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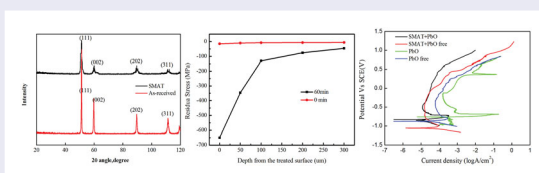
Lead-induced stress corrosion cracking behavior of mechanically surface-treated alloy 690

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ABSTRACT

Lead (Pb) is one of the impurities that cause stress corrosion cracking (SCC) of the tubing material in the steam generator (SG), the corrosion behavior of surface mechanical attrition treatment (SMAT) treated surface was measured for alloy 690 in simulated SG condition with and without Pb contamination. Even though the polarization curves indicated the corrosion potential of SMAT-processed specimens shifted negatively, the positive effect of SMAT on resistance of SCC was observed. The reduced susceptibility to lead-induced stress corrosion cracking may be attributed to the formation of stable passive film and the significant compressive stress on the surface induced by SMAT.



IMPACT STATEMENT

A significant compressive stress induced by the SMAT process effectively reduced the susceptibility of the alloys to lead-induced stress corrosion cracking.

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KEYWORDS

SMAT; PbSCC; Alloy 690; Nanocrystalline

Alloy 690, due to its outstanding robustness against corrosion resistance, is widely used as the steam generator (SG) tubing material in nuclear power plants. However, many types of corrosion still happened since it was exposed to a highly aggressive water chemistry environment containing some oxidizing impurities.[1–7] Among the impurities in the SG crevices, Pb has been recognized as one of the primary contributors to the SCC failure of SG tubing materials. It is well known that the corrosion resistance of alloy is attributed to the formation of a protective passive film on its surface. The Pb contamination results in certain changes of the passive films, and then leads to degradation of the films. The experiments of Lu et al. revealed that PbSCC might dominate rupture mechanisms in the passive film [8] and passivity degradation.[9–11] The potentiodynamic measurements of Pb contamination on the anodic dissolution behavior of nickel-based alloys indicated that the Pb could increase the anodic dissolution.[12–15] Kilian and Roth [15] and

Kim et al. [16] provided the evidence to establish the relation between PbSCC and an anodic dissolution mechanism,[11] which is the potential range of the maximum PbSCC susceptibility corresponding to the position of an anodic current peak. PbSCC behavior highly depends on the solution pH value, and recent investigation indicated that the effect of Pb contamination on the passivity degradation of alloy 690 in an alkaline solution was more significant than that observed in the acidic one.[17] Grain size refinement of the metallic surface is one of the promising ways to control the corrosion behavior of alloy 690.

Surface mechanical attrition treatment (SMAT),[18] in which the repeated multidirectional impacts at high strain rates onto the specimen surface result in severe plastic deformation and grain refinement progressively down to the nanometer regime in the entire specimen surface, is one of the important ways to significantly improve mechanical properties of metallic materials.

Compared with the conventional shot peening method, SMAT deformation directions are random, which can activate multi-slip systems. And SMAT can reach higher kinetic energies to achieve a thicker nanocrystalline surface layer. The influence of SMAT on the SCC at room temperature has been investigated and the results showed that SMAT could improve the corrosion behavior of metallic alloys.[11] However, to our knowledge, no experimental data are reported so far to investigate PbSCC of SMAT-processed materials under high temperature and high pressure. To fill the knowledge gap, it is worth to study the corrosion behavior of SMAT-processed materials under aggressive environment. In this work, the effects of SMAT on the corrosion resistance of Alloy 690 with and without Pb contamination in an alkaline solution have been investigated under high temperature and high pressure.

