

# Combined effects of lead and magnetite on the stress corrosion cracking of alloy 600 in simulated PWR secondary water

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## ABSTRACT

The combined effects of lead and magnetite on the stress corrosion cracking of Alloy 600 were investigated in simulated PWR secondary water at 315 °C. Lead induced fast stress corrosion cracking of Alloy 600, and this cracking was accelerated when the alloy was in contact with magnetite. The coupling with magnetite caused the rapid growth of the oxide layer on Alloy 600, resulting in the formation of a defective oxide layer. Based on these results, the effect of magnetite as an additional contributor to the stress corrosion cracking of Alloy 600 is discussed.

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## 1. Introduction

Mill-annealed Alloy 600 steam generator (SG) tubes have experienced various types of corrosion damage, such as stress corrosion cracking (SCC), intergranular attack, and pitting corrosion, in the secondary side of pressurized water reactors (PWRs) [1,2]. These types of corrosion mainly occur in the heated crevices between the tubes and their support plates or tube sheets, which are covered with magnetite deposits. This is because aggressive chemical impurities can be concentrated within these crevices by local boiling, resulting in the formation of a highly corrosive environment [3]. In addition, the concentration of impurities can cause the conditions to become acidic or alkaline, which is also well recognized as a contributor to the corrosion process [4–6].

The research related to the corrosion of Alloy 600 tubes has mainly focused on the water chemistry in the crevices, such as impurity concentrations, pH, and temperature [7–9]. However, it should be noted that the actual corrosion occurs on the tube surface covered with magnetite deposits under operating conditions. These deposits contain numerous open pores due to boiling chimneys that are formed on the heated tube surface [10,11]. Therefore, only the local tube surface inside the pores at the tube/magnetite

deposit interface is exposed to the corrosive crevice water. Furthermore, magnetite is an oxide, but it shows almost metallic behavior with respect to its electrical properties [12,13]. For this reason, numerous microgalvanic cells can also be formed on the tube surface within the porous magnetite deposits. This suggests that the corrosion degradation of the tubes in the secondary side of SGs will occur under these conditions where galvanic corrosion between the tube and magnetite deposit is activated. Therefore, it is necessary to evaluate the effects of not only crevice water chemistry but also the galvanic coupling with magnetite on the corrosion behavior of the tube, especially SCC that has been the major failure mechanism of Alloy 600 tubes in the secondary side of SGs.

This study aims to evaluate the combined effects of lead and magnetite on the SCC behavior of Alloy 600 in a simulated crevice environment at 315 °C. Lead oxide (PbO) was selected as an impurity that can be concentrated within the crevice, because Pb compounds have been known to induce fast SCC of Ni-based alloys [14–17]. A special U-bend specimen was prepared to evaluate the SCC of Alloy 600 occurring within porous magnetite deposits by using an electrodeposition method. Based on the experimental results, the combined effects of Pb and magnetite on the SCC of Alloy 600 are discussed in relation to the property of the oxide layer.

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## 2. Experimental procedures

### 2.1. Preparation of U-bend specimens

Alloy 600 was melted in a high frequency vacuum induction furnace, and hot-rolled in the temperature range 1150–1250 °C. The plates were cold-rolled to achieve a total thickness reduction of about 70%. The Alloy 600 specimens were machined using these cold-worked plates into a size shown on the left side of Fig. 1. These specimens were annealed at 1060 °C for 2.5 min followed by water quenching, and then were ground with 1000-grit silicon carbide paper. Finally, the specimens were ultrasonically cleaned in acetone and ethanol for 5 min, respectively. The chemical composition of Alloy 600 is given in Table 1.

The U-bend specimen was fabricated in accordance with ASTM G30-97. After the specimen was bent into the shape shown on the right side of Fig. 1, the legs were carefully fastened parallel to each other using a bolt and nut made of Alloy 600 in a bending jig without allowing the relaxation of the tensile elastic strain.

### 2.2. Preparation of magnetite-deposited U-bend specimens

To evaluate the effect of magnetite on the SCC of Alloy 600, it is necessary to continuously maintain a galvanic connection with magnetite in the test solution until the test completed. To accomplish this, a magnetite layer was electrodeposited over the entire surface of the U-bend specimen, except for a width of about 1 mm in the apex region, as shown in Fig. 2. The electrodeposition solution consisted of 2 M NaOH, 0.1 M triethanolamine, and 0.043 M  $\text{Fe}_2(\text{SO}_4)_3$ . The electrodeposition of the magnetite layer was conducted at an applied potential of  $-1.05 V_{\text{SCE}}$  for 2 h at 80 °C by using the conventional three-electrode system. The original technique for the electrodeposition of magnetite was developed by Switzer et al. [18]. In this deposition process, the composition, phase, and shape of the deposited iron oxide layer are dependent on the applied potential [19,20]. In addition, the deposition temperature also affects the shape of the electrodeposited magnetite layer [18]. Considering these factors, the deposition conditions used in this work were selected to deposit a pure and dense magnetite layer with a polyhedral shape on the surface of Alloy 600 from our previous studies [21–23]. The characteristics of the magnetite layers electrodeposited on the U-bend specimens were analyzed using a scanning electron microscope (SEM) and an X-ray diffractometer with Cu-K $\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ).

