Factors Controlling Irradiation Hardening of Iron-Copper Model Alloy*

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The factors controlling irradiation hardening and their contributions to the hardening in electron irradiated pure-iron and Fe-0.15 mass%Cu alloy were determined by means of post-irradiation annealing experiments, such as hardness measurements, positron annihilation spectroscopy (PAS) measurements, transmission electron microscope (TEM) observations and three dimensional atom probe (3DAP) analyses. In pure-iron, almost complete recovering of the hardness was observed after the annealing to 773 K, which was accompanied by disappearing of the interstitial type dislocation loops (I-loops) that were observed in as-irradiated specimen. In contrast, the hardening of Fe-0.15 mass%Cu alloy recovered in a two-step mode; about a half of the hardening recovered by the 773 K annealing, and a complete recovery was observed after annealing to 973 K. Most of the I-loops observed in as-irradiated specimen again disappeared after the annealing to 773 K. These clearly show that the I-loops are one of the main factors controlling irradiation hardening in iron-copper alloy. The residual hardening in the Fe-0.15 mass%Cu alloy after the annealing to 773 K, which is about a half of the irradiation hardening, was attributed to the copper-rich precipitates (CRP) through the direct observation by 3DAP analysis. PAS measurements revealed the disagreement between the recovery behaviors of the hardness and lifetime parameters. Based on the quantitative data analysis, it was concluded that the factor controlling the irradiation hardening of pure-iron is the I-loops, and those in Fe-0.15 mass%Cu alloy are both the I-loops and CRP of which the contributions to the hardening are almost same.

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1. Introduction

The reactor pressure vessel steels (RPVS) of nuclear power plants often contain impurity copper that enhances the irradiation embrittlement through the hardening caused by the formation of copper-rich precipitates (CRP) as well as the formation of matrix defects, such as vacancy clusters (V-clusters) and interstitial type dislocation loops (I-loops).

So far, many researchers have investigated the effect of impurity copper1-9) on the neutron irradiation embrittlement of RPVS using iron-copper model alloys. Although it is expected that V-clusters, I-loops and CRP are the potential hardeners in iron-copper model alloys, each contribution to irradiation hardening has not been made clear yet.

Positron annihilation spectroscopy (PAS) and transmission electron microscope (TEM) have been considered to be able to monitor the behavior of V-clusters and I-loops, respectively. And three dimensional atom probe (3DAP) analysis is expected to be adequate method to detect them directly. Surveillance of the post-irradiation annealing behavior of Vickers hardness and microstructure as well as those at as-irradiated condition may give us information about the role of each defect structure in the observed and/or recovered irradiation hardening.

The objective of this research is to clarify the contribution of the potential irradiation-induced defect structures to irradiation hardening of iron-copper alloys by means of post-irradiation annealing test method, such as Vickers hardness, PAS, TEM and 3DAP analysis.

2. Experimental Procedure

The materials used were pure-iron and iron-copper model alloys of which the concentrations are shown in Table 1. All ingots were made by arc-melting method in high purity argon gas. The ingots were cold-rolled to about 1 mm thick sheets, and then cut into plate specimens (48 mm×10 mm×1 mm). Finally, the specimens were solution annealed at 1073 K for 1 hour, and followed by quenching into iced water.

Electron irradiations were performed with Phodtron 5MeV electron accelerator. The irradiation conditions and temperatures are summarized in Table 2 and Table 3, respectively.

Post-irradiation experiments such as Vickers hardness tests with a load of 0.2 kgf, PAS measurements and TEM

Table 1 Copper and interstitial impurity concentrations of materials.

<table>
<thead>
<tr>
<th>Material (mass%)</th>
<th>Cu</th>
<th>C</th>
<th>O</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>pure-Fe</td>
<td>0.001</td>
<td>0.0002</td>
<td>0.0236</td>
<td>0.0056</td>
</tr>
<tr>
<td>Fe-0.15Cu</td>
<td>0.149</td>
<td>0.0002</td>
<td>0.0226</td>
<td>0.0036</td>
</tr>
</tbody>
</table>

Table 2 Electron irradiation conditions.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Displacement damage, $\psi t$/dpa</td>
<td>$9.0 \times 10^{-4}$, $2.2 \times 10^{-2}$</td>
</tr>
<tr>
<td>Electron dose, $\psi t$/m$^{-2}$</td>
<td>$2.1 \times 10^{23}$, $5.1 \times 10^{24}$</td>
</tr>
<tr>
<td>Displacement damage rate, $\psi$/dpa s$^{-1}$</td>
<td>$2.6 \times 10^{-11}$, $1.7 \times 10^{-11}$</td>
</tr>
<tr>
<td>Dose rate, $\psi$/m$^{-2}$ s$^{-1}$</td>
<td>$6.0 \times 10^{18}$, $3.9 \times 10^{19}$</td>
</tr>
<tr>
<td>Period, t/h</td>
<td>9.7, 356.8</td>
</tr>
</tbody>
</table>

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analyses were performed at a tip temperature of about 60 K.

