The Origin of Residual Stress due to Thermo-Mechanical Treatment in Ni-base alloy

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1. Introduction

Residual stress is defined as the stress remaining after the stress applied to the material is removed. In particular, this residual stress is presumed to be the cause of fracture of welds or cold-worked materials. The stress inside material is determined by measuring the average distance between the lattice planes. The residual stress of a material is interpreted as present if the interplanar distance is greater or less than that without stress. In particular, dissimilar weld metals of light-water reactors are susceptible to primary water stress corrosion cracking (PWSCC), which is presumed to be due to residual stress.

Cold working of alloy 600, a light water reactor steam generator tube material, increases the PWSCC sensitivity of this material. That is, in the Ni-base alloy, thermal and mechanical treatment such as cold working, quenching, and welding increases the intergranular (IG) fracture susceptibility of these materials. Nevertheless, the exothermic reaction caused by thermal mechanical treatment cannot be explained by residual stress.

Therefore, in this study, thermal and mechanical treatment of alloy 600 was performed and differential scanning calorimeter (DSC) analysis confirmed that exothermic reaction appeared in these specimens. This exothermic reaction is due to the entropy reduction process, and it was analyzed whether lattice contraction occurred during this process. Based on these results, the relationship between lattice contraction and residual stress due to entropy reduction was discussed.

2. Experiment

The composition of alloy 600 with a diameter of 11 mm used in this experiment is shown in Table 1. For DSC analysis, the following five specimens of Alloy 600 were prepared; (1) 1095° C-1H treatment followed by water quenching (WQ), (2) 1095° C-1H treatment followed by furnace cool (FC), (3) WQ + 30% cold work (WQ+CW), (4) FC + 30% cold work (FC +CW), (5) WQ + 475^{\circ}C-10,000H aging (WQ+A). These specimens were analyzed for specific heat in DSC at a heating rate of 10 K/min.

WQ alloy 600 was subjected to accelerated aging at 400 $^{\circ}$ C higher than the reactor operating temperature for up to about 5,000 hours, and the lattice change according to the aging time was observed using a neutron diffraction. Ni-base alloys such as Alloy 600 have a face centered cubic (FCC) structure generally. In the FCC structure, the (111) plane is the plane with the

highest atomic density and is likely to have the greatest influence on the material change.

Table 1. Chemical composition of Allov 000 (wt 7	nical composition of Alloy 600 (with	%).
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С	Co	Cr	Cu	Fe	Mn	Ni	S	Si
0.009	0.06	16.25	0.05	8.15	0.32	74.55	0.002	0.31

3. Results and Discussion

Figure 1 shows the comparison of DSC results for alloy 600 subjected to thermal and mechanical treatment under various conditions. (1) WQ, (3) WQ + CW, (4) FC + CW specimens exhibited an exothermic reaction. On the other hand, (5) only endothermic reaction was observed in the WQA specimen.

Both cold-worked and WQ-treated specimens exhibit an exothermic reaction, because this heat is caused by the release of entropy remaining in the material during the cold-working or WQ process. Figure 1 shows that cold working moves the ordering reaction to around 100°C, which is much below the reactor operating temperature. This effect means that cold working makes it easy to cause a ordering reaction(entropy reaction) in the operating environment. It is important to note that this significantly alters the kinetics of damage in the operating condition.



Fig. 1. DSC results of variously treated Alloy 600.

Figure 2 shows the contraction behavior of (111) lattice after aging treatment of (1) WQ and (2) FC-treated materials at 400 °C. By aging treatment, WQ shrinks about 0.035% of (111) plane at 400°C-500 hours. The lattice contraction is saturated. That is, a material that exhibits an exothermic reaction causes

lattice contraction when it is aged. In this respect, entropy reduction is the cause of lattice contraction. They are characterized by exothermic reactions.



Fig. 2. (111) lattice variation in WQ, AC, and FC Alloy 600 aged at 400°C.

Let's discuss how a decrease in entropy causes lattice contraction. We assume that atoms are not actually spheres, but are spheres for convenience. The sphere cannot distinguish the orientation of the atoms. Therefore, this study introduces the concept that atoms are two-dimensional ellipses. The ellipse can explain that the entropy of a material increases when the direction of its major and minor axes changes.

In this study, the heat generated in the thermally mechanically treated material is due to entropy. Figure 3 schematically shows how the lattice distance changes depending on the arrangement of elliptical atoms. The above exothermic materials have an irregular arrangement and for this reason heat is stored. This is a latent heat and also the configuration entropy. When a material in this state is exposed to a temperature at which atomic diffusion is possible, heat is released as the atoms are stably aligned, and at the same time, the lattice distance becomes smaller.



Fig. 3. Atomic arrangement and dimensional variation with entropy in Alloy 600.

If residual stress is the reason for the vulnerability to PWSCC in the reactor operating environment, the exothermic reaction shown in the DSC analysis cannot be explained. Therefore, the cause of increasing sensitivity to PWSCC should be sought from other aspects. Summarizing these discussions, it is possible that the reduction in entropy may be the cause of the vulnerability to PWSCC in the operating environment of a nuclear reactor.

Thermal mechanical treatment increases entropy, which in turn increases the lattice distance. In this case, if the residual stress is obtained by measuring the lattice distance, it appears as a positive residual stress when entropy increases. In conclusion, it is because entropy remains in the exothermic reaction by thermal mechanical treatment. Therefore, the nature of the residual stress is due to the increase in arrangement(configuration) entropy.

4. Conclusion

Thermal mechanical treatment of Alloy 600 increases and retains the entropy of the material. This residual entropy causes the lattice expansion, and when exposed to operating reactor environment, the entropy decreases while the lattice contracts. Thermal and mechanical treatment of Ni-base alloy increases entropy to cause lattice expansion, and in the operating environment, atoms are aligned by atomic diffusion and the lattice is contracted. This behavior is a sufficient condition for generating tensile stress at the grain boundaries of the material and causing grain boundary failure. Therefore, the essence of increasing the sensitivity of PWSCC is the increase in entropy.

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