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Lattice Contraction Behavior due to Atomic Ordering in Alloy 600

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1. Introduction

Alloy 600 had been used as steam generator tubing and head penetration materials in commercial pressurized water reactor (PWR). It was reported that an ordering reaction occurs in Alloy 600 [1, 2]. Recently, a PWSCC (primary water stress corrosion cracking) mechanism based on an ordering reaction has been proposed [3]. However, there has been little investigation on the effects of ordering reaction on the lattice variation due to the ordering reaction in Alloy 600.

To understand the lattice variation due to the ordering reaction in Alloy 600 is important, since this is unavoidable process in nuclear reactor environment. A lattice variation with ordering treatment at 400℃ in Alloy 600 was systematically investigated using neutron diffraction.

2. Experimental

Alloy 600 rod with 10 mm diameter was used. The chemical composition is shown in Table 1. Alloy 600 was water-quenched (WQ), air-cooled (AC), and furnace-cooled (FC) from solution annealing (SA) treatment at 1095℃. These series were ordering-treated up to 5,500 hours at 400℃. These specimens were examined by high resolution neutron diffraction (HRPD), as shown in Fig. 1. The examined volume is roughly 3cm^3. The specimen is rotated during diffraction measurements. It is possible to measure the anisotropic lattice variation, since HRPD provides a focusing free characteristic. The lattice variation is calculated by equation of $(d_{ordered} - d_{as\text{-}received})/d_{as\text{-}received}$.

Table 1. Chemical composition of Alloy 600 (wt.%).

Elements	C	Cr	Fe	Ni	Mn	S
Alloy 600	0.009	16.25	8.15	74.55	0.32	0.002

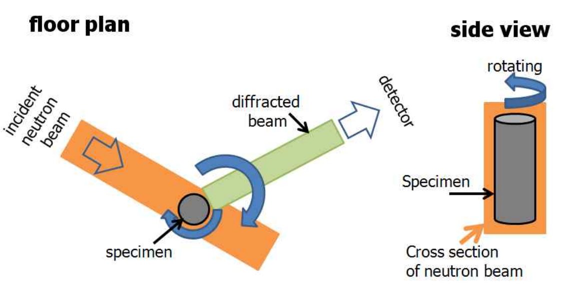

Fig. 1. Schematic illustration of diffraction condition during measurement in high resolution neutron diffraction (HRPD).

3. Results and Discussions

Fig. 2 shows the lattice variation with ordering time at 400°C in WQ specimen. The magnitude of lattice contraction in (111) and (200) appeared up about 0.04% in WQ condition, whereas, that of (220) plane is 0.02%. The lattice contraction saturates after 2,000H at 400°C. This result shows that the lattice contraction occurs anisotropic. This means that the difference in lattice contraction between high angle boundaries is large. This seems to be related to the fact that the PWSCC penetrates along the high angle boundary [4].

The lattice contractions in (111) and (200) in WQ, AC, and FC conditions are compared in Fig. 3 and 4. The magnitude of the lattice contraction in WQ is the largest. This means that the magnitude of the lattice contraction depends on the cooling rate. The magnitude of lattice contraction in AC and FC specimens is lesser than 0.02% regardless of crystallographic planes.

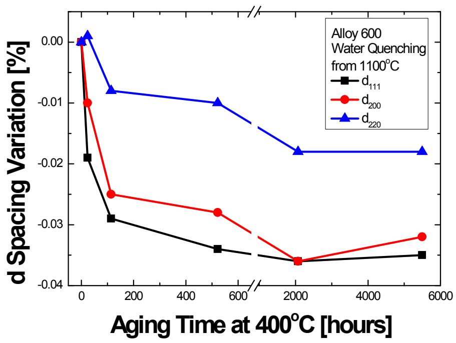

Fig. 2. Comparisons of d_{111} spacing variation with aging time at 400°C in WQ, AC, and FC Alloy 600.

