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<Technical Note>

Effect of Neutron Energy Spectra on the Formation of the Displacement Cascade in α-Iron

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Abstract

This paper describes a computational approach to the quantification of primary damage under irradiation and demonstrates the effect of neutron energy spectra on the formation of the displacement cascade. The development of displacement cascades in α-iron has been simulated using the MOLDY code - a molecular dynamics code for simulating radiation damage. The primary knock-on atom energy, key input to the MOLDY code, was determined from the SPECTER code calculation on two neutron spectra. The two neutron spectra include; (i) neutron spectrum in the instrumented irradiation capsule of the high-flux advanced neutron application reactor (HANARO), and (ii) neutron spectrum at the inner surface of the reactor pressure vessel steel for the Younggwang nuclear power plant No. 5 (YG 5). Minor differences in the normalized neutron spectra between the two spectra produce similar values of PKA energy, which are 4.7 keV for HANARO and 5.3 keV for YG 5. This similarity implies that primary damage to the components of the commercial nuclear reactors should be well simulated by irradiation in the HANARO. Moreover, the application of the MD calculations corroborates this statement by comparing cascades simulation results.

Key Words : primary damage, displacement cascade, molecular dynamics, MOLDY code, primary knock-on atom, SPECTER code, high-flux advanced neutron application reactor (HANARO)

1. Introduction

The effects of neutron irradiation on materials are largely dependent upon two factors; the neutron energy spectra and the irradiation time. For a long period of neutron irradiation to materials, radiation damage can be evaluated experimentally. For example, we can measure the changes in mechanical property from tests and investigate the microstructural changes through electronic microscopy. However, radiation damage is an inherently multiscale phenomenon which involves processes spanning a wide range of time and dimension scales. It is impossible to

cover the whole range of radiation damage through experiments. For this, modeling is required for particular conditions which are not accessible by experiments such as the smallest scales (nanometer and picosecond).

Most damage caused by fast neutrons results from the creation of displacement cascades. The initial interaction of a neutron with a lattice atom can lead to the production of energetic recoils, which are called primary knock-on atoms (PKAs). The energy transfer reactions between PKAs and lattice atoms bring about a series of displacements, taking place in a timescale of the order of 0.1 ps within a region of about 10 nm in diameter [1]. The resulting displacement cascades produce non-equilibrium point defects and their clusters, which are fundamental defects present in the primary damage state. Such defects diffuse over macroscopic lengths and participate in the formation of extended defects, altering the material microstructure. Changes in the microstructure affect the mechanical property, including radiation hardening, radiation embrittlement, etc. Accordingly, the key to understanding mechanical property changes during irradiation depends on the information on the primary damage state.

Progress has been made in computational modeling for the displacement cascade simulations to investigate primary damage using the method of molecular dynamics (MD) [2,3]. MD simulations provide a detailed picture of the evolution of the displacement cascades within a short period of time. This is a valuable technique for investigating the mechanisms of defect production since physical-length and time scales are too small to measure and the quantification of primary defects is essential to establish the multiscale modeling methodology. The method of MD has been applied in simulating displacement cascades in irradiated materials and rapid advances in computing have enabled us to examine higher energy cascades with a number of atoms. The radiation damage parameters of primary interest, obtained from the MD simulations, include: the number of isolated defects and the fraction of the surviving point defects contained in clusters. These parameters can be used as inputs to other calculations, kinetic Monte Carlo and/or reaction rate theory [4,5].

In this paper, we present some of our preliminary work in the field of radiation damage study currently underway. First, using neutron spectra calculated previously, the basic radiation damage parameters are determined from the SPECTER code [6], which include dose rate (dpa/s), gas production, PKA spectrum and its average energy, etc. Then, with the spectrum-averaged PKA energy, we run the MOLDY code [7] and investigate the evolution of radiation defects in α-iron. Two different neutron spectra obtained from the research reactor and the commercial nuclear reactor were employed to examine the difference in primary damage states. This technical paper has given us an opportunity to quantify the primary defects for the given neutron spectrum and to provide the basis of multiscale modeling to the radiation damage study.