Alloy 690 was used as the test material and chemical composition in weight percent is 0.02C, 30Cr, 10Fe, 0.5Mn, 0.5Si and the balance is Ni. They were degreased using acetone and subjected to SMAT using 316L stainless steel balls for 60 min at a fixed frequency 20 kHz at room temperature. The details of the SMAT process were described in an earlier paper.[18] The C-ring test specimens were prepared under the ASTM G 38-37 code [19] and the applied stress was adopted as 414 MPa and the corrosion time is up to 2160 h. The C-ring SCC tests were performed in simulated alkaline SG crevice chemistries at 300°C. The corrosive environment was alkaline SG chemistries with or without 2.2 mM (500 ppm) PbO with pH at 12.7. All the SCC test solutions were deaerated by purging with high-purity nitrogen to remove dissolved oxygen in the test solution for 24 h before testing. An Ag/AgCl electrode was used as the reference electrode, and this reference electrode was immersed in 0.65 M KCl solution, which was the same as the Cl^- concentration in the testing solutions. The potential reported has been

transformed into the SCE scale. A long Pt coil was used as the counter electrode. The phase content and residual stress before and after SMAT processed were carried out using X-ray diffraction (XRD). The residual stress with removal of the surface layer by layer was determined by the $\sin^2\psi$ method. Peaks measured at higher 2θ were chosen to acquire accurate information on residual stress. The chemical composition of specimens in the oxide films was investigated using X-ray photoelectron spectroscopy (XPS); the SIMS analysis was performed to compare the concentration profiles before and after the SMAT process. The scanning electron microscope (SEM) was used to study the surface profile of SCC cracks.

XRD profiles of the as-received and SMAT-processed specimens are shown in Figure 1(a). It can be seen that the untreated specimen consists of complete austenite. Because of the reduction in crystallite size and then inducing high compressive residual stresses in near-surface regions,[20] the Bragg diffraction peaks of the specimen treated by SMAT were broadened and shift slightly to high angle. From XRD results, we also see that the surface layer of the specimen is still mainly composed of single austenite; no obvious phase transition could be observed under such an SMAT condition. Residual stress was measured on the specimen surface before and after SMAT with (311) diffraction of Cu $K\alpha$ X-ray. The electrolytic polishing was used to remove layer by layer from the specimen surface, and depth profiles of the residual stress with and without SMAT specimens are shown in Figure 1(b). Compared with as-received annealed specimens, SMAT treatment led to extremely strong compressive residual stress in the near-surface region, which remain up to a depth of 100 μm .

To investigate the effects of Pb contamination on the polarization behavior of 690 alloys in alkaline SG chemistries, the polarization curve measurements at high temperature and high pressure were conducted: the

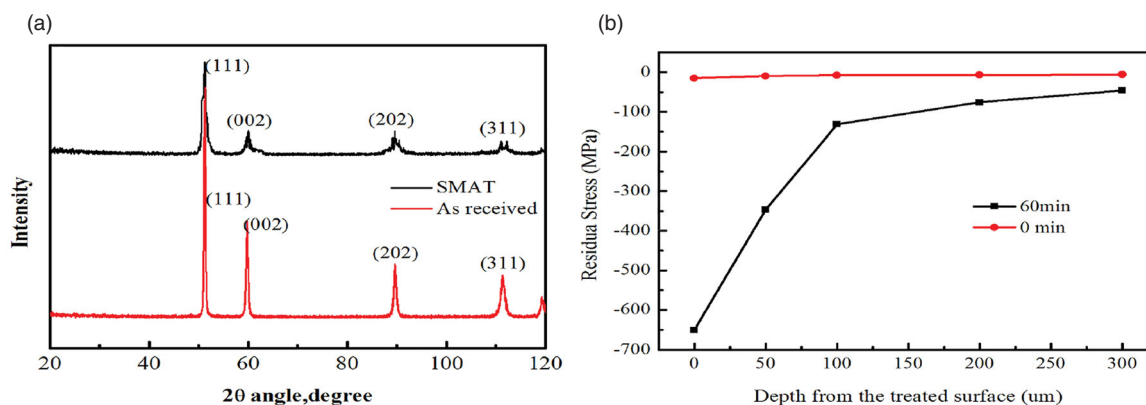


Figure 1. (Colour online) (a) XRD profiles of the untreated and SMAT-processed 690 specimens. (b) Residual stress depth profiles before and after the SMAT process.

