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Effect of Residual Stress on Oxidation Behavior of Stainless Steel 304L in Simulated Secondary Water Environment of PWR

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1. Introduction

Austenitic stainless steel is widely used material in many heat exchanging facilities, including secondary circuit of pressurized water reactors (PWRs). Type 304L stainless steel is one of the representative materials for the feedwater heaters (FWH) that support generation sufficiency of steam generators by pre-heating secondary water with generation-used hot steam. To reduce the corrosion on materials, secondary water chemistry is strictly controlled to obtain pH over 9.5 [1]. However, some failures have been reported before the service lifetime of FWH tubes, even with no trouble on chemistry control. Residual stress applied from plastic deformation and component installation is expected to be the major factor in the occurrence of stress corrosion cracking (SCC). As the corrosion behavior of material can be major factor in SCC initiation, in combination with stress, it should be clearly clarified for prevention and analysis of SCC phenomena in a secondary water environment.

Extensive residual stress shows lower corrosion resistance in stainless steel by electrochemical study [2]. However, the effect of residual stress has not been evaluated with various levels of residual stress in secondary water environment so far. Furthermore, the detailed oxidation mechanism should be deeply explored with microstructure analysis and empirical tests to simulate reported issues. Therefore, this research evaluated the effect of residual stress on corrosion behavior of stainless steel with combinational methods in a secondary water environment of PWR.

In this study, 304L stainless steel was simulated to be subjected to various residual stresses by various levels of cold rolling, and hole drilling method was conducted for measurement. To evaluate corrosion behavior of materials, polarization test and immersion oxidation test were done with oxide thickness measurement after cross-section observation. According to the results, the polarization test results showed the corrosion susceptibility of material with residual stress in the corrosion process, and additional electrochemical impedance spectroscopy (EIS) test results showed instability of passivation layer with residual stress. From microstructure analysis, materials with residual stress showed higher fraction of corrosion-susceptible grain boundaries, including random and high sigma coincidence site lattice (CSL) boundaries, which were also empirically observed by cross-section electron backscatter diffraction (EBSD) analysis.

2. Experimental

2.1 Material

Solution annealed stainless steel 304L plate was prepared, followed by cold rolling with reduction rate of 3 %, 4 %, 6 %, named CW 3, CW 4, and CW 6, respectively. The measured residual stress levels were determined by hole drilling, and chemical composition of each material is listed below. Prior to overall tests, each sample was polished with emery paper up to 800 grits and diamond suspension up to 1 μm. After the polishing, the samples were rinsed in the following order: acetone, ethanol, and distilled water.

Table 1. Chemical composition of 304L stainless steel plate used in this study [wt%]

Fe	C	Si	Mn	P	S
Bal.	0.024	0.45	1.43	0.033	0.003
Ni	**Mo**	**Cr**	**Co**	**Cu**	**Ti**
8.11	0.18	18.35	0.17	0.28	0.002

Table 2. Measured residual stress level of each material [MPa]

CW 3	CW 4	CW 6
118	197	354

2.2 Electrochemical test

Potentiodynamic polarization test was conducted in secondary water chemistry shown on the list below. The scan rate applied was 0.1667mV/sec, with a scan range of -0.6 V to 1.6 V. Dissolved oxygen was controlled by Ar purging for 1 hr before each experiment, and potential stabilization was achieved with open circuit measurement. A 3-electrode system was used, consisting of a Pt mesh counter electrode, a saturated Ag/AgCl reference electrode, and a working electrode.

Electrochemical impedance spectroscopy test was conducted on 2,000 hr corrosion specimens with the same 3-electrode system. The scan range was 3 MHz to 0.1 Hz with a scan voltage of 7 mV.

Table 3. Secondary water chemistry applied on this study

ETA [ppm]	N_2H_2 [ppb]	NH_3 [ppb]	pH
5.73	124.2	207.6	9.82

2.3 Immersion oxidation test

Coupon (10 mm * 15 mm * 3mm) corrosion specimens were exposed to the secondary water environment of 1 gallon autoclave with loop facility. The specimens were hung on SS316L wire covered with zirconia tubes to prevent galvanic corrosion. Corrosion specimens were collected after 600, 1200, 2000 hr of exposure and analyzed to measure oxide thickness and cross-section EBSD. The test water chemistry is shown in table 3, and the operation condition was selected the harshest temperature and pressure.