2. PKA Energy Spectra Calculation - SPECTER Code

It is known that a PKA is created by means of various nuclear reactions. The PKA spectrum is determined by such factors as incident neutron energy, masses involved, and the angle between the incident neutron direction and the recoil direction [8]. We can readily obtain the PKA spectrum in various elements for a given neutron spectrum from the SPECTER code calculation [6]. The SPECTER code contains libraries of pre-processed cross sections for atomic displacements

and gas production, as well as atomic recoil energy distributions, on a specified 100-point neutron energy grid. Accordingly, for a given neutron energy spectrum, the code simply calculates the spectral-weighted radiation damage parameters by converting the master library files into the user group structure. This code provides a convenient tool to obtain basic damage parameters resulting from neutron irradiation. However, the SPECTER code does not account for burnup, which might affect the displacement reactions significantly when the transformed elements produce displacements at a rate comparable to the original elements. In this case, time-dependent neutronics calculations should be made first and then the SPECTER code can be employed for damage calculation.

In order to find out the effect of neutron energy spectra on the basic radiation damage, two sets of neutron spectra, obtained from the neutron transport calculations, were used in this study. The first data represents neutron spectra at the IR hole in HANARO [9], which is located near the core center. In fact, various kinds of alloy, sealed with the instrumented irradiation capsule, have been irradiated and tested in the HANARO facilities for investigating the radiation effect on materials in a short period of time. This neutron spectrum for HANARO was computed by the MCNP-4A code with 94-neutron energy group. The details of neutron transport calculations are described in Ref. [9]. Generally, the surveillance program is underway periodically in the commercial light water reactors (LWRs) for monitoring the degree of irradiation embrittlement of reactor pressure vessel (RPV) steel. This program evaluates the integrity of RPV steel by examining the irradiation conditions and property changes to irradiated steels. In particular, the neutron spectrum at the inner surface of RPV is an important factor to affect the lifetime of RPV. Thus, we adopt the neutron spectrum at the RPV inner-surface in the YG 5 nuclear power plant as sample input to the SPECTER [10]. It is of primary interest to compare differences in radiation damage parameters for iron between HANARO and YG 5. Although RPV steel is composed of various elements except iron, such elements as Ni, Mn, and Mo, do not affect the displacement damage significantly because of similar level of atomic weights and small volume fraction. Hence, this study only deals with pure iron (bcc iron) as a target material.

The normalized neutron spectra for HANARO and YG 5 are plotted in Figure 1, which clearly shows the relative amount of neutron flux as a function of neutron energy. Total neutron flux for HANARO is sixty times as high as that for YG 5. Although the spectral shapes for both cases are quite different in the low energy region (E_n < 1 eV), the low energy neutrons are not influential in inducing displacement reactions. For LWR, the strong moderation due to neutron scattering from hydrogen in the coolant water leads to a relatively higher fraction of low-energy neutrons. The fraction of high-energy neutrons (E_n > 1 MeV) for HANARO and YG 5 is 25 % and 22 %,

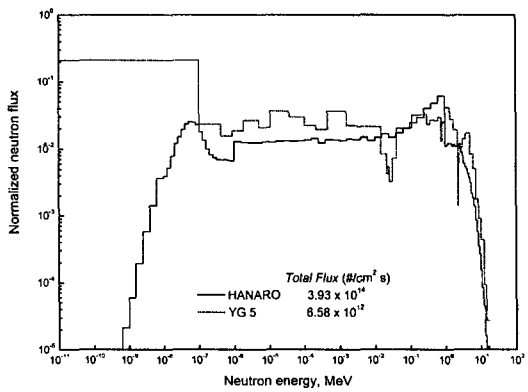

Fig. 1. Energy Dependence of Normalized Neutron Flux in Two Irradiation Environments: HANARO (research reactor) and YG 5 (commercial pressurized water reactor)

Table 1. Basic Radiation Damage Parameters of Iron for HANARO and YG 5. Results Obtained from the SPECTER Calculation

	Displacement rate (dpa⁺/s)	Hydrogen production (appm*/s)	Helium production (appm*/s)	Average PKA energy (keV)
HANARO	1.04×10^{-7}	4.77×10^{-7}	2.50×10^{-8}	4.7
YG 5	1.83×10^{-9}	1.01×10^{-8}	7.06×10^{-10}	5.3

⁺dpa : displacement per atom

*appm : atomic parts per million

respectively. The basic radiation damage parameters are obtained from the SPECTER code calculations on two neutron spectra. The damage parameters for iron such as displacement rate, gas production rate and average PKA energy are listed in Table 1. Since the irradiation time was set to be one second, differences in neutron flux are manifested as differences in radiation damage parameters. The displacement rate and gas production rate are dependent on irradiation time as well as neutron spectrum, whereas the PKA distribution is not related to irradiation time. It should be noted, however, that the spectral-averaged PKA energy of α-iron for YG 5 is greater than that for HANARO, albeit a little different - 5.3 keV for YG 5 vs. 4.7 keV for HANARO. This can be rationalized by comparing

