**In-Situ** Measurement of Internal Stress Distribution Change of TT600 by Energy-Dispersive X-ray Diffraction with White X-ray Micro Beam

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Under the primary conditions of a pressurized water reactor, the intergranular stress corrosion cracking susceptibility of a Ni based alloy is improved by chromium carbide precipitation in grain boundaries. The effects of chromium carbide precipitation treatments are explained by the grain boundary strengthening mechanism, the intergranular corrosion prevention mechanism, and so on. However, few studies have demonstrated these mechanisms, and the effects on intergranular stress corrosion cracking susceptibility are not completely understood. Therefore, for the purpose of demonstrating the change in the internal stress distribution with or without grain boundary carbide precipitation treatment in the 600 alloy, in situ measurements were performed in this study using the X-ray diffraction technique with the BL-28B2 beamline at the SPring-8 synchrotron radiation facility in Japan, to clarify the improvement in the mechanical properties of the alloys by TT treatment.

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### Table 1 Chemical composition of specimens (mass%).

| Element | Specimen |
|---------|----------|
| C       | 0.03     |
| Si      | 0.27     |
| Mn      | 0.29     |
| P       | 0.010    |
| S       | 0.001    |
| Ni      | 72.90    |
| Cr      | 16.20    |
| Fe      | Bal.     |
developed at SPring-8 for examining the inner strain distribution of specimens. The EXDM analyzes strain based on the lattice spacing of an X-ray irradiation position using the X-ray diffraction pattern in a transmission configuration. In this method, X-ray beam size is controlled to be smaller than the grain size because the irradiation position must be considered as a single crystal. The EXDM measurement was carried out on the BL-28B2 beamline at SPring-8. The white X-ray beam on the BL-28B2 can be formed into 10 μm × 10 μm which was sufficiently small for the crystal grain size.

The specimens were set on a small tensile tester with no load, and the tensile tester was installed on the sample stage of a multiaxis diffractometer on BL-28B2. Grain boundary image information was acquired from the transmitted Laue patterns obtained by imaging the specimens with the X-ray beam.10) Figures 2(a) and (b) show the grain boundary images of the TT-untreated and TT-treated specimens, respectively.

The EXDM measurements were performed according to the following procedure. Measurement points were selected with reference to the obtained grain boundary images. The measurement points indicated by the red dots in Fig. 2(a) and (b) were extracted from the grain boundary and trans-grain of each specimen. Sixty points were selected for the TT-untreated specimen, among which 36 points were around the grain boundary and 24 points were in the trans-grain. In the case of the TT-treated specimen, fifty points were selected. Twenty five out of this 56 points were around the grain boundary and the other 25 points were in the trans-grain. Each measurement points was irradiated with the white X-rays beam to obtain transmitted Laue images. Twenty strong spots were selected in each transmitted Laue image. Thereafter, a diffraction pattern was obtained from each spot using a semiconductor detector. The same measurements were performed by applied stress within the elastic strain range determined by stress-strain curves, as shown in Fig. 3. Figure 3 shows the stress-strain curve of the sample prepared via TT treatment from 0 to 15 h. In the strain measurement by the measurements of the EXDM, the stresses applied in the tensile direction were 64 MPa and 150 MPa for the TT-untreated specimen and 50 MPa and 150 MPa for the TT-treated specimen.

The stress in each sample at the X-ray irradiation position was analyzed according to the following procedure: To analyze the elastic strain, the lattice spacings of three or more independent lattice planes were calculated using the X-ray diffraction peaks obtained at each irradiation position. The strain-free lattice plane spacing was used as the average value of the lattice constants obtained from all measured data without applied stress. Tensor analysis was performed by assuming that the stress parallel to the beam direction, that is, in the direction perpendicular to the specimen surface, was released because the grain size and specimen thickness were approximately equal. Here, the elastic constant of Ni obtained from literature was employed as the elastic constant in the tensor analysis; $c_{11} = 25.08 \times 10^{10}$ Pa, $c_{12} = 15.00 \times 10^{10}$, $c_{44} = 12.35 \times 10^{10}$.11)

![Fig. 1 Schematic view of tensile specimen.](image1)

