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# Positron annihilation study and computational modeling of defect production in neutron-irradiated reactor pressure vessel steels

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**Abstract**

Positron annihilation spectroscopy (PAS) and a computer simulation were used to investigate a defect production in reactor pressure vessel (RPV) steels irradiated by neutrons. The RPV steels were irradiated at 250 °C in a high-flux advanced neutron application reactor. The PAS results showed that mainly single vacancies were created to a great extent as a result of a neutron irradiation. Formation of vacancies in the irradiated materials was also confirmed by a coincidence Doppler broadening measurement. For estimating the concentration of the point defects in the RPV steels, we applied computer simulation methods, including molecular dynamics (MD) simulation and point defect kinetics model calculation. MD simulations of displacement cascades in pure Fe were performed with a 4.7 keV primary knock-on atom to obtain the parameters related to displacement cascades. Then, we employed the point defect kinetics model to calculate the concentration of the point defects. By combining the positron trapping rate from the PAS measurement and the calculated vacancy concentrations, the trapping coefficient for the vacancies in the RPV steels was determined, which was about $0.97 \times 10^{15}\ s^{-1}$. The application of two techniques, PAS and computer simulation, provided complementary information on radiation-induced defect production.


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## 1. Introduction

Irradiation with fast neutrons induces various changes in the mechanical and physical properties of materials. It is known that the evolution of microstructures tends to degrade the mechanical properties of reactor pressure vessel (RPV) steels. One effect is irradiation embrittlement; loss of ductility can cause an increase in the probability of brittle fracture. Basically, defects are generated in the form of interstitials and vacancies as a result of the displacement cascades of high-energy particles. The point defects surviving in the cascades can form clusters and migrate through the crystal lattice, to interact with the microstructure including other point defects, solute atoms, dislocations and precipitates. The change of the microstructure under irradiation is the main cause of material degradation. A number of refined methods have been suggested to characterize radiation-induced microstructures including transmission electron microscopy, small angle neutron scattering, atom probe filed ion microscopy, positron annihilation spectroscopy, etc. [1]. Each method is selective to a specific type and the density of the radiation-induced defects.

Positron annihilation spectroscopy (PAS) is a sensitive method for the study of open-volume type defects which include vacancies, vacancy agglomerates, and dislocations [2]. This technique has been widely used for detecting point

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defects in various materials irradiated by high-energy particles. The measured positron annihilation lifetimes of a sample are linked to the size of the open-volume defects and the relative intensities of each defect are related to the defect concentrations. Thus, PAS can provide information on the size of open-type defects and their number density. Since a displacement cascade, initiated by primary knock-on atoms (PKAs), has dimensions of 1–10 nm with a lifetime of only several ps, the spatial and temporal resolution required to investigate its evolution directly is not achievable by any of the known experimental methods [3]. In this work, we applied PAS to study the nature and properties of defects formed during neutron irradiation.

A point defect kinetics model has been used to estimate the concentration of radiation-induced point defects, which simply describes the spatially-averaged point defect behavior as a consequence of the production of displacement cascades, a solid state diffusion and interactions between point defects and extended defects [4]. The point defect kinetics model requires information on the displacement cascades as input parameters. We have, therefore, performed molecular dynamics (MD) calculations to determine the cascade parameters. Of special interest from the MD calculation results is the fraction of the point defect clustering and the cascade efficiency. The two methods, MD simulation and point defect kinetics model, are important factors consisting of multiscale modeling for radiation damage studies, which enable us to determine the defect concentration in a theoretical way.

An attempt was made in this study to complement two techniques, PAS for the experimentation and computer simulation for the theoretical modeling. This coupling method was applied to examine neutron-irradiated RPV steels. The positron trapping rates for vacancies were determined by PAS and the vacancy concentration was calculated from the point defect kinetics model along with MD simulation. This combined methodology presents a unique way for a radiation damage study.