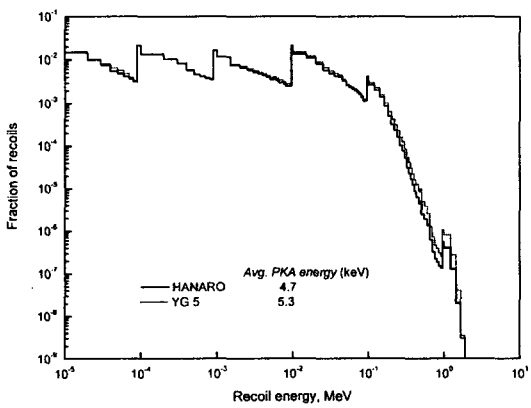

Fig. 2. Normalized Iron PKA Spectra for HANARO and YG 5. The Spectral-averaged PKA Energy is 4.7 and 5.3 keV, Respectively

the PKA recoil spectra generated by the SPECTER code. The PKA recoil spectra for iron in HANARO and YG 5 are shown in Figure 2. It is seen that more PKAs are distributed in the higher energy region (> 0.1 MeV) for YG 5. It is expected that there would be no substantial differences in primary damage when comparing the PKA energy distribution and its average energy. The cascade simulations will be performed with these energy values in the following section.

3. MD Cascade Simulations-MOLDY Code

As stated previously, the MD method is an appropriate technique for simulating displacement cascades. The rapid growth in computer modeling makes it possible to simulate the atomic behavior as a function of several variable such as temperature, PKA energy etc. The physics of primary defect production in displacement cascades has been extensively studied by a number of groups using MD techniques [1-5]. It should be pointed out that there are some limitations to the MD methods in characterizing the radiation defects. The cascade simulation of high energy PKA (greater than 50 keV) normally requires huge amount of computing time and large atomic block size. And, this method is not effective in observing the impact of neutron flux and impurity elements on primary damage due to the small timescales and the scarcity of proper interatomic

potentials. The description of the interatomic potential in many-body system containing impurity atoms is a demanding issue in the field of computational materials science. Nevertheless, the MD simulation is of use in quantitatively defining the state of the primary damage (within tens of picoseconds) in the restricted space. In addition, the MD simulation results can be used as inputs to other model calculations which are linked to the multiscale modeling approach [5].

The MD code used in this study was the MOLDY [7], modified to run on a PC. This code has been used widely for simulations of displacement cascades in metals. The classical equations of motion of the atoms are integrated via a Gear 4-value predictor-corrector algorithm [11] and the many-body interatomic potential for α-iron derived by Finnis and Sinclair [12] is embedded in the MOLDY. The process of performing the MOLDY simulation requires the following steps. Before running the MOLDY, a proper size of computational block should be designated depending on the PKA energy. Then, the block of atoms should be equilibrated at a given temperature. When the displacement reaction is initiated, the atomic block should be in the equilibrated phonon mode. The cascade simulations are initiated by imparting a kinetic energy to an atom in a particular direction. This atom corresponds to the PKA created by a collision with an initial neutron. The cascade simulation lasts until intra-cascade recombination of point defects is complete. Generally, it is a common thing to obtain representative damage parameters by averaging several output results for a given cascade energy and temperature.

The 4.7 keV and 5.3 keV cascade simulations have been completed at 290℃ up to 15 ps. As discussed previously, the values of 4.7 keV and 5.3 keV represent the average PKA energies for the HANARO and YG 5 irradiation conditions, respectively. We cannot input these PKA energies directly to the MOLDY code due to the following reason. The code does not account for energy loss due to ionization and electronic excitation. Only a certain fraction of the PKA energy (E_p) contributes to the cascade reaction with elastic collisions. This initial kinetic energy, used as an input energy in the MOLDY computation, is analogous to the damage energy (T_{dam}) in the standard NRT model [6,13]. The relationship between the PKA energy E_p and the damage energy Tdam is expressed as:

$$\frac{T_{dam}}{E_p} = \frac{1}{1+\lambda\, w(E^*)} \tag{1}$$

where λ is given by $\lambda = 0.0876\, Z^{1/6}$, Z = atomic number. The function w(E*) and its variable E* can be written in terms of E_p and Z:

$$w(E^*) = E^* + 0.402(E^*)^{3/4} + 3.4(E^*)^{1/6} \tag{2}$$

$$E^* = \frac{E_p}{0.0869\, Z^{7/3}},\ E_p \text{ in keV} \tag{3}$$

Note that the MD simulation energy is identical to the damage energy, T_{dam} in Eq. (1). Accordingly, the PKA energy of 4.7 and 5.3 keV corresponds to 3.65 and 4.09 keV MD simulation energy for iron, respectively.

