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Multiscale Modeling of Radiation Damage: Radiation Hardening of Pressure Vessel Steel

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Abstract

Radiation hardening is a multiscale phenomenon involving various processes over a wide range of time and length. We present a multiscale model for estimating the amount of radiation hardening in pressure vessel steel in the environment of a light water reactor. The model comprises two main parts: molecular dynamics (MD) simulation and a point defect cluster (PDC) model. The MD simulation was used to investigate the primary damage caused by displacement cascades. The PDC model mathematically formulates interactions between point defects and their clusters, which explains the evolution of microstructures. We then used a dislocation barrier model to calculate the hardening due to the PDCs. The key input for this multiscale model is a neutron spectrum at the inner surface of reactor pressure vessel steel of the Younggwang Nuclear Power Plant No. 5. A combined calculation from the MD simulation and the PDC model provides a convenient tool for estimating the amount of radiation hardening.

Key Words : multiscale model, molecular dynamics (MD), point defect cluster (PDC) model, radiation hardening

1. Introduction

Radiation damage is an inherently multiscale phenomenon that involves various processes spanning a broad range of time and length scales. When a high-energy particle collides with lattice atoms in a solid, the displacement reactions produce non-equilibrium point defects within a range of nanometer for several picoseconds. Such defects diffuse over macroscopic length and time scales and develop into a full-fledged microstructure by interacting with other extended defects. The pertinent processes include dimensions from atomic size to structural component that span more than 15 orders of magnitude. The time scale also extends over a wide magnitude, from femtoseconds (10^{-15} s) to decades [1].

Irradiation changes the microstructure and property of materials, thereby degrading the integrity of structural components and causing safety problems. Evaluating the behavior of the

internal structures of light water reactors (LWRs) in the nuclear industry is therefore important. Most problems of irradiated materials originate from the atomic collision of high-energy particles and lattice atoms. This collision leads to the displacement cascades through the energy transfer reaction and causes various types of defects such as vacancies, interstitials, and clusters. The behavior of the point defects created in the displacement cascades is important because the defects play a major role in the microstructural evolution and in the changes to the material property.

A lattice atom that a projectile first strikes and displaces is called a primary knock-on atom (PKA). A PKA possesses sufficient kinetic energy to initiate further atomic displacements. The cascade that a PKA generates usually occupies a region about several nanometers in diameter and occurs in a time scale of several picoseconds. Primary damage refers to the microscopic changes in materials, such as the number and configuration of the point defects within several picoseconds. Radiation damage is closely linked to the state of the primary damage.

Investigating the state of the primary damage through quantitative experimentation is impossible. However, with the advancement of computer capability and the development of many-body potentials, molecular dynamics (MD) method enables us to simulate the displacement cascade on an atomic scale. To study the evolution of displacement cascades in α-iron occurring under LWR environments (290℃), we used the MOLDY code [2]. From the MD simulations, emphasis is placed on the quantifying the primary damage state - the residual defect distribution.

A theoretical model is presented that estimates the amount of radiation hardening under neutron irradiation [3]. We used a reaction rate theory to describe the evolution of microstructures in iron and a dislocation barrier model to predict an increase in yield strength. The model is based on the assumption that point defect clusters (PDCs) are the primary cause of hardening and that these defects eventually act as a barrier to the dislocation motion. The displacement cascade can directly create small clusters, and the interaction of mobile defects and clusters can develop the small clusters into extended defects. Basically, the model calculates the PDC concentration and size as a function of irradiation time. To estimate the PDC concentration, we need to input the parameters related to the primary damage state: for example, the cascade efficiency and the fraction of defect clustering. We used the results of our MD calculation as input for the equation of the PDC model. By combining the MD simulation and the PDC model, we can theoretically quantify the primary defects and present the basis of multiscale modeling for the study of radiation damage. Moreover, with the aid of the multiscale model, we can calculate the amount of radiation hardening in pressure vessel steel at Younggwang Nuclear Power Plant No. 5 (YG5).