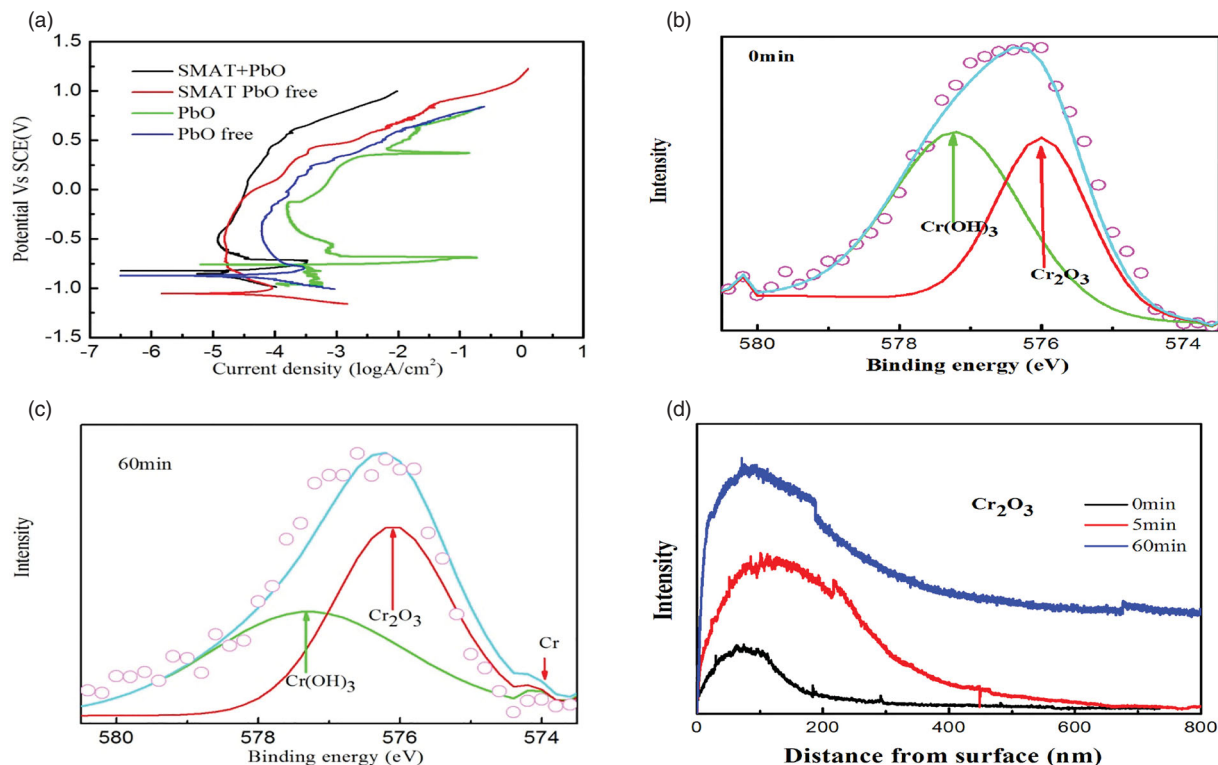


Figure 2. (Colour online) (a) Potentiodynamic curves for alloy 690 with and without SMAT treatment in PbO-free and PbO solution at 300°C; Cr 2p peaks of XPS obtained from alloy in solution containing PbO without (b) and with (c) SMAT treatment; (d) Cr₂O₃ concentration–depth profile of SMAT specimens in solution containing PbO measured by the SIMS technique. The time in (b)–(d) indicates the duration for SMAT.

specimens with and without SMAT were preconditioned at -1 V(SCE) in (1) Pb-free solution and (2) in the Pb-containing solution. Typical results are superimposed on Figure 2(a).