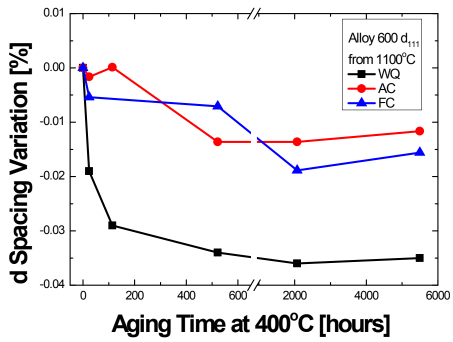

Fig. 3. Comparisons of d_{111} spacing variation with aging time at 400°C in WQ, AC, and FC Alloy 600.

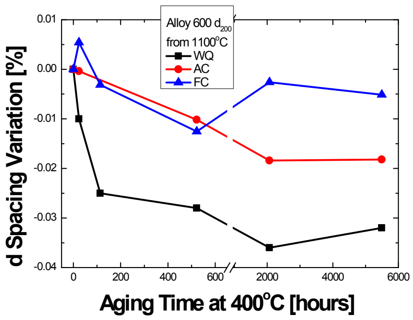

Fig. 4. Comparisons of d_{200} spacing variation with aging time at 400°C in WQ, AC, and FC Alloy 600.

Disordering reaction occurs during SA treatment at 1095°C. The lattice contraction due to the ordering treatment at 400°C is natural, since the increase in the number of ordered bonds makes the atomic distance. Specially, the number of disordering bonds in WQ is increased by rapid cooling from 1095°C. The number of disordering bonds depends on the cooling rate. The FC condition provides enough time to form the ordering bonds. Therefore, the magnitude of lattice contraction in WQ specimen is the largest. It is understood that the magnitude of lattice contraction depended on the cooling rate, and decreased with cooling rate.

The lattice contraction appeared to be anisotropic according to the crystallographic planes. This suggests that the compatibility of grain boundary (GB) could not be maintained when the lattice contraction occurs due to the ordering reaction in the component made of Alloy 600 in the nuclear reactor environments.

The ordering reaction can be detected by DSC [1, 3, and 5]. The appearance of exothermic reaction is indirect evidence for the ordering reaction in WQ Alloy 600. The ordering reaction is a kind of chemical reaction. Thus, the rate of ordering reaction is governed by Arrhenius relationship. The kinetics ordering reaction in nuclear reactor condition may be slower. The ordering reaction occurs regardless of cooling rate in nuclear reactor environment, although the exothermic reaction is observed at 450°C in WQ specimen.

There are two kinds of ordering reaction in Alloy 600 [5]. One is a thermal ordering. This is occurred by thermal diffusion without strain. The other is a strain induced ordering (SIO). This occurs during deformation at high temperature. The SIO occurs immediately when strain is applied at high temperature, whereas the first one is relatively slow because this is driven by pure diffusion.

The SIO and strain induced disordering (SID) occurs simultaneously when deformation occurs. The SID is dominant at below 150°C, whereas the SIO is dominant at above 150°C [5]. On the other hand, SID occurs predominantly at above 600°C. This temperature depends on the strain rate, since the deformation changes the atomic position continuously.

The ordering reaction is an unavoidable process in nuclear reactor operating condition. Therefore, the effect of lattice contraction due to ordering reaction should be considered in the assessment of integrity in the components made of Alloy 600, since the ordering reaction is a spontaneous and an unavoidable phenomenon below 520℃ in Alloy 600.

4. Conclusions

1. The magnitude of lattice contraction due to the ordering reaction is lager when the cooling rate is faster from the disordering temperature.
2. The ordering treatment causes anisotropic lattice contraction according to the crystallographic planes. The ratio of minimum to maximum lattice contraction in WQ Alloy 600 is about 2.
3. The maximum lattice contraction occurs in (111) and (200) planes are similarly about 0.04% in WQ Alloy 600. This saturates after 2,000H at 400°C.
4. The effect of ordering reaction should be considered in the assessment of integrity of primary boundary materials made of Alloy 600, since the ordering reaction is an unavoidable phenomenon in nuclear reactor environment.

Acknowledgements

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