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Multiscale Modeling of Radiation Hardening in Pressure Vessel Steels

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Abstract

Radiation hardening is a multiscale phenomenon involving various processes, spanning a wide range of time and dimension. We present a multiscale model for estimating the amount of radiation hardening in pressure vessel steels under light water reactor environments. The model consists of largely two parts: molecular dynamics (MD) simulation and point defect cluster (PDC) model. MD simulations have been applied to investigate the formation of primary damage due to the displacement cascades. The evolution of microstructures has been described by interactions between point defects and their clusters, which are formulated by PDC model. Then, the hardening due to point defect clusters was calculated using a simple dislocation model. Key input into this multiscale model includes neutron spectrum at the inner surface of the reactor pressure vessel steels for the Younggwang nuclear power plant No. 5 (YG 5). The average energy of primary knock-on atoms (PKAs) produced from YG 5 irradiation condition was used as an initial value for the following MD calculation. The combination of MD simulation and PDC model calculation provides a convenient tool in estimating the amount of radiation hardening. The calculation results suggest that the yield strength is increasing with neutron dose, but not significant at this moment.

1. Introduction

Radiation damage is an inherently multiscale phenomenon involving processes spanning a broad range of time and length scales. When a high-energy particle collides with lattice atoms in a sold, the resultant displacement reactions produce non-equilibrium point defects within a range of nanometer for several picosecond. The defects diffuse over macroscopic length and time scales, developing into full-fledged microstructure. Pertinent processes include the atomic size all the way to the dimension of structural component, spanning more than 15 orders of magnitude. Time scale also extends over a wide range of magnitude from femto-second to tens of years.[1]

It is well known that irradiation changes the microstructure and property of materials, which tends to degrade the integrity of structural components and further causes safety problems. It is, therefore, a big concern to evaluate the behavior of internal structures of operating light-water reactors (LWRs) in the nuclear industry. Most problems of irradiated

materials originate from the atomic collision between the high-energy particles and lattice atoms. This collision leads to the displacement cascades through the energy transfer reaction and results in various types of 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. The production and behavior of point defects created in the cascade are important because they play a major role in the microstructural evolution and material property changes. The cascade generated by a PKA generally occupies a region about several nm in diameter and occurs 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 called primary damage. The radiation damage is closely linked to the state of primary damage.

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] to study the evolution of displacement cascades in α-iron occurring under LWR environments (290^{o}C). Cascades of 5.3 keV in PKA energy are simulated at 290^{o}C in α-iron, which was determined by the SPECTER code using the neutron spectrum on the inner-surface of a pressurized-water reactor (PWR). The emphasis is placed on the quantification of primary damage state – the residual defect distribution.

A theoretical model is presented that estimates the amount of radiation hardening under neutron irradiation. A reaction rate theory was employed to describe the evolution of microstructure in iron leading to the increase in yield strength. The model assumes that point defect clusters (PDCs) are the primary sources of hardening and these defects eventually act as barriers to the dislocation motion. Small clusters can be created directly from the displacement cascade and be developed by diffusive mechanism between mobile point defects and clusters. The model below was developed by Stoller to describe the microstructural evolution of ferritic steels under irradiation.[3] Basically, this model calculates the PDC concentration and its size as a function of irradiation time. In estimating the PDC concentration, the parameters related to the primary damage state are required as inputs which include the cascade efficiency, the fraction of defect clustering, etc. Here, we used the MD calculation results as inputs to the PDC model equation. By combining the MD simulation and the PDC model, 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. And, the amount of radiation hardening of irradiated pressure vessel steels in YG5 is calculated by applying the multiscale model described here.

2. Modeling Approach

The modeling approach involves a hierarchical scheme integrating basic damage calculation, MD simulation and PDC model calculation. Since the PDC model was explained in detail in the previous meeting,[4] we will focus on the first part of multiscale modeling approach in this study. In the section of basic damage calculation, the damage parameters are calculated using the SPECTER code.[5] The parameters of our interest are the displacement rate and the average PKA energy for given neutron spectrum. Then, the MD simulation is performed to define the primary damage to iron. In the MD simulation, we used the average PKA energy obtained from the SPECTER code. Then, we estimate

the increase in yield strength of irradiated iron by the PDC model, where the calculation results from the MD simulation are applied.

