

Effect of surface roughness on corrosion behavior of Alloy 690TT in simulated primary coolant of PWRs

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1. Introduction

Nickel-based alloy has been widely used for structural materials in the nuclear power plants (NPPs) due to its superior corrosion resistance and high temperature strength. In particular, Alloy 690TT is an excellent material, which is the superior resistance to an intergranular attack (IGA) and stress corrosion cracking (SCC), for the steam generator tubes in pressured water reactors (PWRs).¹⁾ However, a corrosion degradation of the steam generator tubes in primary coolant system is inevitable despite its superb properties for corrosion resistance.¹⁻²⁾ Such initiation of corrosion degradation during operation causes the formation of corrosion products and release the metal ions to primary water.³⁻⁴⁾ The corrosion products transported into the primary coolant may be activated in the core or re-deposited on the surface of components in primary coolant system including fuel cladding. Nickel cations released from the steam generator tubes cause the serious problems in PWRs, for instance, radiation exposure during maintenance operation, occurrence of axial offset anomaly (AOA) and reduction in plant operation.⁵⁻⁶⁾

Some studies have been carried out to characterize the oxide layer grown on a Ni-base alloy in primary water chemical condition, especially on morphology and chemical composition.⁷⁻⁹⁾ The oxide layer was deemed to a duplex structure composed to an inner Cr rich layer and an outer spinel layer rich in iron or nickel. The composition and morphology of oxide layer are affected by surface finish, operating condition, and water chemistry. Above all, surface roughness, which is one of the commercial requirements for steam generator tube of PWRs, is extremely important in terms of corrosion degradation and corrosion products.

In this study, we report the effect of surface roughness on corrosion behavior in simulated primary water at 330 °C and at 150 bar. The surface roughness was controlled to evaluate its difference in corrosion property using mechanical grinding and polishing method. It was measured using a surface profiler and morphology of oxide layer was investigated with FE-SEM. The corrosion rate of Alloy 690TT samples was calculated by descale-gravimetric analysis.

2. Methods and Results

2.1 Material and sample preparation

The specimens (15×45×1.5mm) used in this experiment were meal annealed at 1100 °C for 5 min and then thermally treated at 715 °C for 10 hr. The chemical composition of Alloy 690 used in this experiment is listed in Table 1. The specimens ground with SiC abrasive paper to #220, #440, #1000, #2000 and mechanically polished with 0.3 μm Al₂O₃ powder. The surface roughness was measured using surface profiler. Fig. 1 shows the surface roughness RMS (root mean square) value of the samples. The RMS values were declined as decrease of particle size in SiC sand paper and Al₂O₃ powder.

Element	Ni	Cr	Fe	Mn
wt%	61.2	28.3	9.4	1.1

Table 1 Chemical composition of alloy 690TT samples

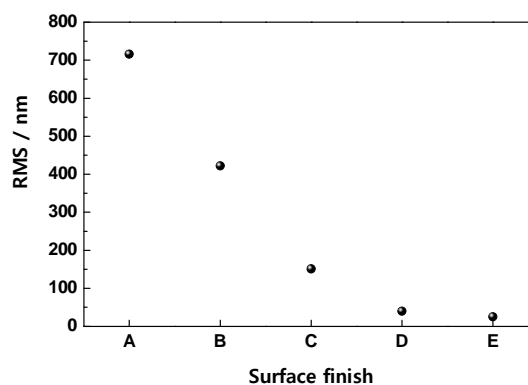


Fig. 1 Surface roughness controlled of alloy 690TT samples with different states

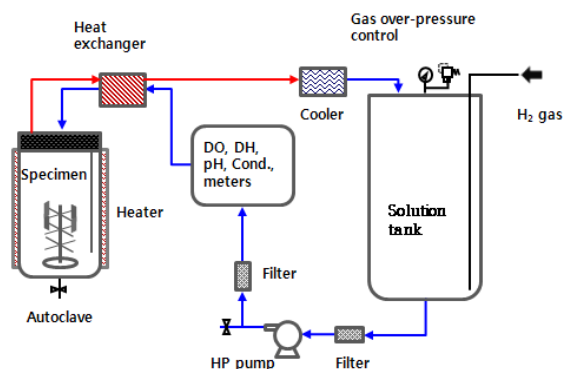


Fig. 2 Schematic diagram of the loop of simulated primary coolant for the corrosion rate test