2. Modeling Approach

The modeling approach involves a hierarchical scheme that integrates the basic calculation of radiation damage, the MD simulation and the calculation of the PDC model. Because the PDC model has been explained in detail elsewhere, we focus on calculating the radiation damage taking place at the early state of irradiation [4]. In the calculation, we used the SPECTER code for the parameters of the displacement rate and the average PKA energy for a given neutron spectrum [5]. With the average PKA energy obtained from the SPECTER code, we then performed the MD simulation to define the parameters for the primary damage of iron. Finally, with the primary damage parameters from the MD simulation, we

used the PDC model to estimate an increase in the yield strength of the irradiated iron.

2.1. Basic Calculations of Radiation Damage

PKAs can be created by means of various nuclear reactions. The PKA spectrum is determined by such factors as the incident neutron energy, the masses involved, and the angle between the incident neutron and the recoil direction. We can readily obtain the PKA spectrum of various elements for a given neutron spectrum from the SPECTER code calculation [5]. The SPECTER code contains libraries of preprocessed cross sections for atomic displacements and gas production, as well as atomic recoil energy distributions on a specified hundred-point neutron energy grid. Accordingly, for a given neutron energy spectrum, the code simply calculates the parameters of the spectral-weighted radiation damage by converting the master library files into a user group structure. Although the SPECTER code is a convenient tool for obtaining the basic damage parameters of neutron irradiation, it does not account for burnup. This can significantly affect the displacement reaction when a transformed element produces displacement at a rate comparable to the original element. In this case, we first make a time-dependent neutronics calculation and then use the SPECTER code to calculate the radiation damage.

To obtain the basic radiation damage parameters, we used a typical set of neutron spectra for the inner surface of a reactor pressure vessel (RPV) at the YG5. A normalized neutron spectrum for YG5 is plotted in Figure 1, which clearly shows the relative amount of neutron flux as a function of neutron energy. The portion of high-energy neutrons (E_n > 1 MeV), which is influential in inducing the displacement cascades, is about 22 percent. RPV steel comprises various elements such as Fe, Ni, Mn, and Mo. However, the presence of minor elements such as Ni, Mn, and Mo does not significantly affect the displacement damage because of the similar level of atomic weights and the small volume fraction of these elements. Hence, this study only deals with pure iron (bcc iron) as target material. We obtained the basic parameters of radiation damage from the SPECTER code calculations. The calculated displacement rate is 1.83 × 10^{-9} dpa/s and the average PKA energy for iron is 5.3 keV. Figure 2 plots PKA recoil spectra for iron at the YG5.

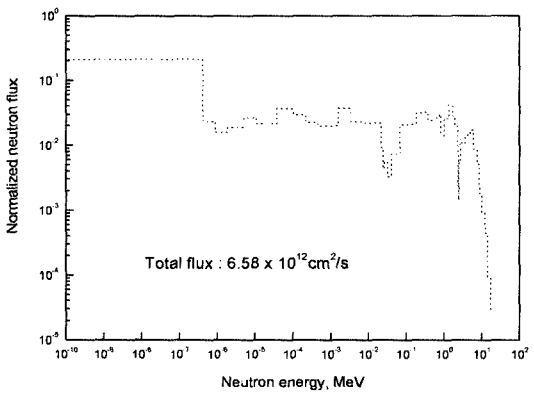

Fig. 1. Normalized Neutron Spectrum for the Inner-surface of a Reactor Pressure Vessel at YG5

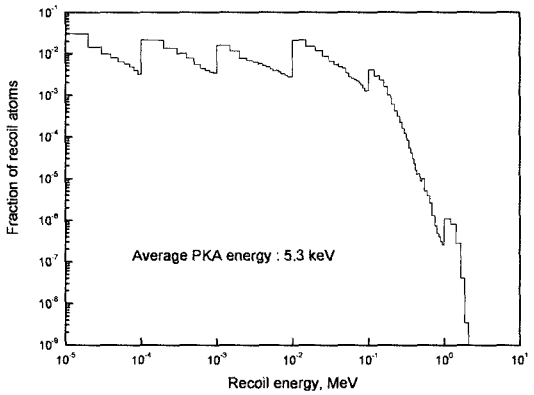

Fig. 2. Energy Distribution of Iron Recoils at YG5. The Spectral-averaged PKA Energy is 5.3 keV

2.2. Molecular Dynamics Simulation

The MD method is a suitable technique for simulating displacement cascades in ordered materials. It is relevant because the time and physical-length scales for displacement cascades are so small that the use of experimental technique is difficult. With the rapid growth in computer modeling, we can simulate atomic behavior to a degree of tens of picoseconds. The MD simulation is particularly useful for quantitatively defining the state of primary damage in a restricted space. Moreover, the MD simulation results provide input for the PDC model.

