Determination of the Positron Trapping Coefficients for Neutron-Irradiated Reactor Pressure Vessel Steels

Junhyun Kwon*, Yong-Min Kim, Whungwhoe Kim
Korea Atomic Energy Research Institute, 150 Deokjin, Yuseong, Daejeon 305-353
*E-mail: jhkwon@kaeri.re.kr**

1. Introduction

Irradiation of fast neutrons induces various changes in the mechanical and physical properties of reactor pressure vessel (RPV) steels. Basically, point defects are generated in the form of interstitials and vacancies as a result of the displacement cascades by neutrons. These point defects surviving in the cascades interact with various microstructures such as solute atoms, dislocations and precipitates. A formation of microstructures under irradiation is the main cause of a material degradation.

Positron annihilation spectroscopy (PAS) and a computer simulation were used to investigate a defect production in RPV steels irradiated by neutrons. It is known that PAS is a sensitive method for investigating open-volume-type defects such as voids and vacancies. We measured the positron lifetimes and relative intensities of the defects through PAS and calculated the positron trapping rate. A cluster dynamics model, based on the reaction rate theory, has been used to estimate the concentration of radiation-induced defects. The positron trapping coefficient, a parameter which relates the defect concentration to the positron trapping rate, was determined by combining the two techniques of PAS and a computer simulation.

2. Experimental

The materials used for this study are commercial-grade RPV steels of which the pressure vessels for the YoungGwang nuclear power plants were made. The chemical compositions of the steels are as follows: 0.21 w/o C, 1.24 w/o Mn, 0.007 w/o P, 0.002 w/o S, 0.24 w/o Si, 0.92 w/o Ni, 0.21 w/o Cr, 0.49 w/o Mo, 0.005 w/o V, 0.03 w/o Cu and 0.022 w/o Al. 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 x 10^{19} n/cm^2, which corresponds to ~0.02 dpa.

The PAS measurements for two samples (unirradiated- and irradiated-RPV steels) were performed at room temperature by means of a fast-fast coincidence timing spectrometer. We employed a ^{22}Na β^+-source of about 1 MBq and collected about three million counts for each test. The positron lifetime can be measured by detecting the time difference between the birth γ-radiation of the ^{22}Na β^+-source and one of the annihilation γ-quanta with energy of 511 keV. The collected γ-quanta are converted by scintillator-photomultiplier detectors into electrical pulses, which are processed by discriminators. These output pulses start and stop a time-to-amplitude converter as a stopwatch. The generated amplitude which is proportional to the time difference between two γ-quanta stands for a measure of the positron lifetime. The scheme of the positron lifetime measurement is shown in Fig. 1. The time resolution of the system is 260 ps in full width at half maximum. The positron lifetime data was analyzed by subtracting the source components and background. All the spectra were decomposed into two lifetime components, which were denoted by τ_1 (shorter) and τ_2 (longer).

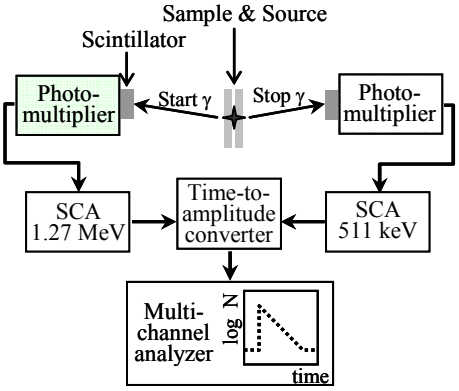

Fig. 1. Scheme of the positron annihilation lifetime spectroscopy (SCA: single-channel analyzer).

3. Cluster Dynamics Calculation

In parallel with the PAS measurements, theoretical approaches were made in order to determine the concentration of the point defects by combining the molecular dynamics (MD) simulation and the cluster dynamics calculation. The MD calculations have been performed to simulate the neutron irradiation conditions in the HANARO, which corresponds to an average PKA energy of 4.7 keV at 250°C. From the MD simulation, we obtained two parameters: the cascade efficiency and the point defect clustering fraction. These parameters are

important in that the surviving defects can contribute to a radiation-induced microstructural evolution. The cascade efficiency was 0.45, and the clustering fractions were 0.1 and 0.33 for interstitial and vacancy clusters, respectively.