The cascade simulations have been performed until the phase of in-cascade recombination of interstitials and vacancies is finished. However, after recombination phase, the atomic block has not returned to complete thermal equilibrium because of the high temperature. Therefore, we had to run the code until the number of point defects does not change considerably. In this case, the required time was over 15 ps. The parameter of primary interest to this simulation is the number of point defects that survive from in-cascade recombination since microstructural changes to irradiated materials are caused by these surviving defects. The evolution of the displacement cascade

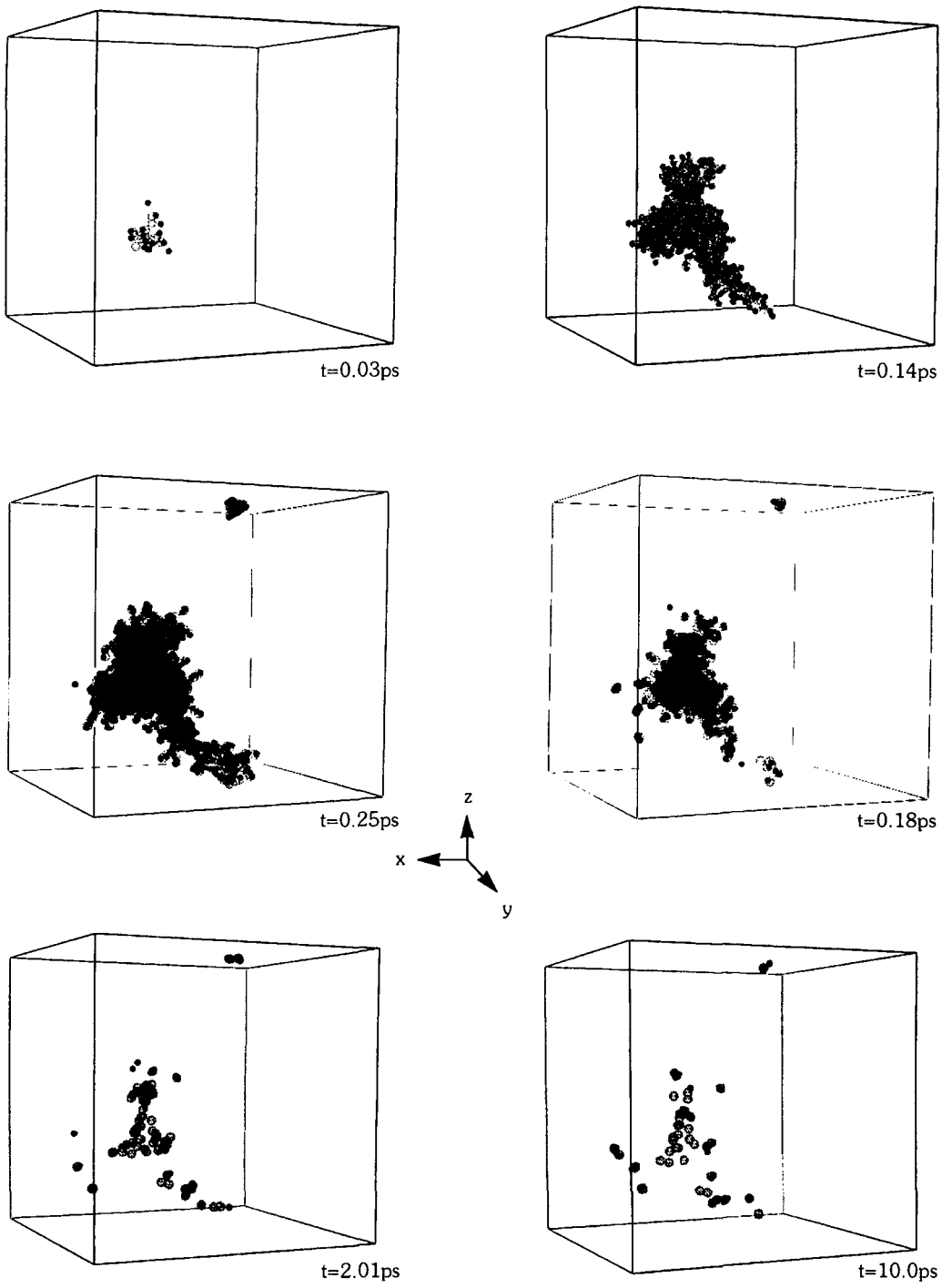

Fig. 3. Illustration of Displacement Cascade Evolution from a 5.3 keV PKA Simulation in Iron at 290℃ as a Function of Time. The Block Size is $40a_o \times 40a_o \times 40a_o$ (a_o: lattice constant) and the Initial PKA Direction [135]. Filled Spheres Represent Interstitials and Empty Ones Vacancies

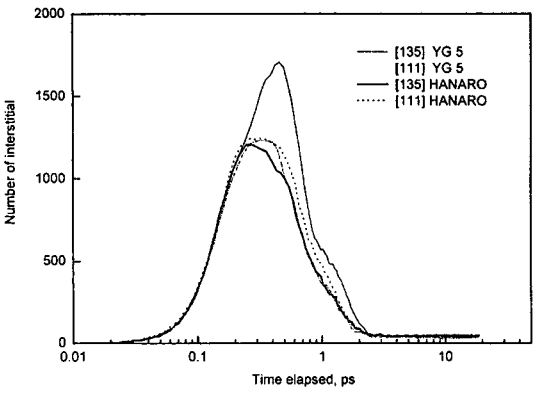

Fig. 4. Time Dependence of the Number of Surviving Interstitials as a function of Initial PKA Direction. [uvw] Represents the Direction of a PKA in α-iron

observed in the 5.3 keV PKA cascade is illustrated in Figure 3, which shows atomic behaviors from the beginning of a cascade to its relaxation in iron at 290℃. The number of point defect reaches a peak at t = 0.25 ps, and then its the number starts to decrease gradually due to the recombination of interstitials and vacancies. The constant number of stable defects could not be obtained until t = 18 ps. About 40 interstitials remain at the end, of about 1200 displaced atoms at the peak of the cascade, which is the same case with vacancies.