For the MD calculation, we used MOLDY code [2]. The MOLDY code has the following features: The classical equations of the motion of the atoms are integrated via a Gear four-value predictor-corrector algorithm [6] and the many-body interatomic potential for α-iron derived by Finnis and Sinclair is embedded in the MOLDY code [7]. The proper size of the computational block should be designated according to the PKA energy. The cascade simulations are initiated by giving one of the lattice atoms a defined amount of kinetic energy in a specified direction. The selected atom is equivalent to the PKA mentioned previously. Due to the statistical variability of selecting either the position of an atom or the PKA direction, at least six different cascades are typically required to represent the average behavior at any specified energy and temperature [8].

As seen from Figure 2, the value of 5.3 keV represents the average PKA energy for the YG 5 irradiation conditions. However, this value cannot be used as the damage energy in the MOLDY code because the MOLDY code fails to account for the loss of energy caused by ionization and electronic excitation. Only a fraction of the PKA energy (E_{pka}) contributes to the cascade reaction with elastic collisions. As the input energy in the MOLDY computation, this initial kinetic energy is analogous to the damage energy (T_{dam}) in a standard Norgett-Robinson-Torrens (NRT) model [5,9]. The relationship between the PKA energy and the damage energy is given by

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

where λ is given by $\lambda = 0.0876\, Z^{1/6}$ and Z is the atomic number. The function w (E^+) is

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

and the variable E^+ can be written in terms of E_{pka} and Z as follows:

$$E^+ = \frac{E_{pka}}{0.0869\, Z^{7/3}}, \quad E_{pka} \text{ in keV} \tag{3}$$

The MD simulation energy is identical to the calculated value of the damage energy. The PKA energy of 5.3 keV corresponds to 4.09 keV of energy for the MD simulation of iron.

We performed cascade simulations until the phase of the in-cascade recombination of interstitials and vacancies was finished. After the recombination phase, however, the atomic block did not return to the thermal equilibrium state because of the high temperature. As a result, the MD simulation should be terminated when little change occurs in the number of point defects. For the MD simulation, the parameter of primary interest is the distribution of residual point defects after the in-cascade recombination. These residual defects are known to cause microstructural changes to irradiated material. Figure 3 illustrates the evolution of the displacement reactions in the 5.3 keV PKA cascade; the figure shows each step from the beginning of a cascade to its relaxation in iron at 290℃. The number of point defects reaches a peak at about t = 0.30 ps. When the

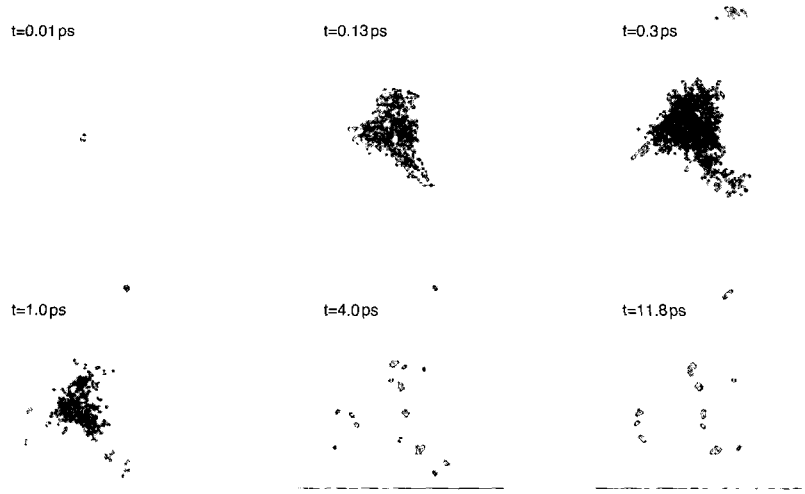

Fig. 3. Displacement Cascade Evolution from a 5.3 keV MD Simulation in α-iron at 290℃ as a Function of Time. The block size is $40a_o \times 40a_o \times 40a_o$ (a_o: lattice constant) and the initial PKA direction is [135]. The filled spheres represent interstitials; the empty spheres represent vacancies

