Ion irradiation-induced the radiation hardening in Fe and Fe-0.3Si alloys

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Abstract. Depth profile of Helium irradiation effects in reactor pressure vessel (RPV) model material is identified using a nano-indentation technique in order to develop a new methodology to clarify the solute-vacancy interaction. 2 MeV He ion was irradiated in pure iron at room temperature with and without an energy degrator. The indentation hardness was measured by nanoindentation changing the load at 9 levels. Hardening in irradiated materials was clearly observed in all indentation loads as well as the well-known indentation size effect. It was interesting that the significant increase of hardness was recognized at 0.8 μm depth, which is corresponding to the obstacle of deformation at 3~4 μm depth, that would be the cause of helium implantation. Furthermore, dose effects on 3MeV Si irradiated RPV model materials were confirmed.

1. Introduction

Fast neutron irradiation induces lattice defects in metal such as interstitial atoms, vacancies, and their clusters [1][2][3]. The diffusion of the irradiation-induced defects may cause nano-scale clustering of solute atoms as well as the further clustering of the defects, and these clusters obstacle dislocation motion and lead hardening. In reactor pressure vessel (RPV) made of low-alloy ferrite steels, which is one of the most important components in nuclear power plants suffered by a fast neutron, solute atoms such as Cu, Mn, Ni and Si gather to form solute clusters, and these clusters cause irradiation embrittlement of the RPV. Clustering of these elements is mainly controlled by the diffusion mechanism of irradiation-induced vacancies. Though it is important to observe the interaction between vacancies and solute atoms, experimental techniques to directly observe the vacancy motion is limited.

Helium babble is another important cluster related to the vacancy motion. Helium atoms are generated from nuclear transmutation reactions in materials during neutron irradiation. Transmuted helium gathers with vacancies to form the bubbles. Helium babble formation in ferrite steels is an interesting topic for advanced reactors though is not observed in the RPVs of light water reactor. Furthermore, it is relatively easier to observe helium babbles using a transmission electron microscope (TEM) comparing with the other experimental technique to observe vacancy-type defects, such as residual resistivity method or positron annihilation [4][5].

Ion irradiation is widely used due to well-controlled irradiation condition such as irradiation temperature and dose rate [6][7]. We use ion accelerator to understand the effect of solute elements on vacancy migration by focusing the helium babble development behaviour. However, the weak point of ion irradiation is a large gradient of irradiation defects as well as the injected ions along with the depth
direction. Therefore, as the first step, the objective of this study is to develop the instant detection of the depth profile of irradiation effects using a nano-indentation technique using the model materials of RPV.

2. Materials and methods

As the RPV model materials, pure-Fe and Fe-0.3Si, 5 x 10 mm were used in this study. The chemical composition of the materials is shown in Table 1. The samples were heated to 1000 °C for 1 h and followed by 800 °C for 1 h before quenching in water. After that, it was kept at 600 °C for 24 h to reduce the residual stress. After the heat treatment, the samples were polished one-side using polishing paper and lastly buff polishing with a solution of 0.05 µm. Finally, it was electrochemical-polished by ethanol 90% and perchloric acid 10% to reduce stress concentration on the surface may be generated by the polishing process.

| Table 1. The chemical composition of Fe based materials (wt %). |
|-------------------|---|---|---|---|---|---|---|---|---|
|                  | C  | Si | Mn | P   | S   | Ni  | Cr  | Cu  | O   | N   |
| Fe                | 0.0065 | <0.01 | <0.01 | <0.003 | 0.0021 | <0.02 | <0.02 | <0.02 | 0.0087 | 0.0006 |
| Fe-0.3Si          | 0.0085 | 0.30 | <0.01 | <0.003 | 0.0019 | <0.02 | <0.02 | <0.02 | 0.0018 | 0.0006 |

Stopping and Ranges of Ions in Matter (SRIM) is the Monte Carlo simulation code used for calculating ion deposition profiles in materials by ion implantation [8]. Ion energy plays a significant role in implantation depth of ion into materials. Mazumder et. al. [9] performed irradiation many times with different ion energy in order to irradiate the materials in the uniform depth. In this study, the energy degrader material was used to control the irradiation depth.

2 MeV He\(^{2+}\) ion implantation by 1.7 MV Tandem accelerator at Nagaoka University of Technology was performed to pure-Fe at room temperature with helium of 5 x 10\(^{15}\) ions/cm\(^2\) and 1.5 x 10\(^{16}\) ions/cm\(^2\). These fluences are corresponding to the concentration at a peak of 0.2% and 0.6%, respectively. Titanium foil was used as energy degrader to reduce stopping range of implantation depth. The estimated depth profile of implanted He is about 3.15 µm at the displacement damage peak. Furthermore, the use of 2 µm thickness titanium foil decreases the implantation depth to approximately 1.9 µm at the displacement damage peak. Figure 1 shows the estimation of helium depth distribution calculated by SRIM.