All specimens showed an active–passive behavior in the Pb-free and Pb-containing solution under the applied potential. It can be noted in Figure 2(a) that the corrosion potential increased for both SMAT and untreated specimens in Pb-contaminated solution compared with Pb-free solution. The corrosion potential of SMAT specimens decreased compared with the untreated ones in the same environment, while the passive current densities are obviously lower for SMAT ones regardless of the presence of Pb in the solutions. SMAT specimens have a wider passive range, which means that a more stable oxide layer was formed on the surface. According to the E–pH diagram at a high temperature,[21] Pb ions in a solution were electrodeposited during the cathodic potential scan. Hwang's study showed that the corrosion potentials were increased by the addition of Pb because metallic Pb is stable in alkaline solution at an elevated temperature.[22] The lower corrosion potential of SMAT specimens can be explained in terms of surface roughness, defect density and residual stresses induced by SMAT.[23–25] In Figure 2(a), a current peak,

named peak 1, appeared on the polarization curve of untreated specimens in Pb-contaminated solution, which is also related to the Pb electrodeposition during the cathodic potential scan. This electrodeposited Pb can be oxidized and dissolved, leading to the increase in anodic current.[26] Peak 2 is related to the oxidation of Pb, which appears at $V = 0.48$ (SCE) in the Pb-containing solution.[14] But these two peaks were not found in SMAT-treated specimens.

According to Okamoto et al.,[27,28] there are generally three types of bonds in passive films, i.e. O–M, OH–M, and H₂O–M. The distribution of oxide and hydroxide in the passive film can be identified by XPS and SIMS. Figure 2(b) and 2(c) shows Cr 2p core level spectra of XPS and their decompositions for passive film formed on alloy 690 without and with SMAT, respectively. It can be seen that the spectra were decomposed into three peaks after SMAT and corrosion: the intensity of the signal of Cr₂O₃ remarkably increased with Cr(OH)₃ decreased sharply and Cr-rich signal appeared, which indicates that SMAT leads to a significant increase in the amount of Cr oxide in the passive film. The Cr₂O₃ level spectra can also reflect the trend of OH[−]/O^{2−} distribution in the films. In the passive films, Cr oxides increased with SMAT duration and a large amount of Cr₂O₃ appeared in the film after