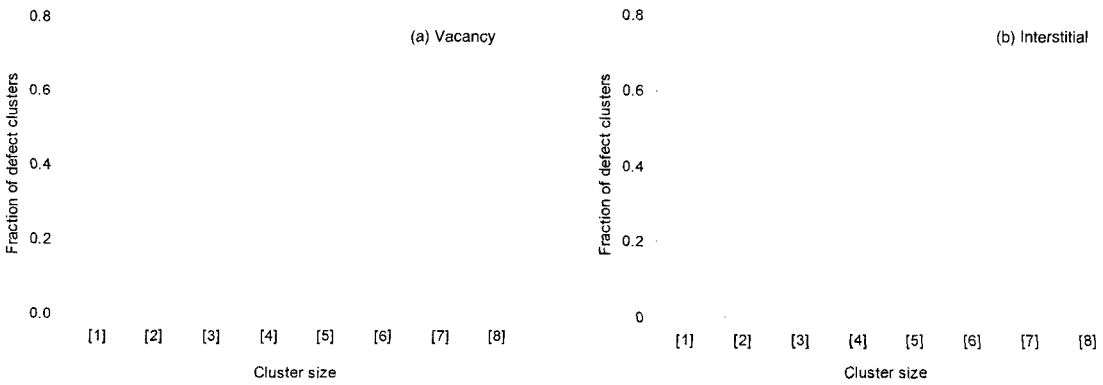

Fig. 4. Distribution of Residual Point Defects in Cascades: (a) vacancy and (b) interstitial. The data are averaged over six MD simulations in α-iron at 5.3 keV PKA energy

recombination of interstitials and vacancies starts to occur, the number of point defects decreases gradually. The constant number of stable defects could not be obtained until t = 10 ps for the 5.3 keV cascade. Of about 1200 displaced atoms at the peak of the cascade, about 40 interstitials remained at the end, which is a phenomenon similar to that of the vacancies.

For given value of PKA energy, we performed six simulations with different positions and different initial PKA directions. For two different initial PKA positions, we considered three directions: [135], [123], and [111]. We derived the size distribution of the PDCs by averaging the six simulation results, which fail to show a strong dependency on the initial direction and the PKA position. Figure 4(a) shows the vacancy cluster distribution in α-Fe. Seventy percent of the

Table 1. Data for the Mean Number of Residual Point Defects Per 5.3 keV Cascade in α-iron at 290°C (each value is the average for six cascade simulations)

Cascade efficiency	0.64
Vacancy clustering fraction	0.31
Number of vacacies per initial vacancy cluster	3
Di-interstitial clustering fraction	0.32
Tri-interstitial clustering fraction	0.1
Tetra-interstitial clustering fraction	0.06

residual vacancies tend to form single point defects in the primary damage state. On the other hand, as shown in Figure 4(b), a higher portion of interstitials form clusters, albeit small ones. The clustering formation of the interstitials is significant because these defects are thermally stable and can migrate away from the region of their parent cascade to be absorbed preferentially at such sinks as dislocations and grain boundaries. In contrast, vacancy clusters are not stable and tend to be dissociated into single point defects at high temperature.

Several parameters were derived from the results of the MD computation. We obtained the cascade efficiency by dividing the number of Frenkel pairs produced in the primary damage state by the number obtained from the NRT formula [9]. Generally, the cascade efficiency, which accounts for the in-cascade recombination, decreases as the PKA energy increases. In the MD simulation, we found a certain fraction of point defects exists as clusters. For interstitial clusters, we evaluated the in-cascade clustering fractions according to the size of the interstitials, while we determined the vacancy-clustering fraction as a single value by assuming that all vacancy clusters were created with the same size. Table 1 lists the important parameters from the MD computation that are used in the calculation of the PDC model.