For comparison, the Si\(^{2+}\) ion irradiation was conducted by 1.7 MV Tandem Cockcroft-Walton accelerator at the HIT facility of the University of Tokyo [10] up to the fluence of 1.12 x 10\(^{15}\) and 3.34 x 10\(^{16}\) ions/cm\(^2\) at 400 °C. They are corresponding to the average dose of 5 dpa and 15 dpa, respectively. Figure 2 shows the SRIM calculation displacement depth profile for 3MeV Si ion implantation in Fe. The implantation reaches to approximately 1.3 µm from the surface. Pure-Fe and Fe-0.3Si were irradiated to evaluate the effect of solute material Si to the irradiation hardening.

Nanoindentation tests were performed to determine the mechanical properties of irradiation-induced hardening for both He and Si irradiation samples. Dynamic Ultra Micro Hardness Tester (DUH-211) with Berkovich trigonal pyramid shape indenter tip used in this study. The Oliver and Pharr method was applied to calculate indentation hardness [11]. The schematic diagram of the nanoindentation load applied as shown in Figure 3. For He irradiation samples, nine different loads were performed to analyse the indentation size effect. For Si irradiation sample, the load was set at 5 mN.
Figure 1. The estimation depth of helium concentration level of 0.6 at% profile calculated by SRIM for He\textsuperscript{2+} irradiation in Fe with energy degrader 2\textmu{}m Ti foil and without Ti foil.

Figure 2. SRIM calculation displacement depth profile for 3MeV Si\textsuperscript{2+} ion implantation in Fe with fluence 1.12x10\textsuperscript{16} n/cm\textsuperscript{2}.

Figure 3. The schematic diagram of the nanoindentation procedure used.

3. Results and discussion
Figure 4 shows the relation between nano-hardness of pure Fe irradiated by He\textsuperscript{2+} ion with indentation applied load to the irradiation depth. We can clearly observe that applying low indentation load resulted in high of nano-hardness. This is due to the indentation size effect (ISE) [12]. The figure also shows irradiation hardening. The irradiated pure Fe by 0.2\% and 0.6\% of helium concentration showed an increase of nano-hardness compared to unirradiated pure Fe. However, the nano-hardness was slightly decreased when the concentration of He increases from 0.2\% to 0.6\%. It was reflected to study by Chen et al. [13] that found the nano-hardness decreasing after He concentration increases from 0.1\% to 0.2\%. They observed the formation of small cavities when increasing the He concentration. In our case, the slight recovery of hardening may also be due to the formation of cavities and voids in the irradiated region. The indentation load also affects the penetration of irradiation depth. It has a trend similar to how hardness changes with increasing depth in the case of non-irradiated Fe. The anomalous bumps observed in the hardening in Figure 4 at approximately 0.8 \textmu{}m could be representing the region where the hardening is the most obvious. Usually, the deformation region of nano-indentation is 4–5 time deeper than the penetration depth of the indenter. The bumping region represents the obstacle of dislocation at the depth of approximately 3–4 \textmu{}m. In the region, injected helium would exist at the highest concentration. The depth profile of irradiation-induced hardening represents the microstructure related to helium.
Figure 4. The relation between nano-hardness of He irradiation pure Fe and indentation load in the function of indentation depth.

The 5mN load with maximum indent depth of approximately 0.3 µm was applied to Si2+ ion irradiated specimens. The dose dependence of irradiation hardening for pure Fe and Fe-0.3Si alloys irradiated at 400 °C was shown in Figure 5. Hardness change is the difference of the hardness before and after irradiation; \( \Delta H = H_{\text{irr}} - H_{\text{unirr}} \). The hardening increase with increasing dose. The results may show the change in hardening as a function of the square root of dose. The solute element Si affect the irradiation hardening. The Si element contained in the alloys resulted in decreases of irradiation hardening compared to pure Fe. Liu et al. [14] also reported the same trend where the presence of Si solute element in Fe-Cu alloys decreases the irradiation hardening. Moreover, the findings are in line with the findings from Murakami et al. [15] which stated the presence of solute element Si enhance the recovery of damage defects.
Figure 5. Irradiation hardening of Si$^{2+}$ irradiation pure Fe and Fe-0.3Si for 5dpa and 15dpa at 400°C.

4. Conclusion
We irradiated pure Fe and Fe-0.3Si with He$^{2+}$ and Si$^{2+}$ ion and found that both ion irradiation enhances the irradiation hardening. He$^{2+}$ ion irradiation performed to pure Fe for 0.2% and 0.6% He concentration at room temperature. Nano-hardness decreases with the increase of He concentration from 0.2% to 0.6% through both are harder than the unirradiated material. For Si$^{2+}$ ion irradiation, we irradiated 5dpa and 15dpa for Fe and Fe-0.3Si at 400 °C. The irradiation hardening increases with increasing of dose at 400 °C for both materials, pure Fe and Fe-0.3Si. Further, the addition of solute element Si in Fe resulted to reduce the irradiation hardening. The present of Si may contribute to the enhancement of radiation damage recovery.

5. References
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