In the final product shown on the right side of Fig. 2, the undeposited Alloy 600 surface at the apex region simulates the local tube surface surrounded by the pores within the porous magnetite deposits. Therefore, the galvanic couple in this magnetite-deposited specimen will be formed consistently along the interface between Alloy 600 and magnetite during the test, simulating a porous magnetite-covered tube in an operating SG.

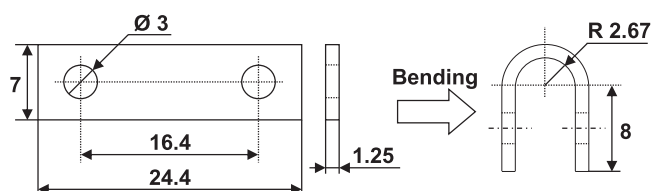


Fig. 1. Dimensions of a U-bend specimen for the SCC tests (unit: mm).

**Table 1**  
Chemical composition of Alloy 600 (wt %).

C	Cr	Fe	Si	Mn	Ti	Al	Ni
0.02	15.7	10.0	0.1	0.3	0.1	0.06	Bal.

### 2.3. SCC test

Table 2 shows the experimental details for evaluating the combined effects of Pb and magnetite on the SCC of Alloy 600. Two types of specimens were used in the SCC tests: magnetite-free and magnetite-deposited U-bend specimens. Two different solutions were also used in the SCC tests: PbO-free and 100 ppm PbO solutions. The magnetite-free specimens were located on a specimen holder made of Alloy 600 in the autoclave. In the case of the magnetite-deposited specimen, a magnetite layer was also electrodeposited on the surface of the Alloy 600 specimen holder to prevent galvanic corrosion between the specimen and holder. The SCC tests for each condition shown in Table 2 were conducted in three different Ni-autoclaves with a capacity of 3.8 L.

The base solution used for the SCC tests was deionized water adjusted to a pH of 9.0 at 25 °C using NaOH. In case of the PbO solution, PbO of 100 ppm in weight was added to the base solution. The solution pH was not additionally adjusted after the addition of PbO. Before the SCC tests, the inside of the autoclave was ground first using silicon carbide papers and carefully cleaned using acetone and deionized water. The test solution of 2 L was injected into the autoclave and deaerated with high-purity argon gas (99.999%) at a flow rate of 300 mL/min in the closed autoclave at room temperature for 3 h. The test times were measured from when the solution temperature reached 315 °C. To examine the stress corrosion cracks produced in the specimens, the autoclave operation was stopped after periods of 650 h and 1000 h. Before the autoclave operation was restarted for the 1000 h test after the inspection of the specimens, the inside of the autoclave was cleaned again, and the test solution was refreshed and deaerated using the same procedure.

After each test was completed, at least two specimens were cross-sectioned into two pieces of half the specimen width. The cross-sectioned specimens were mounted in a resin and polished with 0.5  $\mu\text{m}$  alumina powder. The mounted specimens were etched in a 2% bromine–98% methanol solution, and the maximum crack depth and crack morphology were observed using optical microscope. To analyze the properties of the oxide layer formed on Alloy 600, transmission electron microscope (TEM) specimens were prepared by a focused ion beam technique, and the layer was investigated using TEM and energy dispersive X-ray spectroscopy (EDS).

## 3. Results and discussion

### 3.1. Characteristics of the electrodeposited magnetite layer

Fig. 3 shows the X-ray diffraction (XRD) patterns and SEM images of the electrodeposited magnetite layer on the Alloy 600 specimens. As shown in Fig. 3(a), the XRD patterns of the electrodeposited magnetite layer correspond to all crystalline magnetite (JCPDS card no. 01-080-7683). The surface morphology of the electrodeposited magnetite was polyhedral and dense, as shown in Fig. 3(b). This magnetite layer was homogeneously electrodeposited over the surface of the Alloy 600 substrate. The average thickness was about 40  $\mu\text{m}$ , and no defects such as cracks and holes could be observed in the cross section of the magnetite layers

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