2.3. Estimation of Radiation Hardening

The details of the mathematical model for the PDC evolution are described elsewhere and are not repeated here [3,4]. Our discussion emphasizes the parameters related to the MD simulation and the calculations of the PDC model. Briefly, the model numerically integrates the rate equations that describe the time dependence of the vacancy and interstitial concentrations, the interstitial-cluster and vacancy-cluster concentrations, and the vacancy cluster size. In solving the rate equations, we referred to other literature to help determine the kinetic parameters of the point defects, which include the migration energy and diffusivity pre-factor [10]. Table 2 shows the kinetic parameters we used; these parameters differ from the standard values that Stoller suggested [3].

Table 2. Migration Energies and Diffusivity Pre-factors for Point Defects in α-iron

	Vacancy		Interstitial	
	standard[3]	this work[10]	standard[3]	this work[10]
Migration engergy(eV)	1.25	0.87	0.25	0.167
Diffusivity pre-factor(cm^2s^{-1})	0.5	1.15×10^{-2}	0.05	2.09×10^{-3}

We calculated the hardening increment due to the PDC distribution on the assumption that PDCs act as barriers to dislocation motion. A change in the shear stress that is due to PDCs, $\Delta\tau$ can be expressed as follows:

$$\Delta\tau = \frac{\mu b}{\chi \ell} \tag{4}$$

where μ is the shear modulus, b is the Burgers vector, χ is a factor inversely proportional to the barrier strength, and ℓ is the average barrier spacing between the PDCs. The average spacing is determined by the barrier size, d_n, and barrier

concentration, N_n, which are given by

$$\ell = \left(\sum_n d_n N_n \right)^{-1/2} \quad (5)$$

To estimate the changes in stress, we needed to know the size and concentration of the PDCs. We obtained these values as a result of the calculation of the PDC model. We used a Taylor factor of 3 to convert the shear stress to a change in the uniaxial yield strength, σ_{ys}. Figure 5 plots the estimated change in yield strength for the given displacement rate (1.83×10^{-9} s^{-1}). The results show that the yield strength increases with reactor exposure but the yield strength is saturated with about one month' s irradiation. From the whole calculation, the contribution of the vacancy clusters to the total hardening was negligible. The interstitial clusters appear to be the major source of radiation hardening.

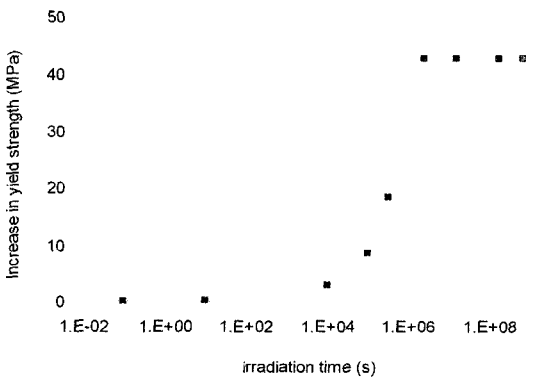

Fig. 5. Estimated Yield Strength Increase of Steel from a YG5 Pressure Vessel at 0-T Surface at 290℃ (displacement rate = 1.83×10^{-9} s^{-1})

3. Discussions

We present a multiscale model for estimating the amount of radiation hardening in pressure vessel steel. The methodology involves a computational approach that integrates the calculation of basic radiation damage, MD simulations and the calculation of a PDC model. The multiscale model enables us to predict the increase in yield strength of neutron-irradiated steel without resorting to a test. However, accurately determining the various kinetic parameters that are linked to point defect behavior is critical.

We used our multiscale model to evaluate the increase in yield strength of RPV steel at YG5. The yield strength increases by up to 43 MPa and is saturated because of the balance between the production and loss rate of the PDCs. Because this model only treats the PDCs as a hardening source, the application of this model to alloys is limited. In fact, PDCs can grow into other types of microstructures such as dislocations, voids, and precipitates. Although there is some uncertainty in determining the kinetic and material parameters for ferritic steel and in applying the simple dislocation models, the results suggest the applicability of the multiscale model for predicting radiation hardening. Future work will focus on improving the present model and on introducing the Monte Carlo method for simulating radiation-induced microstructures.

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