2.1 Basic Radiation Damage Calculation

A primary knock-on atom (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. We can readily obtain the PKA spectrum in various elements for a given neutron spectrum from the SPECTER code calculation.[5] 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 calculation should be made first and then the SPECTER code can be employed for damage calculation.

In order to obtain the basic radiation damage parameters, one set of neutron spectrum was used in this study, which represent the typical neutron spectrum at the reactor pressure vessel (RPV) inner-surface in the YG 5 nuclear power plant. The normalized neutron spectrum for YG 5 is plotted in Figure 1, which clearly shows the relative amount of neutron flux as a function of its energy. The fraction of high-energy

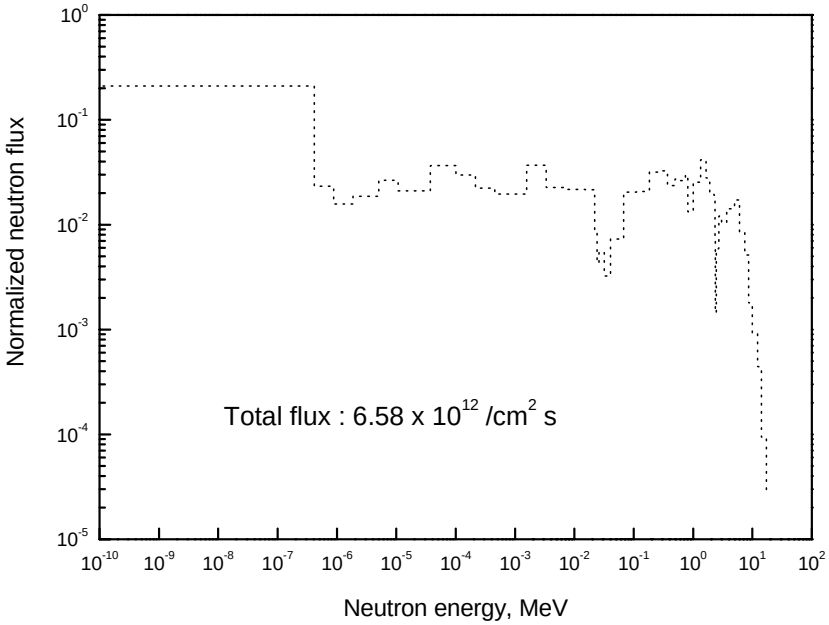

Figure 1. Normalized neutron spectrum at the inner-surface of pressure vessel for the pressurized-water reactor, YG 5

neutrons (E_n > 1 MeV), which are influential in inducing the displacement cascades, is about 22 %. 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 basic radiation damage parameters are obtained from the SPECTER code calculations. The calculated displacement rate is 1.83 x 10^{-9} dpa/s and the average PKA energy for iron is 5.3 keV. The PKA recoil spectra for iron in YG 5 is plotted in Figure 2.

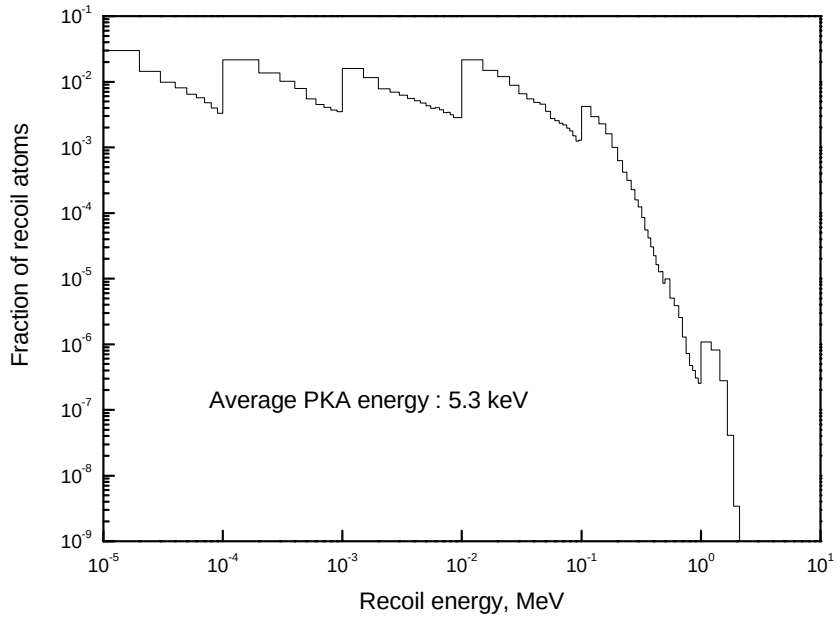

Figure 2. Energy distribution of iron recoils in YG 5. The spectral-averaged PKA energy is 5.3 keV.