Table 4. Operation condition of the harshest part of feedwater heater

Chemistry	Temperature [℃]	Pressure [MPa]	DO [ppb]
Table 3.	235	10.0	< 5

3. Results and Discussions

3.1 Potentiodynamic polarization test result

Polarization test results for each specimen are shown on the graph below. Although the residual stress level showed no distinguishable difference in the results, materials with residual stress showed lower corrosion potential and higher corrosion current density than solution annealed material. This tendency can be explained by the wider interatomic distance caused by residual stress, which make the material more susceptible to corrosion [2].

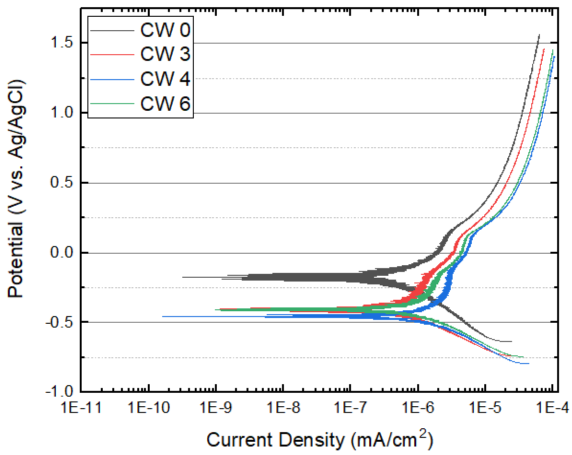

Figure 1. Potentiodynamic polarization test result

3.2 Immersion corrosion test result

The thickness of the passive Cr oxide layer was measured after exposure for 600, 1200, 2000 hr in the secondary water environment. Until the 1200 hr, there was no significant difference in thickness observed throughout the entire specimen. However, after 2000 hr of oxidation, higher oxide thickness was observed on CW 4 and CW 6, indicating that the protective roll of the passive layer was impaired due to residual stress.

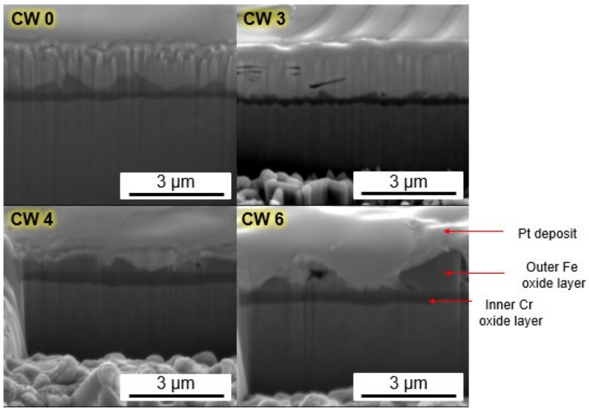

Figure 2. Cross-section SEM image of 2,000 hr oxidized specimens

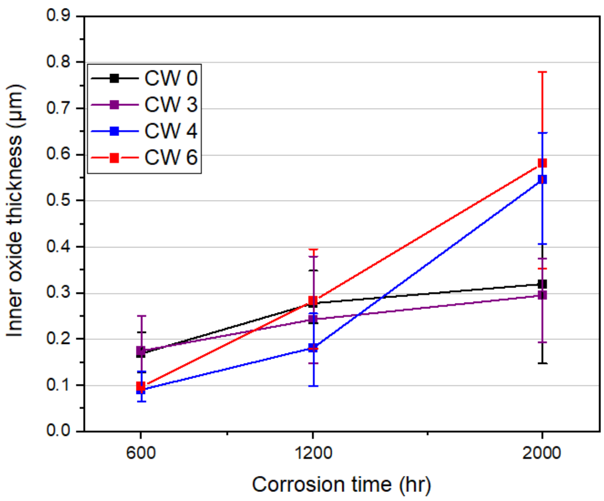

Figure 3. Inner oxide thickness measured during exposure time to 2000 hr

EBSD analysis was conducted on cross-section of oxidized specimen to investigate its oxidation behavior. Figure 4 shows a representative image of CW 3 with CSL boundary, and a comparison of the CSL boundary fraction throughout surface of entire specimens. Oxidation acceleration was observed along corrosion susceptible paths such as slip and random/high CSL boundaries. These results are showing the corrosion resistance decreased with increased residual stress.