2.2 Corrosion rate test

Simulating loop of primary coolant internal circulating for the corrosion release test was designed as shown in Fig. 2. The primary water was prepared by dissolving 2 ppm LiOH and 1500 ppm H_3BO_4 into deionized (DI) water and then adjusting pH_{330°C} of 6.7. The dissolved oxygen was controlled to be less than 5 ppb and dissolved hydrogen maintained 35 cc/kg. The corrosion rate test was performed at 325 °C and 150 bar for 500 hr. After corrosion rate test, samples were kept in a vacuum desiccator for 1day and then corrosion rate was calculated by descale-gravimetric analysis using two-step alkaline permanganate-ammonium citrate (AP/AC) descaling process.¹⁰⁾ AP solution was consisted of 1% $KMnO_4$ and 5% NaOH and AC solution was made up 5% $(NH_4)_2$ citrate. Each solution was prepared in 1L water bath at 90 °C. Descaling carried out during 5 min in each solution for 4 times, making a total process time of 40 min.

2.3 Analysis of oxide layer

Fig. 3 shows the SEM image of the oxide layer after immersion test. For the grounded samples, polyhedral oxide particles having the size of 80~120 nm were formed randomly on the ground surfaces and oxidation mainly occurred along the scratches as shown in Fig. 3 (a) and (b). On the other hand, the sample polishing mechanically having mixed oxides of polyhedral and needle-like structure, which have about length of 50 nm and width of few nm, were formed on the polished surface as shown in Fig. (c).

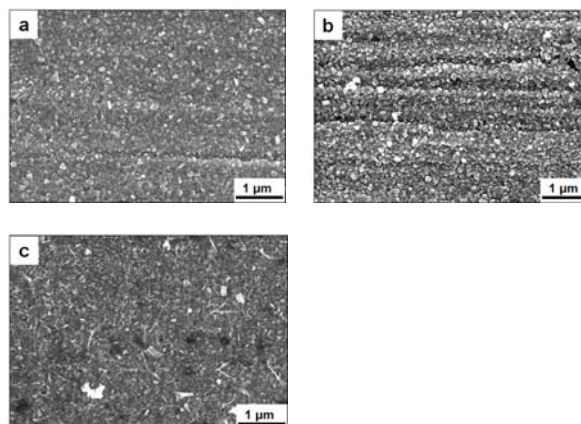


Fig. 3 SEM images of alloy 690TT samples with different surface states, (a) ground to #400 (b) ground to #2,000 (c) mechanically polished with 0.3 μm Al_2O_3 powder.

2.4 Corrosion rate

Descaling-gravimetric analysis was performed to calculate the corrosion rate of samples using a weight loss of specimens. It carried out 2-step descale of AP/AC for effective removal of oxides on a stainless steel or a nickel-based alloy. Repeated treatment was required for complete removal of oxides. It also was necessary to choose an inflection point for accurate corrosion rate.¹¹⁾ Fig. 4 shows the corrosion rate with the difference of surface roughness. The corrosion rate declined gradually in accordance with the decrease of RMS value of samples. Corrosion rates were appeared from the maximum of 0.37 mg/m^2h to the minimum of 0.12 mg/m^2h . Above all, corrosion rates of samples which have the RMS value than 151 nm (Fig. 4 (c) ~ (e)) were similar. The lowest corrosion rate (Fig. 4 (e)) was 0.13 mg/m^2h and it had the minimum RMS value. The corrosion rate of samples was declined approximately 160% as RMS values decreased from about 450 to 150 nm.

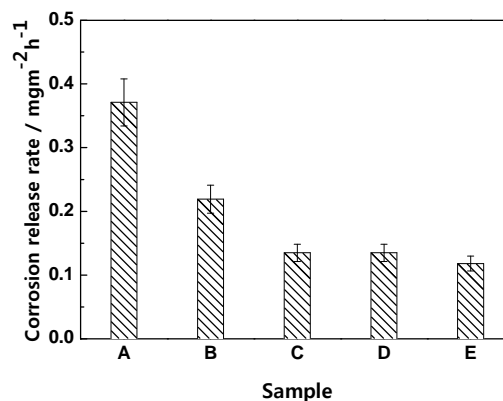


Fig. 4 Variation of the corrosion rate as surface states

3. Conclusions

We investigate the effect of surface roughness on corrosion behavior of Alloy 690TT in the simulated primary coolant environments of a pressurized water reactor.

- 1) Surface roughness of samples were controlled from 716 nm to 25 nm to evaluate for the effect of surface roughness on corrosion behavior
- 2) Polyhedral oxide particles were formed on the grounded surfaces, whereas the mixed oxides of polyhedral and needle-like structure formed on the polished surface.
- 3) The corrosion rate of samples was declined approximately 160% as RMS values decreased from about 450 to 150 nm.

Acknowledgements

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