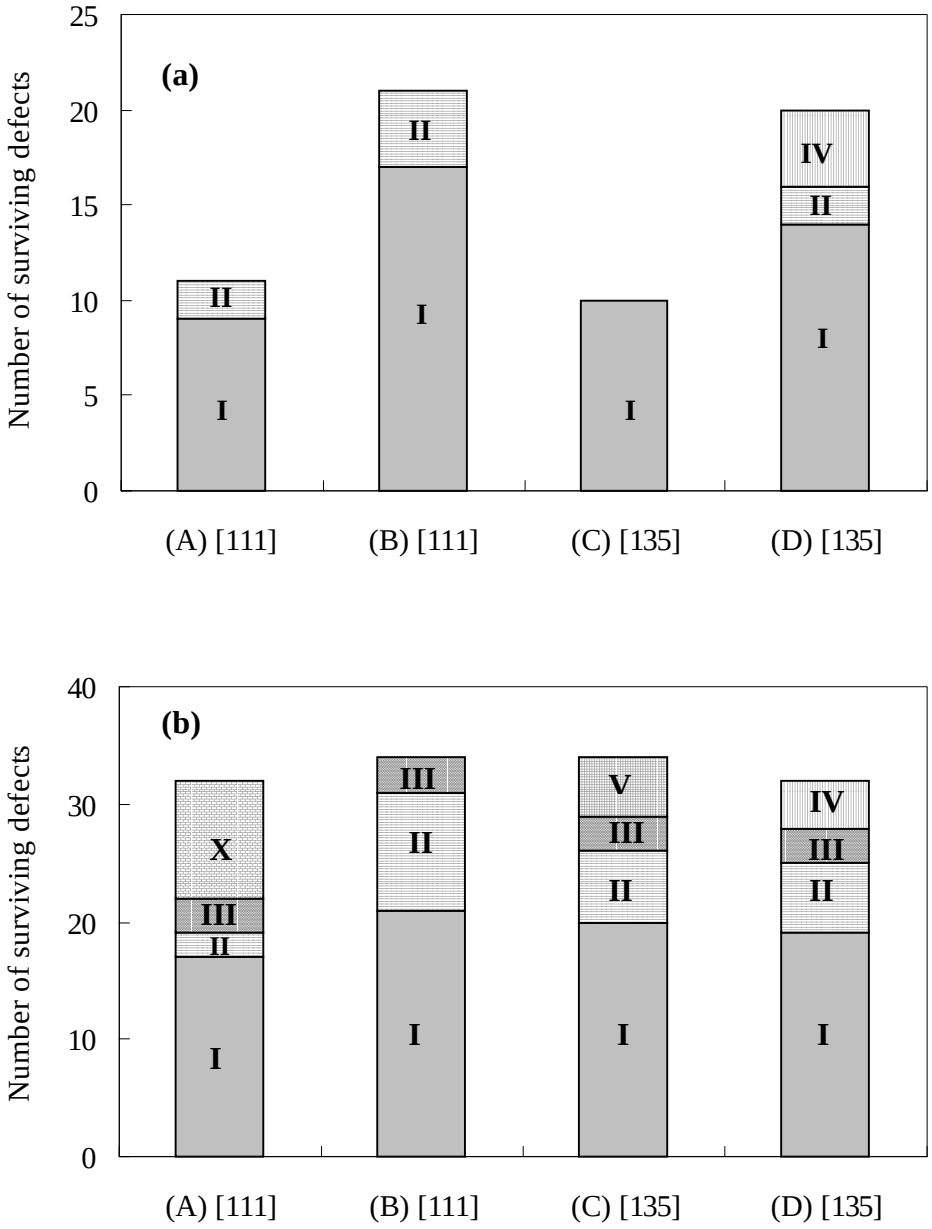

Figure 4. Vacancy cluster spectrum for α-iron at 290°C at the end of an MD simulation. (a) 2 keV Iron cascade and (b) 4 keV Iron cascade.

from the MD simulations are used as input parameters [8,9]. The two parameters of interest are: the number of FMD and the in-cascade clustering fraction, which are shown in Figure 3 and 4. Since these parameters are primarily dependent on the PKA energy and temperature for given materials, the use of MD simulation is essential. More MD simulations are under consideration, which correspond to the LWR operating conditions in order to predict the radiation hardening. In addition to the determination of damage parameters, the MD simulations also provide insight into the details of primary defect production.

In theoretically predicting the irradiation effect on materials, the multiscale modeling approach has been established to complement test reactors [10]. This multiscsale modeling methodology includes MD simulations, kinetic Monte Carlo methods, and reaction rate theory. Integrating each method over the relevant dimension and time scales enables us to model the behavior of radiation-induced defects and microstructures, and to quantitatively predict the property changes in materials exposed to radiation. MD simulations provide information on the primary damage state and physical insight into cascade processes, as described in this paper. Emphasis will be placed on the improvement of MD simulation methods and the extension to treating irradiation effects in alloying systems.

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