For given PKA energy, three simulations have been carried out with different initial directions of a PKA. Three directions are considered: [135], [111], and [100]. Figure 4 shows the changes in the number of surviving interstitials as a function of initial PKA direction and its energy. In all cases, most point defects are annihilated through recombination reactions within a short time (about 2~3 ps). As expected from the SPECTER code calculation, there is not much difference in the number of surviving defects between the two conditions (YG 5 and HANARO). The details of the simulation results, including time to reach peak number in displaced atoms, peak number of displaced atoms, and number of surviving defects at t = 10 ps, are summarized in Table 2. It is, however, observed that there is a correlation between PKA directions and the number of surviving defects. The PKA with a low-index direction [100], as compared to a [135]-direction, tends to bring about a lesser number of surviving defects.

4. Discussions

We have presented a methodology to investigate the primary damage under neutron irradiation. The methodology is based on the utilization of computer codes to simulate radiation damage production in displacement cascades. By combining the SPECTER and the MOLDY code,

Table 2. Summary of MOLDY Simulation Results. Primary Damage Parameters as a Function of Initial PKA Direction for YG 5 and HANARO

	Initial PKA direction	Time to reach peak in displaced atoms (ps)	Peak number of displaced atoms	Number of surviving defects after t = 10 ps
YG 5	[135]	0.46	1706	44
	[111]	0.36	1228	30
	[100]	0.32	1205	25
HANARO	[135]	0.25	1204	41
	[111]	0.31	1243	35
	[100]	0.32	1454	28

we have tried to demonstrate how to determine the primary damage parameters under different irradiation conditions.

The first point to note is that two sets of neutron spectrum have been used as sample calculations. From the fact that radiation embrittlement of RPV is a primary concern in the nuclear industry, we chose neutron spectrum at the inner surface of RPV steel for the commercial PWR (YG 5). And, we obtained another neutron spectrum at the irradiation capsule for the research reactor (HANARO). In fact, irradiation tests are being performed in the HANARO facilities for investigating the degree of RPV embrittlement. Both neutron spectra are calculated ones.