## 2. Experimental

### 2.1. Materials and irradiation

The materials used for the PAS test are commercial-grade steels of which the pressure vessels for the Young-Gwang nuclear power plants were made. The chemical composition of the steels is as follows: 0.21 wt% C, 1.24 wt% Mn, 0.007 wt% P, 0.002 wt% S, 0.24 wt% Si, 0.92 wt% Ni, 0.21 wt% Cr, 0.49 wt% Mo, 0.005 wt% V, 0.03 wt% Cu and 0.022 wt% Al. The specimen was prepared according to the proper size ($10 \times 10 \times 0.5\ mm^3$) for the PAS measurement. After machining, the sample was annealed in vacuum at 600 °C for 10 h. The irradiation was carried out with fission neutrons at 250 °C in a high-flux advanced neutron application reactor (HANARO, Korea). The fast neutron fluence ($E_n > 1.0$ MeV) was around $1.69 \times 10^{19}\ n/cm^2$, which corresponds to ~0.02 dpa (displacement per atom). The normalized neutron spectrum in HANARO is shown in Fig. 1. The PKA spectrum was obtained from the SPECTER code calculation [5] for the neutron spectrum shown in Fig. 1 and the average PKA energy was found to be 4.7 keV for Fe. This PKA energy value will be used for the MD simulations to determine the cascade-related parameters.

### 2.2. Positron annihilation spectroscopy measurements

The positron lifetime measurements for two samples (unirradiated- and irradiated-RPV steels) were performed at room temperature by means of a conventional fast–fast coincidence spectrometer. We employed a $^{22}Na$ $\beta^+$-source of about 1 MBq and collected about two million counts for each sample. The positron lifetime is measured by detecting the time difference between the birth $\gamma$-radiation of the $^{22}Na$ $\beta^+$-source and one of the annihilation $\gamma$-quanta with energy of 511 keV. The time resolution of the system is 180 ps in full width at half maximum. The positron lifetime data was analyzed by using the PALSFIT program [6] by subtracting the source components and background.

The coincidence Doppler broadening (CDB) spectra were measured with two high-purity Ge-detectors. The energies of the annihilating $\gamma$-quanta pair, denoted by $E_1$ and $E_2$, were simultaneously measured by two detectors located at an angle of 180° relative to each other. The overall energy resolution of the system was about 0.9 keV, which corresponds to a momentum resolution of $\sim 3.52 \times 10^{-3} m_0 c$ in the direction of $cP_L$ ($= E_1 - E_2$), where $m_0$ is the electron rest mass and $c$ is the speed of the light. We analyzed the CDB spectra to derive the $S$- and $W$-parameters in order to investigate the general characteristics of the irradiated samples. In this analysis, the $S$- and $W$-parameters are defined as the ratio of the number of the low-momentum quanta ($P_L < 4 \times 10^{-3} m_0 c$) and the high-momentum quanta ($18 \times 10^{-3} m_0 c < P_L < 30 \times 10^{-3} m_0 c$) in the spectra to the total quanta, respectively.

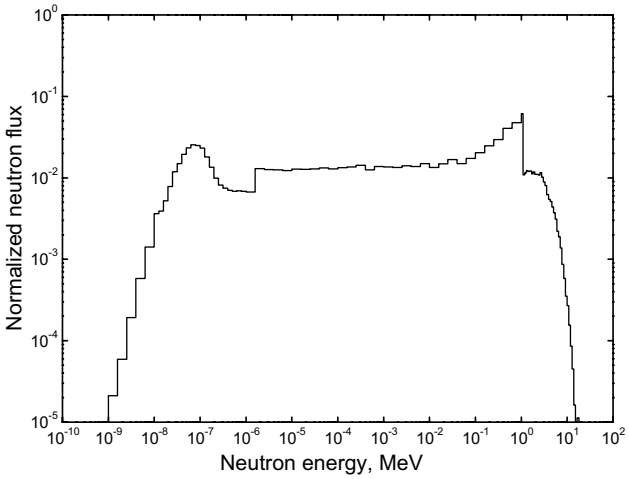


Fig. 1. Energy dependency of a normalized neutron flux in the HANARO ($\phi_{total} = 3.93 \times 10^{14}\ cm^{-2}\ s^{-1}$).