![Fig. 2 Grain boundary images visualized from the specimen. The images includes the measurement positions. Red lines are the grain boundaries. Red points indicate measurement points. Grain boundary images of (a) TT-untreated 600 alloy and (b) TT-treated 600 alloy.](image2)

![Fig. 3 Time dependence of the stress-strain curves for thermally treated 600 alloy. Numerical values represent the time required for thermal treatment.](image3)
3. Results and Discussion

3.1 Change in principal stress of TT-untreated specimen due to applied stress

Figures 4(a), (b), and (c) show the principal stress conditions at each point of the TT-untreated specimen at applied loads of 0 MPa, 64 MPa, and 150 MPa, respectively. The red points indicated the measurement points, the red arrows indicate tensile stress and the blue arrows indicate compressive stress. Here, the stress at the red points without red and blue arrows could not analyzed because the information about the three independent lattice plane spaces could not be obtained. The 31 points for grain boundaries and the 19 points for interior of grains were analyzed consistently during the experiments.

As shown in Fig. 4(a), stress was observed at the measurement points even under no load. As a lattice number of distorted Laue spots were observed in the acquired transmission Laue images, it was probable that the residual stress formed at the time of sample preparation was detected. Figures 4(b) and (c) show that the main stress direction changed by applying applied tensile stress in the horizontal direction. Comparing the stress conditions between grain boundaries and trans-grains, the stress around the grain boundaries showed an increasing trend. Table 2 shows the average of the maximum principal stress at each applied stress and the ratio of the tensile stress and maximum principal stress at the grain boundaries and trans-grains. The measured stress increased with the applied stress, and the number of the points indicated that the maximum principal stress of tension in the grain increased significantly. The principal stress increased considerably at the grain boundaries and trans-grain.

3.2 Change in principal stress of TT-treated specimen due to applied stress

Figures 5(a), (b), and (c) shows the principal stress at each point of the TT-treated specimen at applied loads 0 MPa, 50 MPa, and 150 MPa, respectively. The points indicated by only red circles are the measurement points that could not be analyzed using the data obtained in the experiments. The number of points that could not be analyzed was larger for the TT-untreated specimen compared with the TT-untreated specimen. As inclusions precipitated with the grain boundaries, the diffraction patterns from the inclusions and base material were mixed, and the three independent lattice spacings required for the strain analysis could not be obtained. The 14 points for grain boundaries and the 17 points for interior of grains were analyzed consistently during the experiments. Stress was observed at each analysis point even without applied load, as shown in Fig. 5(a). It was considered that the residual stress formed during sample preparation was detected. As shown in Fig. 5(b) and (c), the principal stress direction changed according to the stress distribution in the specimen depending on the applied tensile stress.

| Position       | 0   | 64  | 150  |
|----------------|-----|-----|------|
| External stress (MPa) |     |     |      |
| Ratio of tensile stress in maximum principal stresses (%) | 89.3 | 94.1 | 96.4 | 88.2 | 89.3 | 100 |
| Mean value of maximum principal stresses | 391.9 | 386.2 | 522.3 | 413.4 | 784.6 | 760.7 |
 stress in the horizontal direction. Table 3 shows the average of the maximum principal stress at each analysis points and the maximum principal stress at the grain boundaries and trans-grains. The number of points indicated that tensile stress increased with the external force in the horizontal direction. Even though the maximum principal stress increased with the applied stress, the maximum principal stress at the grain boundaries and trans-grains was lower compared with the TT-untreated specimen.

### 3.3 Equivalent stress changes

The difference in the internal stress distributions of the TT-treated and TT-untreated specimens under the applied tensile stress was measured by the EXDM. According to the changes in principal stress obtained through tensor analysis, the following results were confirmed: In the TT-untreated specimen, the stress in the grain boundary increased and the maximum principal stress increased with strain. In the case of the TT-treated specimen, the principle stress changed negligibly compared with the TT-untreated specimen. The magnitude and direction of the principal stress at each measurement point changed according to the applied stress, as shown in Fig. 4 and 5. These behaviors were related to the individual shapes of the crystal grains and the difference between each three-dimensional crystal grain arrangement and connectivity. Thus it was considered that the direction of a stress changed in a complex manner. To compare the stress between the grain boundaries and trans-grains, the principal stress, which was a complex vector with directionality, was converted into equivalent stress, which was a scalar value. The von Mises equivalent stress at each point was calculated from the eq. 1.