Figure 1 shows the recovery behavior of Vickers hardness of (a) pure-iron; and (b) Fe-0.15Cu alloy during annealing to 923 K. The alphabets shown in the figure correspond to the annealing conditions for TEM observations which will be discussed in the Figs. 3 and 4.

observations were carried out at room temperature. 3DAP analyses were performed at a tip temperature of about 60 K under an ultrahigh vacuum condition (~2 × 10^-8 Pa) with a ratio of pulse voltage to the static voltage of 0.2. Post-irradiation annealing behavior of these properties was also examined through isochronal annealings for 30 minute in a vacuum at each temperature from 598 to 973 K.

3. Results

3.1 Irradiation hardening

Figure 1 shows the recovery behavior of irradiation hardening. For pure-iron, only one recovery stage appeared below 773 K. On the other hand, two recovery stages appeared in Fe-0.15Cu alloy below and above 773 K. This clearly indicates that the latter stage is due to the recovery of CRP induced by the irradiation. Although the factors controlling the hardening below 773 K are expected to be matrix defects and CRP, the magnitude of the contributions are not cleared yet. It is shown that the recovery stage under 773 K shifts to a higher temperature with increasing electron dose both in pure-iron and Fe-0.15Cu alloy. In contrast, the recovery stage over 773 K does not depend on electron dose.

3.2 Microstructure change

Figure 2 shows the recovery behaviors of PAS for pure-iron(a) and for Fe-0.15Cu alloy(b), respectively. The results show the average of positron lifetime ($\tau_m$) decreased with increasing annealing temperature for all the specimens. And longer lifetime components ($\tau_2$) are saturated at 623 K for 30 min annealing except for pure-iron irradiated to 0.9 mdpa, and the intensity of $\tau_2$ decreased with increasing annealing temperature for all the specimens. There is no good correlation between recovery behavior of irradiation hardening and that of PAS parameters.

4. Discussion

4.1 Contribution of matrix defects

The obtained experimental results concerning about the mechanism of the matrix defect hardening suggest that I-loops have a larger contribution to irradiation hardening than V-clusters. In order to determine the contribution of each defect structure to irradiation hardening, the amount of the irradiation hardening was estimated from the microstructures obtained by TEM and PAS, using the following equations:

$$\Delta \tau_m = h \times \tau_m$$

$$\Delta I_{\tau_2} = i \times I_{\tau_2}$$
where $\alpha$, $\mu$, $b$, $N$ and $d$ are dislocation barrier strength factor for each defect, shear modulus, Burgers vector, density and diameter of dispersed particles, respectively. The $\alpha$ values were determined so that the estimated $\Delta HV$ became equal to the measured $\Delta HV$ by hardness test. As expected from the PAS results, which suggested no good correlation of the recovery behaviors between hardness and PAS, the $\alpha$ value of V-cluster is very small (0.05), while that of I-loops is 0.30. This strongly indicates that the factor controlling irradiation hardening of pure-iron is I-loops. Contributions of I-loops and V-clusters to the irradiation hardening of pure-iron are 79% and 21%, respectively.
4.2 Contribution of copper-rich precipitates (CRP)

The obtained experimental results for as-irradiated and post-irradiation annealed Fe-0.15Cu alloy indicate the invisible CRP by TEM induce a large irradiation hardening for Fe-0.15Cu alloy. The contribution of each defect structure to irradiation hardening of the Fe-0.15Cu alloy was also calculated by the above equation, using $\alpha_{\text{I-loop}} = 0.30$ and $\alpha_{\text{V-cluster}} = 0.05$. The $\alpha_{\text{Cu}}$ is calculated to be 0.065 from the diameter and density of CRP estimated by 3DAP data with the assumption that the matrix copper concentration is 0.08% and the average sizes of the precipitates are those obtained by 3DAP analysis. The obtained value is considered to be smaller than the expected value. This is probably due to over estimation of both the size and density of CRP that causes the reduction of $\alpha$ value at a total copper concentration (0.15 mass%). In this calculation, contributions of I-loops, V-clusters and CRP to irradiation hardening are 44%, 8% and 48%, respectively.

5. Conclusion

Effects of irradiation and post-irradiation annealing on the irradiation hardening and microstructural change in pure-iron and Fe-0.15Cu alloys were investigated to clarify the quantitative contribution of each defect structure to the hardening. These results and the quantitative data analysis strongly indicated that the factor controlling the irradiation hardening of pure-iron is I-loops, and those in Fe-0.15Cu alloy are both I-loops and CRP of which the contributions to the hardening are almost same.

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REFERENCES


Fig. 6 Comparison of the Vickers hardness measurement and estimated hardness from the PAS, TEM and 3DAP, showing (a) pure-iron (0.9 mdpa); (b) pure-iron (22 mdpa); and (c) Fe-0.15Cu alloy (22 mdpa).