From a viewpoint of displacement damage, the neutron energy distribution is the most important factor. However, not a big difference in neutron spectra has been found between the two cases except total neutron flux. For this, we have obtained similar distribution of PKA recoil energy and average PKA energy from the SPECTER calculations. Although neutron flux, which is directly related to the dose rate, is another important factor to control the radiation damage, the present work cannot handle this effect due to the limitations to the MOLDY code. In order to include the dose rate effect on radiation damage, another modeling technique will be required such as reaction rate theory or kinetic Monte Carlo, which are beyond the scope of this study.

The primary defects generated by the displacement cascade take the form of two types; either isolated point defects or defect clusters. These defects play important roles in subsequent microstructural evolution and eventually affect material property changes. The isolated defects are free to migrate over relatively long distances, called freely-migrating defects, which are linked to microstructural changes. Defect clusters, created directly from the displacement cascades, are significant in that these small clusters provide nuclei for the growth of larger defects. The existence of small clusters implies that the extended defects can evolve more rapidly than the classical nucleation process. It is, therefore, important to quantify the net production of isolated defects before any correlation between radiation damage and microstructural evolution is made. For example, in the case of 5.3 keV PKA simulation shown in Figure 3, 44 interstitials remain as either isolated defects or clusters in the period of thermal relaxation. Nine out of 44 interstitials become isolated defects, whereas the others form clustering such as di-, tri-, and tetra-interstitials. It is well known that these results are strongly dependent on the PKA energy. More details on the MOLDY calculation results, involving the clustering fraction of surviving defects, their spatial distribution, etc. as a function of PKA energy, will be given in a later publication.

At present, the Reactor for Virtual Experiments (REVE) project is in progress, which is a collaborative effort between Europe, United States and Japan. This project aims at developing tools for computational simulation of irradiation effects on materials [14]. The first phase of the REVE project deals with RPV steel embrittlement, which is relatively well understood about degradation mechanism and many tools - computer codes have been developed or are under development. The intent of this paper is to introduce some part of multiscale modeling approach to quantitative simulation of radiation damage and to demonstrate the code calculations. Although we did not obtain productive results regarding primary damage due to the neutron spectral similarity, a new attempt is being made to investigate the radiation damage using computational modeling methods.

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References

1. A.F. Calder and D.J. Bacon, "A molecular dynamics study of displacement cascades in α-iron," *J. Nucl. Mater.* 207, 25 (1993).
2. W.J. Phythian, R.E. Stoller, A.J.E. Foreman, A.F. Calder and D.J. Bacon, "A comparison of displacement cascades in copper and iron by molecular dynamics and its application to microstructural evolution," *J. Nucl. Mater.* 223, 245 (1995).
3. D.J. Bacon, A.F. Calder, F. Gao, V.G. Kapinos and S.J. Wooding, "Computer simulation of defect production by displacement cascades in metals," *Nucl. Instr. Meth. B* 102, 37 (1995).
4. R.E. Stoller, "Evaluation of neutron energy spectrum effects and RPV thru-wall attenuation based on molecular dynamics cascade simulations," *Nucl. Eng. Des.* 195, 129 (2000).
5. B.D. Wirth, M.J. Caturla, T. Diaz de la Rubia, T. Khraishi and H. Zbib, "Mechanical property degradation in irradiated materials: A multiscale modeling approach," *Nucl. Instr. and Meth. B*, 180, 23 (2001).
6. L.R. Greenwood and R.K. Smither, "SPECTER: Neutron damage calculations for materials irradiation," ANL/FPP/TM-197, Argonne National Laboratory (1985).
7. M.W. Finnis, "MOLDY6 - A molecular dynamics program for simulation of pure metals," Harwell Report AERE R-13182 (1988).
8. L.R. Greenwood, "Neutron interactions and atomic recoil spectra," *J. Nucl. Mater.* 216, 29 (1994).
9. C. Seo, KAERI Internal Memo, HAN-RR-CR-920-01-76 (2001).
10. B. C. Lee, Private communication.
11. R. W. Hornbeck, Numerical Methods, p.199, Quantum Publishers, Inc. New York (1975).
12. M.W. Finnis and J.E. Sinclair, "A simple empirical N-body potential for transition metals," *Phil. Mag. A*, 50, 45 (1984).
13. M.J. Norgett, M.T. Robinson and I.M. Torrens, "A proposed method of calculating displacement dose rates," *Nucl. Eng. Des.* 33, 50 (1975).
14. S. Jumel, C. Domian, et al., "Simulation of Irradaition Effects in Reactor Pressure Vessel Steels: the Reactor for Virtual Experiments (REVE) Project," *J. Test. Eval.* 30, 37 (2002).