2.2 Molecular Dynamics Simulation

The MD method is an appropriate technique for simulating displacement cascades in ordered materials. This method is relevant in that the time and physical-length scales for cascades are too small to be open to experimental studies. The rapid growth in computer modeling makes it possible to simulate the atomic behavior up to tens of picoseconds. It is possible to investigate the cascade characteristics depending on temperature and cascade energy. The MD simulation is of use in quantitatively defining the state of the primary damage (within tens of picoseconds) in the restricted space. The MD simulation results will be used as inputs to PDC models subsequently.

The MD code used in this study was the MOLDY,[2] which was modified to run on a PC. This code has the following features. The classical equations of motion of the atoms are integrated via a Gear 4-value predictor-corrector algorithm[6] and the many-body interatomic potential for α-iron derived by Finnis and Sinclair[7] is embedded in the MOLDY. A proper size of computational block should be designated depending on 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. This atom is equivalent to the

PKA following a collision with a neutron. Due to the statistical variability by selecting either the position of an atom or PKA direction, typically at least six different cascades are required to represent the average behavior at any specified energy and temperature.[8]

As determined previously, the value of 5.3 keV represent the average PKA energy for the YG 5 irradiation conditions. However, we cannot input this value to the MOLDY code due to the following reason. The code does not account for energy loss because of 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.[5] The relationship between the PKA energy E_p and the damage energy T_{dam} 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:

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

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

Note that the MD simulation energy is identical to the calculated value of damage energy, T_{dam}. The PKA energy of 5.3 keV corresponds to 4.09 keV MD simulation energy for iron.

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 10 ps. The parameter of primary interest to this simulation is the distribution of residual point defects after in-cascade recombination. It is known that microstructural changes to irradiated materials are caused by these residual defects. The evolution of the displacement cascade observed in the 5.3 keV PKA cascade is illustrated in Figure 3, which shows each step from the beginning of a cascade to its relaxation in iron at 290^{o}C. The number of point defects reaches a peak at time t = 0.25 ps. As 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 = 15 ps for the 5.3 keV cascade. 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, six simulations have been carried out with different position and initial direction of PKA. Three directions are considered, which are [135], [123], and [111] for two different positions. By combining the MD simulation results, the distribution of point defect clustering is derived by averaging data. As mentioned previously, clustering of point defects in the primary state of cascade damage affects their behavior and role in subsequent evolution of the microstructure. Depending on the initial direction and position of PKA, the statistics show significant variability. The cluster statistics for vacancy in a-Fe is demonstrated by the histogram in Figure 4 (a). It could be seen that 70% of residual vacancies tent to form single point defects in the primary damage state.

