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STRESS CORROSION CRACKING PROPERTIES OF STEAM GENERATOR TUBING ALLOYS IN CREVICE ENVIRONMENT

The safe and reliable operation of pressurized water reactors (PWRs) depends on the integrity of structural material. In particular, the failure of steam generator (SG) tubes on the secondary side is one of the major concerns of operating nuclear power plants. To establish remediation techniques and manage damage, it is necessary to articulate the mechanism through which various impurities affect the SG tubes. This research aims to understand the effect of impurities (e.g., S, Pb, and Cl) on the stress corrosion cracking of Alloy 600 and 690.

Keywords: Stress corrosion, Nickel base alloy, Nuclear plants, Secondary side, Passive film

1. Introduction

The steam generator (SG) of a nuclear power plant is a heat exchanger that generates steam that drives turbines on the secondary side. If the SG is damaged due to corrosion, radioactive materials leak from the primary system to the secondary system, which affect the safety of nuclear power plant [1].

Alloy 600 (Ni-15Cr-9Fe) has been used until now as a material for SG tubes due to its slow corrosion rate and excellent resistance to stress corrosion cracking in primary and secondary cooling water environments of pressurized water reactors(PWR). However, issues such as pitting corrosion, stress corrosion cracking (SCC), and intergranular attacks (IGA) are known to have occurred in Alloy 600 during long-term use [2,3].

Alloy 690 was developed to solve this problem. As a result of its superior SCC resistance, Alloy 690 has replaced Alloy 600 steam generator tubes [4].

The secondary system of a nuclear power plant contains few impurities (e.g., S, Pb, and Cl) introduced during the initial construction, maintenance, and normal operation. The concentrations of these impurities are very low; However these impurities can become concentrated in the gap between the tube and the top of the tubesheet because the coolant flow is restricted and a sludge is collected. This environment is chemically complicated and has a wide range of pH values from acidic to alkaline [5].

In this study, we aimed to assess the SCC behavior and characterize the oxide film after corrosion experiments by using heat transfer tube material in an aqueous solution containing Pb, Cl and S.

2. Experimental

Alloy 600 and 690 were used as SG tubing material in this work. Table 1 lists their chemical compositions. A tube was machined for reverse U-bend (RUB) sample preparation. The radius of RUB was 12.975 mm. According to a previous report [6], the stress level of ultimate tensile stress (UTS) can be applied to the apex of the RUB specimen in a RUB specimen radius range of 9-13 mm.

The test specimens were immersed for 15 weeks in 3.78L C276 autoclaves. The test matrix is shown in Table 2. The pH(T) of the solution was adjusted to be 9.5 at 310°C using NaOH.

A hydrogen concentration of 6 ppm was achieved in addition to deoxygenation for all autoclave tests [7]. An optical

TABLE 1

Chemical Compositions of Alloy 600 and 690

Material (Wt.%)	C	Si	Mn	P	Cr	Ni	Fe	Co	Ti	Cu	Al	B	S	N	Nb
Alloy 600	0.025	0.05	0.22	0.07	15.67	75.21	8.24	0.005	0.39	0.011	0.15	0.0014	0.001	0.0103	—
Alloy 690	0.02	0.22	0.32	0.009	59.57	58.9	10.54	0.01	0.26	0.01	0.019	0.004	0.001	0.017	0.01

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microscope was used to observe cracks in the specimen, and a transmission electron microscope (TEM) was used to observe the surface oxide film.

TABLE 2

Chemical Compositions of Test Solution

	Pb	Cl	S	Remark
With Pb	O	X	X	Pb
	O	O	X	Pb+Cl
	O	X	O	Pb+S
	O	O	O	Pb+Cl+S
Without Pb	X	X	X	Alkaline
	X	O	X	Cl
	X	X	O	S
	X	O	O	Cl + S

3. Results and Discussion

Optical microscope observation results are shown in Fig. 1 for the Pb + Cl + S group and Cl + S group. As shown in Fig. 1, the largest cracks were found for Alloy 690 in a solution containing Pb while no cracks were found for Alloy 690 in a solution without Pb. The shape of the cracks was complex, and most of the cracks were observed to be transgranular, similarly to other reports [8]. Unlike Alloy 690, cracks were found for Alloy 600 in all conditions. However, the cracks for Alloy 600 were significantly shorter than those for Alloy 690 in the solution containing Pb. It is presumed that the shape and existence of the surface oxide film can affect the SCC.

Fig. 2 displays the results of the experiments conducted in various environments. No cracks were observed for the Alloy 690in the solution without Pb while cracks were found for Alloy 600 regardless of the presence of Pb. The crack growth rate for Alloy 690 in the solution containing Pb was higher than that for Alloy 600. Moreover SCC rate in leaded solution was increased with chlorine indicating that chlorine shows synergistic effect on SCC rate. Chlorine showed a greater effect on SCC than sulfur in the solution containing Pb. When sulfur, chlorine, and lead were added together, the SCC rate was the highest.