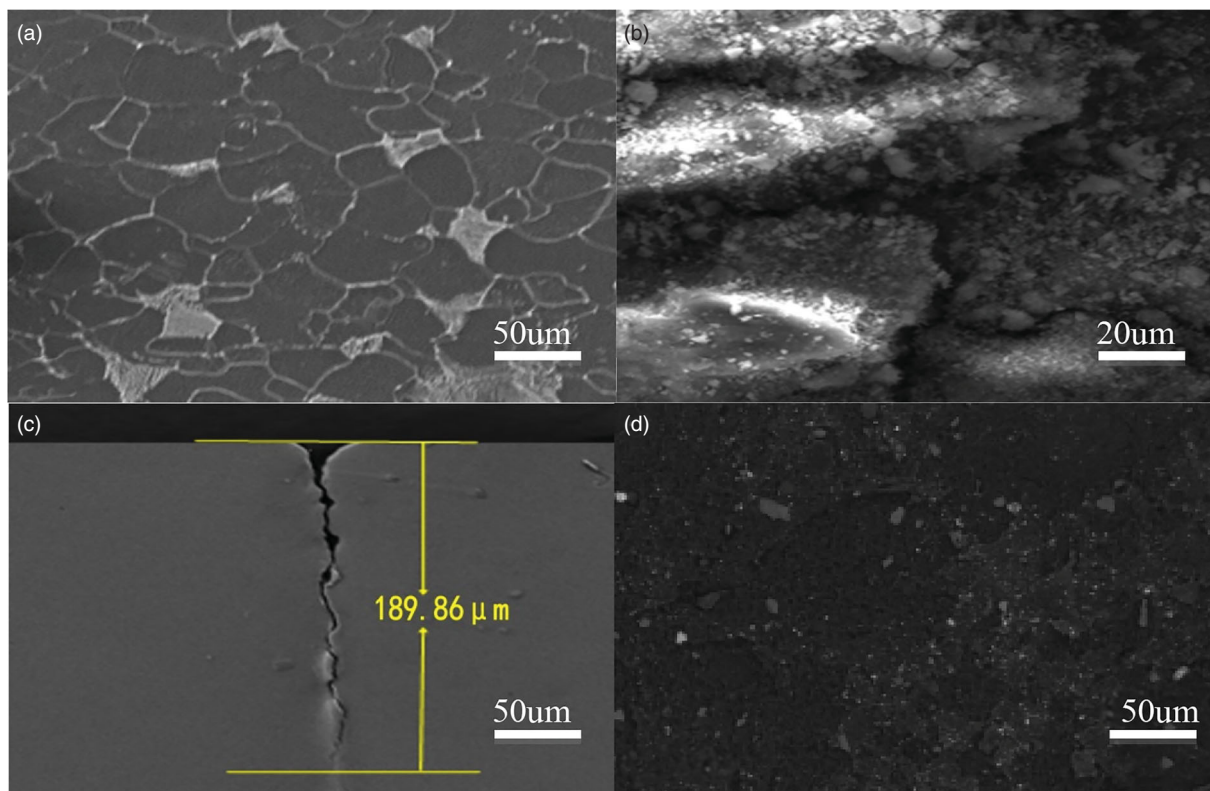


Figure 3. Surface and cross-sectional observations of C-ring with and without SMAT in solution containing PbO. (a) Surface of C-ring without SMAT before corrosion; (b) surface of C-ring without SMAT after corrosion; (c) cross-sectional observations of C-ring without SMAT after corrosion; and (d) surface of C-ring with SMAT after corrosion.

60 min SMAT, as illustrated in Figure 2(d). According to Luo et al., the presence of Pb contamination in alkaline solutions leads to a significant increase in the amount of hydroxyl in the passive film, [11,14] while it hardly affects the ratio of $\text{OH}^-/\text{O}^{2-}$ in the passive films formed with rich Cr oxide, which results in the fact that the role of Pb is not pronounced in SMAT specimens compared with the effect of Pb on the untreated ones. This agrees with the results from the polarization curve.

Figure 3 shows surface and the cross-sectional observations of C-ring specimen with and without SMAT after SCC tests in the solution containing Pb. The typical image of coarse grains was seen on the un-SMAT specimen surface before the SCC test in Figure 3(a). In Figure 3(b) SCC cracks occurred after the PbSCC test for specimen without SMAT, and a crack with a length of around 200 μm was observed on the cross-sectional image shown in Figure 3(c). In contrast, no obvious cracks were observed on SMAT-treated specimens shown in Figure 3(d).

It has been reported that the crack initiation is closely related with the property of the surface oxide film. Hwang et al. [21] reported that Pb was strongly related to crack initiation on Alloy 690 in an alkaline solution due to the acceleration of Cr dissolution from the passive oxide film. If Cr content at these locations was too small to

reach a certain critical value, SCC cracks would initiate with high stress concentration under the oxide films. Lots of new grain boundaries and a high density of dislocations were created on the surface during the SMAT process. The grain refinement on the surface affects the content and microstructure of the passivity film, as a high density of grain boundaries provide a large number of diffusion pathways for Cr, which contributes to the formation of stable Cr oxides. The formation of the protective film with rich Cr oxide on the surface of SMAT specimens resulted in the improvement of PbSCC resistance. Additionally, the high-level compressive residual stress generated during the SMAT process increased SCC resistance regardless of the presence of Pb because the compressive residual stress prevents both crack initiation and growth in SMAT specimens. Park [29] also found the similar phenomena in the shot-peened specimens.

In this work, we have investigated the effect of SMAT on SCC behavior under high-temperature and high-pressure condition in the Pb-free and Pb-containing solutions, respectively. The experimental results showed that the SMAT treatment leads to strong compressive residual stresses on the surface of specimens and a significant increase in the amount of Cr oxide in the passive film. SMAT-processed alloy 690 exhibited better

corrosion resistance in the Pb-free and Pb-containing alkaline solutions.

Disclosure statement

No potential conflict of interest was reported by the authors.

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