$$
\sigma_e = \sqrt{\frac{1}{2}((\sigma_1 - \sigma_2)^2 + \sigma_1^2 + \sigma_2^2)}
$$

Here, \(\sigma_e\) is the equivalent stress at each point and \(\sigma_1\) and \(\sigma_2\) are the maximum and minimum principal stresses at each point, respectively. The average values of the equivalent stress at the grain boundaries and the trans-grains, \(\sigma_e(GB)\) and \(\sigma_e(TG)\), were calculated. The ratio of these values, \(\alpha\), was estimated as shown in eq. 2. Figure 6 shows the changes in the equivalent stress and the \(\alpha\) dependent on the applied stress based on the absence of applied stress on the TT-untreated and TT-treated specimens, respectively.

$$
\alpha = \frac{\sigma_e(GB)}{\sigma_e(TG)}
$$

As shown in Fig. 6, the internal stress of both specimens increased with the applied stress. These tendencies varied depending on thermal treatment and the analysis positions of the specimens. The changes in the equivalent stress of the TT-untreated specimen with applied stress was larger around the grain boundary compared to the trans-grain. In contrast, the change in the equivalent stress of the TT-treated specimen with applied stress was larger in the trans-grain compared to the grain boundary. The changes in the equivalent stress and the \(\alpha\) are shown in Fig. 6.

### Table 3 Maximum principal stress at the grain boundary (GB) and trans-grain (TG) of the thermally treated specimen.

| Position | 0              | 50             | 150            |
|----------|----------------|----------------|----------------|
| GB       | 77.3           | 100.0          | 94.4           |
| TG       | 73.7           | 100.0          | 81.0           |
| Ratio of tensile stress in maximum principal stresses (%) | 150.0 | 279.8 | 316.5 |
| Mean value of maximum principal stresses | 120.9 | 215.6 | 322.2 |

![Fig. 5 Principal stresses under applied tensile stresses acting in horizontal direction on the thermally treated specimen. Red and blue arrows indicate tensile stress and compressive stress, respectively. The points represented by only red dots cannot be analyzed. (a) No applied stress, (b) 50 MPa and (c) 150 MPa.](image)
with the grain boundary. A significant difference was observed in the equivalent stress in the grain boundary between the TT-treated and TT-untreated specimens. The equivalent stress of the TT-untreated specimen was larger than that of the TT-treated specimen, and the stress of the TT-untreated specimen was concentrated in the grain boundary. This can be clearly seen in the behavior of $\alpha$. In the TT-untreated specimen, $\alpha$ increased with applied stress, whereas it remained almost constant when applied stress was applied to the TT-treated specimen. That is, even though stress was concentrated close to the grain boundary in the TT-untreated specimen, the stress concentration was relieved by the TT treatment. As a result of the relaxation of stress around the grain boundary, IGSCC sensitivity decreased in the TT-treated specimen.

Bruemmer et al., Miglin et al., and Kawamura et al. pointed out that the stress concentration close to the grain boundary might be suppressed by dividing the inclusions of the grain boundary. The difference in the stress behavior close to the grain boundary with and without the TT treatment provided a clear experimental fact against the prediction that the TT treatment would disrupt stress propagation at the grain boundary.

4. Conclusion

The change in internal stress under applied stress was examined via EXD to clarify the effect of TT treatment on mechanical properties. The following conclusions were obtained:

(1) In the TT-untreated 600 alloy, the vicinity of the grain boundary was more sensitive to the applied stress changes compared with the grain.
(2) The stress concentration close to the grain boundary was relieved by the TT treatment.
(3) Internal stress analysis based on the EXDM measurement was confirmed as an effective tool.

The effect of the TT treatment is that the stress due to bias a stress toward the grain boundaries of TT-untreated material is separated from the progression of dislocations caused by stress in the presence of grain boundary inclusions. This is consistent with the idea of stress homogenization in the structure.

This was the first time that the stress concentration closed to the grain boundary was alleviated by the TT treatment, and this was proved through the in-situ stress distribution measurement.

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