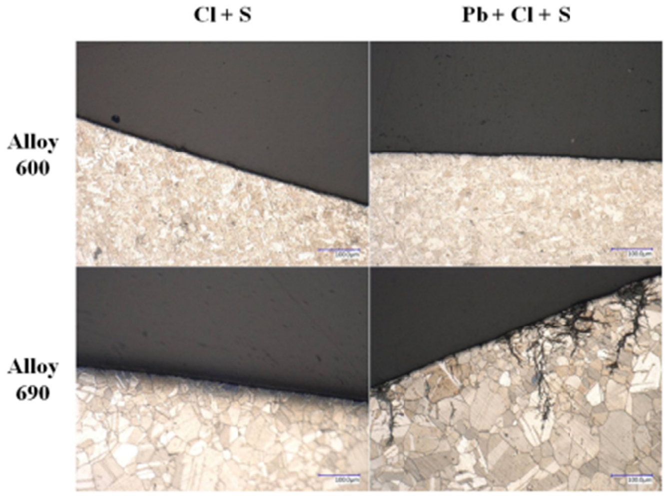

Fig. 1.Optical microscope images of Alloy 600 and 690 in Cl + S and Pb + Cl + S solutions

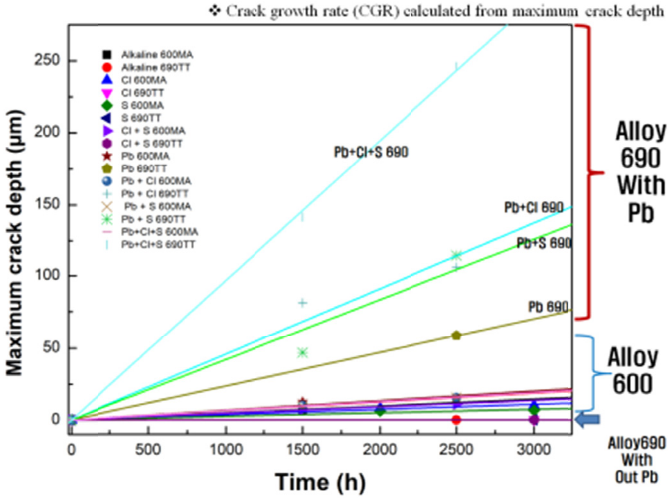

Fig. 2.Crack depth with immersion time for Alloy 600 and Alloy 690 in solutions containing different impurities

Fig. 3 shows the results of TEM for the surface oxide layer of Alloy 690. Alloy 690 had a uniform oxide film on the surface in the solution without Pb. However, the Pb oxide was detected and a uniform oxide was degraded on the surface of Alloy 690

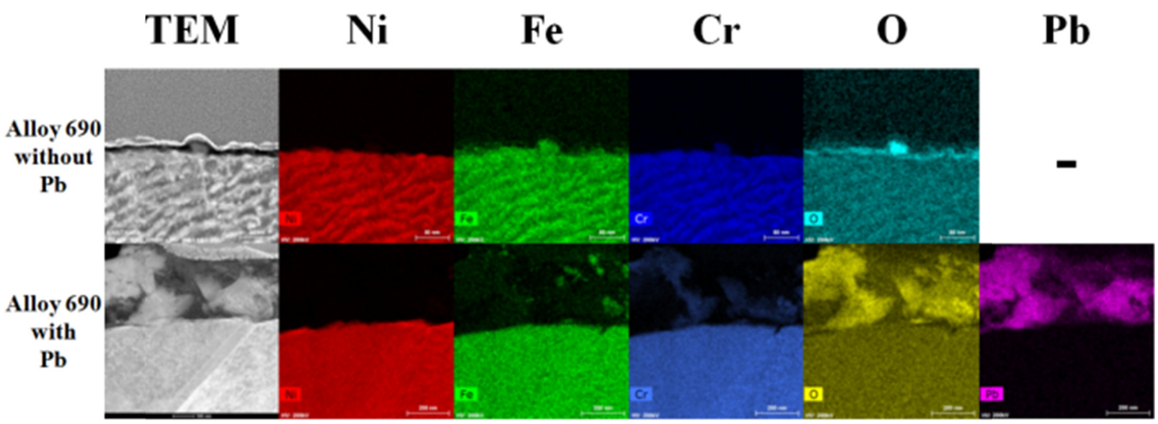

Fig. 3. STEM-HAADF images and EDX elemental maps obtained from the apex of Alloy 690 both with and without Pb

when Pb was added to solution. It is expected that the shape and existence of the uniform oxide film affected the SCC in Alloy 690 considering that surface oxide plays a significant role in SCC behavior [9].

4. Conclusion

While Alloy 690 was very resistant to SCC in test solutions without Pb, it became highly susceptible to SCC in the leaded solution of pH(T) 9.5 at 310^{o}C and was more sensitive to SCC by addition of other impurities (Cl and (or) S) in the leaded solution. Although it was possible to detect cracks for the Alloy 600 irrespective of impurities, the SCC rate for Alloy 690 was higher than that for the Alloy 600 in leaded solutions. From the observation of the surface oxide formed in the solutions containing Pb, it was found that non-uniform surface oxide film was observed for Alloy 690 in the solution containing Pb, which is suspicious of aggravation of SCC behavior.

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