# A High Temperature Neutron Diffraction Study in Aged Alloy 690

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## 1. Introduction

There is an order-disorder reaction in Alloy 600. This has been proven by a DSC (differential scanning calorimetric). The short range order (SRO) forms during an aging under a critical temperature for ordering reaction [1, 2]. The type of order phase is a $Ni_2(CrFe)$ based on $Ni_2Cr$ with orthorhombic structure.

It is known that Alloy 690 has a better resistance to PWSCC (primary water stress corrosion cracking) in nuclear reactor environment. From this point of view, the Alloy 690 is a standard material for steam generator in Korean standard nuclear plant (KSNP). Therefore, it is important to understand the effects of ordering on physical properties in the high temperature in Alloy 690.

The ratio of Ni:Cr in Alloy 690 is close to 2:1 like $Ni_2Cr$. However, Alloy 690 contains about 10% Fe. Marucco has reported that the presence of Fe has a strong delaying effect on the ordering kinetics, based on results of the study of an ordering transformation in Ni–Cr base alloys from the study of $Ni_2Cr$ and Ni–Cr–Fe alloys [3]. Furthermore, it is reported that most commercial Ni–Cr based alloys have an ordering reaction below critical temperature [4,5].

In this study, in order to understand the effects of an ordering reaction in Alloy 690 alloy, a high temperature neutron diffraction study was carried out from room temperature (RT) to 700 °C.

## 2. Experimental

Alloy 690 steam generator tubing was water quenched from the solution annealed temperature at 1095°C. Because it is expected that an ordering progresses with an aging under Tc, Alloy 690 tube were aged at 474°C for 80,000 hours in order to identify the effects of a long term aging.

This aged specimen was cut along the axial direction with 5 mm width and 50 mm long, and these pieces were put in the vanadium can. This specimen was examined by neutron diffraction from RT to 700 °C under a vacuum. The wavelength of the neutron beam was 1.837225 A. The axial direction specimen was aligned vertically and it was rotated along the center line of the rod during a measurement.

The ordered state is maintained below 600 °C in the high temperature neutron diffraction during heating, since a disordering reaction in alloy 690 occurs at 700 °C. Therefore, a disordered state of Alloy 690 is measured during cooling. The measurements temperatures were at RT, 300, 450, 500, 550, 600, 700 °C.

The d spacing was calculated by a integrated center of gravity. The thermal expansion coefficients according to crystallographic direction were calculated in each temperature interval.

$$\alpha_{hkl} = (d_{THhkl}-d_{TLhkl})/(d_{TLhkl}*(T_H-T_L)) \qquad (1)$$

where, $\alpha_{hkl}$ is a average thermal expansion coefficient, $d_{THhkl}$ is a d spacing at a higher temperature, $d_{TLhkl}$ is a d spacing at a lower temperature, $T_H$ is the higher temperature, and $T_L$ is the lower temperature.

## 3. Results and Discussions

Fig. 1 and 2 shows shifts of (111) and (311) diffraction peaks. This is due to the expansion of inter-planar spacing with temperature. This is natural that the inter-atomic distance increases with temperature because of increasing of thermal vibration.

It is possible to calculate the thermal expansion coefficient in each direction, as shown in equation (1). Those in [111] direction are shown in Table 1. The thermal expansion coefficient increases with temperature. The diffraction intensity increased at 700 °C in Fig. 1 and 2. This seems to be due to the rearrangement of atom by disordering reaction and the annealing effect at this temperature.

The ordered Alloy 690 does not show superlattice peaks, although an aging treatment at 474 °C for 80,000 hours makes $Ni_2(CrFe)$ ordered phase. However, this specimen maintains the ordered state until its temperature reaches Tc (600 $^{o}$C). Thus, the diffraction results during a heating and a cooling can be thought simply as the ordered and the disordered or lesser ordered states.

The other difference between the ordered and the disordered state is a contraction of lattice in the ordered state and an expansion of the lattice in the disordered state. The positions of peaks at RT are not identical after cooling. This means that LRO is not formed by the FC process only. This behavior is consistent with the previous results [5].

Each step of diffraction measurement takes two hours except for the temperature controlling time. Therefore, the disordered specimen stayed at least for 8 hours at 300~550°C in the neutron diffraction measurements during cooling.

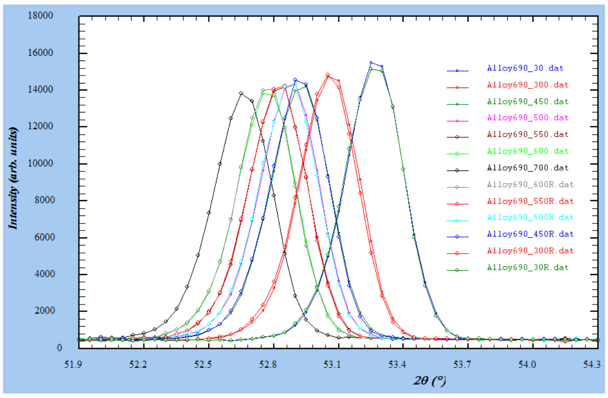


Fig. 1. Comparisons of position of (111) peaks with temperature by neutron diffraction in Alloy 690.

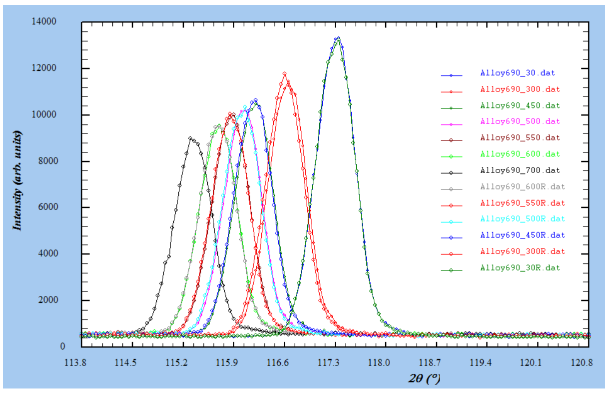


Fig. 2. Comparisons of position of (311) peaks with temperature by neutron diffraction in Alloy 690.

Table 1. Thermal expansion coefficient in [111] direction during heating in Alloy 690 aged for 80,000H at 475 $^{o}$C.

| Temperature [$^{o}$C] | Thermal Expansion Coefficient in [111] during heating , averaged between measurement interval [$K^{-1}$] |
|---|---|
| 30 → 300 | 1.329E-05 |
| 300 → 450 | 1.673E-05 |
| 450 → 500 | 1.818E-05 |
| 500 → 550 | 1.821E-05 |
| 550 → 600 | 1.825E-05 |
| 600 → 700 | 2.007E-05 |

## 4. Summary

The high temperature neutron diffraction study up to 700°C in Alloy 690 shows a hysteresis in thermal expansion coefficient. In addition, the thermal expansion coefficient is anisotropic according to direction of crystal. This can be explained by ordering and disordering reaction. Ordering process in Alloy 690 induces a line broadening in all plane, however, the magnitude of the broadening varies from plane to planes. Furthermore, the lattice contraction occurs during an ordering process. In contrast, lattice expansion occurs during a disordering process above Tc at 700°C.

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