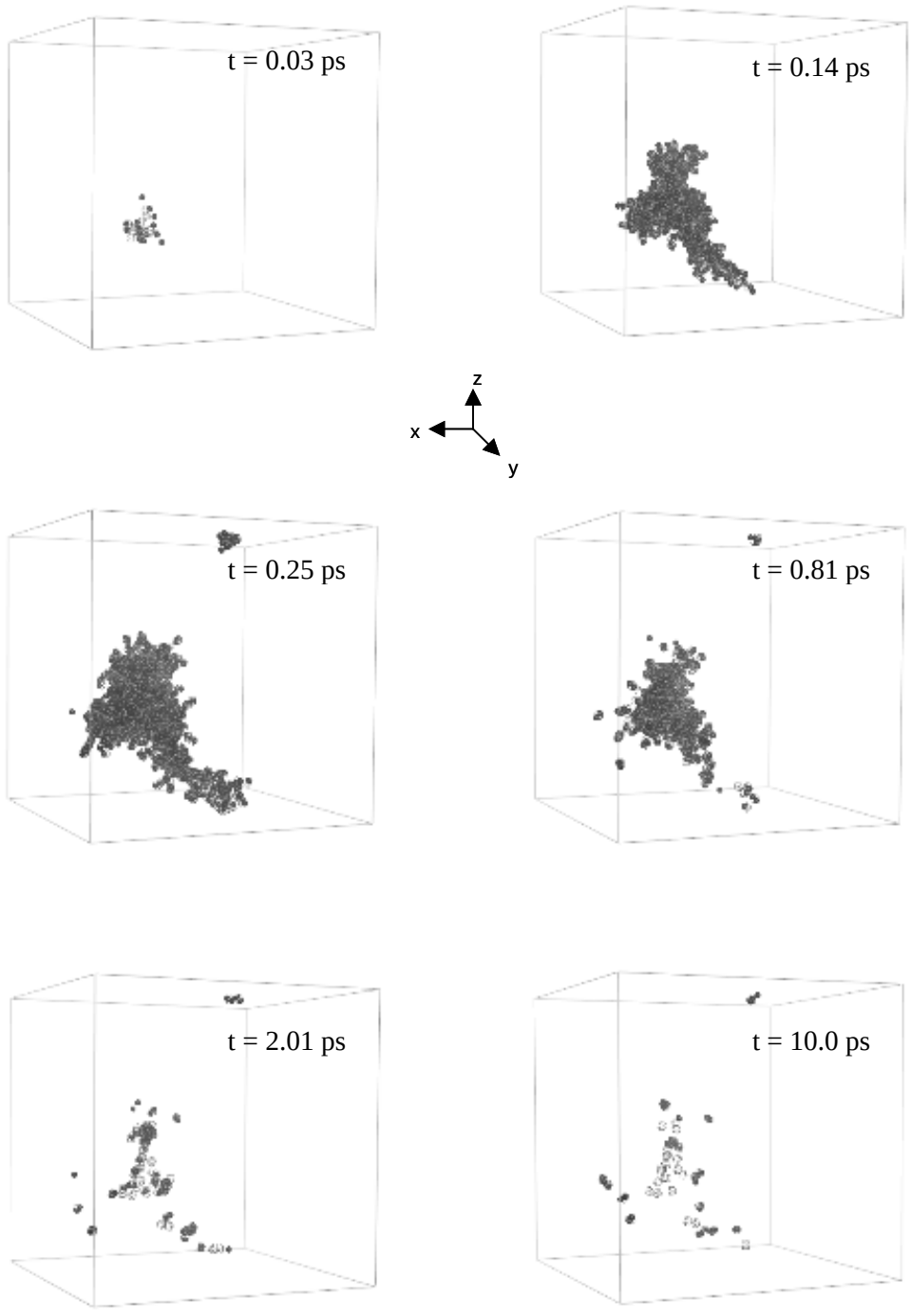

Figure 3. Illustration of displacement cascade evolution from a 5.3 keV PKA simulation in α-iron at 290 ^{o}C as a function of time. The block size is $40a_o$ x $40a_o$ x $40a_o$ (a_o: lattice constant) and the initial PKA direction [135]. Filled spheres represent interstitials and empty ones vacancies.