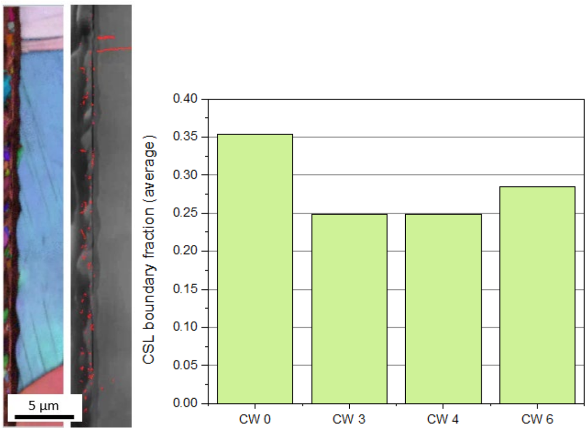

Figure 4. Cross-section EBSD image of inverse pole figure and CSL with fraction comparison

3.3 Electrochemical impedance spectroscopy test result

The effect of oxide passive film and solution was considered in circuit [3] for EIS results, and the fitted Bode plot showed a good fit across the entire data set. The R_{ct} collected from the Nyquist plot showed a decrease with an increase in residual stress. This tendency towards a low R_{ct} can be explained by the fact that the passivity of oxide layer was lower with residual stress, even though the thickness of passive layer was thicker [4].

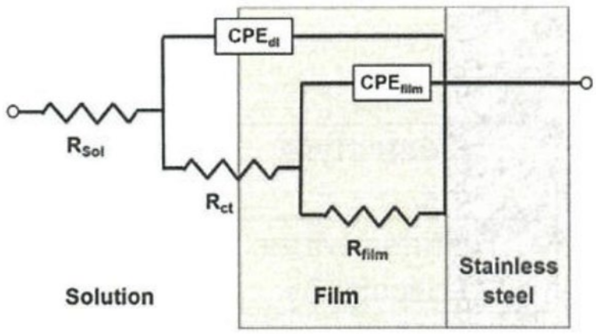

Figure 5. Circuit used in this study for considering film and solution effect[3]

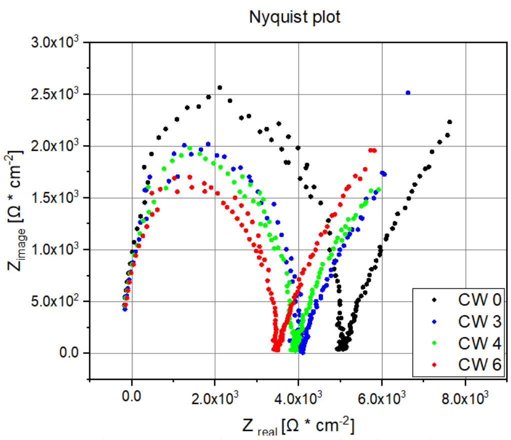

Figure 6. Nyquist plot data of each specimen

5. Conclusion

In this study, the effect of extensive residual stress on the oxidation behavior of stainless steel in a secondary water environment was studied using electrochemical and empirical methods. Polarization tests and comparison of passive layer thickness showed lower corrosion resistance in specimens with residual stress. Additionally, formed passive layer after oxidation on CW 3, 4 and 6 exhibited a lower R_{ct}, indicating instability of the passive layer. EBSD analysis conducted on cross-sections of immersion corrosion specimens showed oxidation acceleration along slip and corrosion susceptible grain boundaries. It was revealed that residual stress on stainless steel increased the fraction of corrosion susceptible path and led to less protective passive layer formation.

To investigate the SCC behavior as a future study, SCC test has been conducting with the addition of external load using proving ring structure.

7. Acknowledgement

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