The cluster dynamics model describes the evolution of the point defect clusters as a consequence of the cascade events, solid state diffusion and interactions of the point defects with sinks by combining it with the point defect kinetic model. Since the mathematical formulation for the cluster dynamics model is described elsewhere [1], we do not describe the details here. Basically, the cluster dynamics model integrates the rate equations that describe the time dependency of the point defect concentrations, the interstitial-cluster and vacancy-cluster concentrations, and the vacancy cluster size. The determination of the kinetic and material parameters is from estimating the concentration of the point defects.

4. Results

The positron lifetime data obtained from the samples is listed in Table 1. No significant difference in the mean positron lifetime (τ_m) was found between the two samples; τ_m (unirradiated) = 146.3 ps and τ_m (irradiated) = 143.1 ps. In a comparison of the τ_2 components with the two samples, it is likely that large voids are present in the steels before irradiation although their number density is low. However, after irradiation the magnitude of τ_2 decreases and its intensity I_2 increases. The lifetime τ_2 of the irradiated steels is close to the lifetime of the positrons in the mono- and di-vacancies in iron [2]. From the PAS results, we can see that irradiation does not induce large vacancy clusters in the RPV steels, whereas a number of small-size vacancies are formed. Because it is impossible to obtain the absolute density of the defects from the positron lifetime measurements, we have performed theoretical calculations by applying the cluster dynamics model.

When positrons are trapped in open-volume defects, the positron lifetimes change depending on their concentrations and identities. There is a linear relationship between the positron trapping rate and the defect density. For vacancy defects produced by irradiation, the positron trapping rate κ_v can be expressed by the vacancy concentrations C_v as:

$$\kappa_v = \mu_v \cdot C_v \quad (1)$$

where the proportionality constant μ_v is the trapping coefficient. The positron trapping rate κ_v can be obtained from the lifetime data by an application of the two-state model. The vacancy concentration was calculated from the cluster dynamics model, which is plotted in Fig. 2. The steady-state C_v is 2.05×10^{-7} /atom. By comparing the two values of κ_v and C_v, the trapping coefficient μ_v for a vacancy in irradiated RPV steels was determined, which was 5.27×10^{-15} /s.

Table 1. Positron lifetimes and relative intensities of RPV steels

	Unirradiated	Irradiated
lifetime τ_1 (ps)	125.4 ± 0.5	117.3 ± 3.2
Intensity I_1 (%)	92.2 ± 0.3	70.2 ± 4.8
lifetime τ_2 (ps)	396.8 ± 6.1	204.6 ± 8.8
Intensity I_2 (%)	7.8 ± 0.3	29.8 ± 4.8
mean lifetime τ_m (ps)	146.3 ± 1.4	143.1 ± 11.8

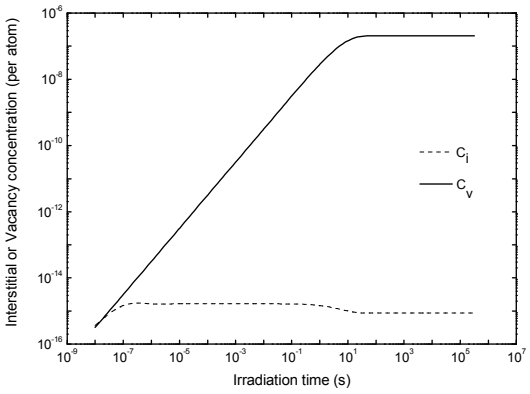

Fig. 2. Time dependence of the interstitial and vacancy concentrations at 250°C.

5. Conclusions

The PAS method and a mathematical model calculation were applied to determine the trapping coefficient for the vacancies in RPV steels. The calculated trapping coefficient of the vacancy for the RPV steels was in the order of 10^{15} s^{-1}, which is close to that for pure iron. This new method for determining the trapping coefficient provides a unique way for a radiation damage study.

REFERENCES

[1] R.E. Stoller, Modeling the influence of irradiation temperature and displacement rate on hardening due to point defect clusters in ferritic steels, Effects of radiation on materials: 16th Int. Symp., ASTM STP 1175 (1993) 394-423.

[2] V. Slugen, G. Kögel, P. Sperr, W. Triftshäuser, Positron annihilation studies of neutron irradiated and thermally treated reactor pressure vessel steels, *J. Nucl. Mater.* 302 (2002) 89-95.