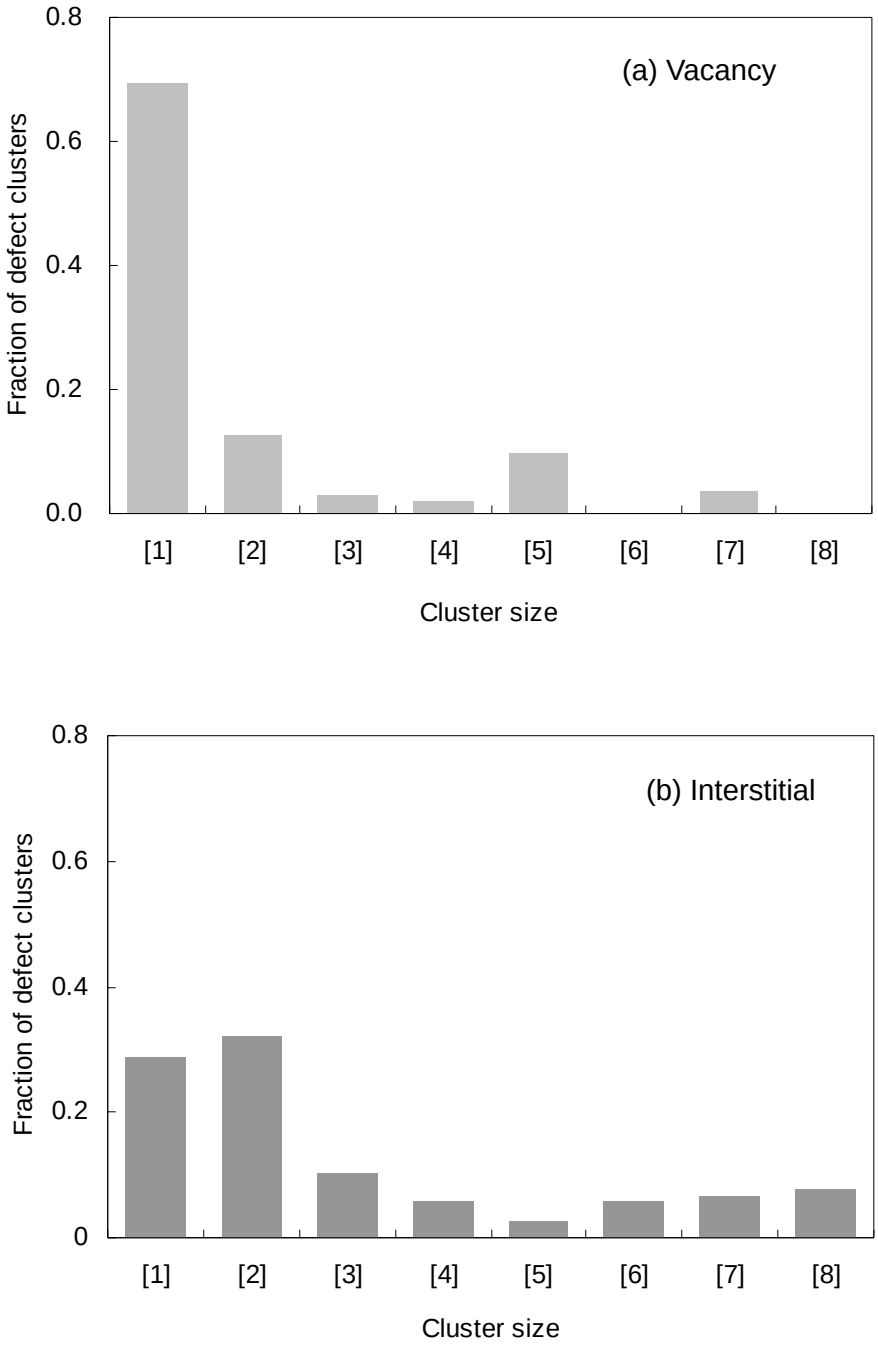

Figure 4. Distribution of residual point defects in cascades: (a) vacancy and (b) interstitial. The data are averaged over six MD simulations in α-iron at 5.3 keV PKA energy for six different cases.

On the other hand, higher portion of interstitials form clusters, albeit small clusters in size, shown in Figure 4 (b). The clustering formation of interstitials is significant in that these defects are thermally stale and can migrate away from their parent cascade region to be absorbed preferentially at such sinks as dislocations, boundaries, etc. Vacancy clusters, in contrast, are not stable and dissociate into single point defects at high temperature.

Several parameters are derived from the MD computation results, which are used in the PDC model calculation. The cascade efficiency is calculated by dividing the number of Frenkel pairs produced from the MD simulation by that calculated from the NRT formula.[9] Generally, the cascade efficiency, accounting for in-cascade recombination, decreases with increasing PKA energy. A large fraction of point defects are found in clusters, seen from the MD simulation. The in-cascade clustering fractions are evaluated depending on the defect size for interstitials, while the vacancy clustering fraction is determined as one value by taking average under the assumption that all vacancy clusters are created with the same size. The important parameters evaluated from the MD computation are listed in Table 1.

Table 1. Data for the population of residual point defects produced by 5.3 keV cascades in α-iron at 290^{o}C (each value is the average for six-cascade simulation).

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

3. Estimation of Radiation Hardening

The details of the mathematical model for PDC evolution are contained in the references[3,4] and will not be repeated here. The discussion will emphasize the parameters related to MD simulation and calculation results. Briefly, the model numerically integrates the rate equations describing the time dependence of the vacancy and interstitial concentrations, the interstitial- and vacancy-cluster concentrations, and the vacancy cluster size. In solving the rate equations, we referred to other literature[10] for determining the kinetic parameters of point defects, which include the migration energy and diffusivity pre-factor. Table 2 shows the kinetic parameters used in this study which are different from the standard values suggested by Stoller.[3] The hardening increment due to the PDC distribution is computed assuming that they act as barriers to dislocation motion.

Table 2. Migration energies and diffusivity pre-factors for iron defects used in this calculation.

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

A change in shear stress $\Delta\tau$ due to PDCs can be expressed as:

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

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 PDCs. The average spacing is determined by the size d_n and concentration N_n of barriers, given by:

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

We obtain the size and concentration of PDCs as a result of the PDC model calculation, which are required to estimate the changes in stress. The Taylor factor of 3 is used to convert the shear stress to a change in the uniaxial yield strength $\Delta\sigma_{ys}$. The change in yield strength for the given displacement rate (1.83×10^{-9} s^{-1}) is plotted in Figure 5. The results show that the yield strength increases with reactor exposure but it is saturated for about one-month irradiation. It could be seen, from the calculation, that the contribution of vacancy defect clusters to total hardening is negligible. Interstitial clusters are major sources of radiation hardening.

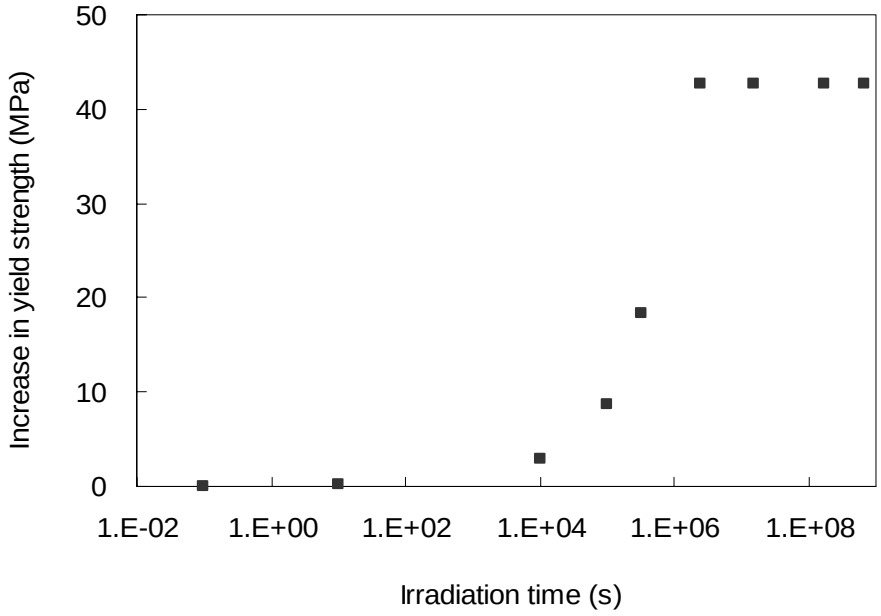

Figure 5. Estimated yield strength increase of YG 5 pressure vessel steel at 0-T surface at 290°C (displacement rate = 1.83×10^{-9} s^{-1})

4. Discussion

This study presents multiscale model for estimating the amount of radiation hardening in pressure vessel steels. The methodology involves computational approach

integrating basic radiation damage calculations, MD simulations and PDC model calculation based on the reaction rate theory. This model enables us to predict the increase in yield strength of neutron-irradiated steels without resorting to test. It is, however, critical to determine the various parameters liked to point defect behavior.

We applied the present multiscale model to evaluate the yield strength increase of reactor pressure vessel at YG 5 power plant. The yield strength increases up to 43 MPa and then saturates due to the balance between the production and loss rate of PDCs. Since the PDC model regards point defect cluster as the only evolved microstructure, there is limitation to this model. In fact, it is possible for PDCs to grow into other types of microstructure including dislocations, voids, precipitates etc. Although there is some uncertainty in determining the kinetic and material parameters for ferritic steels and in applying the simple dislocation models, the calculation results suggest the applicability of multiscale model for predicting radiation hardening. Future work will focus on the continued improvement of the present model and the introduction of kinetic Monte Carlo method for simulating radiation-induced microstructures, as well as PDCs.

5. Acknowledgments

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6. References

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