

Comparative Analysis of Electrochemical Noise Generated during SCC Initiation and Propagation of Nickel-based Alloys for SG Tubing

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1. Introduction

Nickel-based alloys such as Alloy 600 and 690 have been used as the steam generator (SG) tubing materials in a pressurized water reactor (PWR) due to their high corrosion resistance. However, many types of corrosion have occurred in highly caustic environments containing such impurities as Pb and Cu especially in SG sludge piles, because the highly caustic conditions can be developed in the heated crevices of PWR SG's. This work is aimed to analyze the electrochemical noise (EN) generated during Pb-assisted stress corrosion cracking (SCC) of nickel-based alloys at high temperature for a proactive management of SG tubings.

2. Experimental

The C-ring specimens were manufactured from Alloy 600 SG tubing as low temperature mill-annealed (LTMA) with an outer diameter of 22.23 mm and a thickness of 1.27 mm, and from Alloy 690 SG tubing as thermal treated (TT) with an outer diameter of 19.05 mm and a thickness of 1.09mm. The C-ring was stressed to 150% of its room temperature YS at the apex using an Alloy 600 bolt and nut according to ASTM G38.

EN measurement was carried out with a Zahner IM6e equipped with a Zahner NProbe. Two C-ring specimens were galvanically coupled using the zero-resistance ammeter (ZRA) mode of IM6e. One was stressed and the other was unstressed. The measurements were performed in a potential-controlled current noise (PCCN) mode. The electrochemical current noise (ECN) was recorded by applying anodic potential of 100 mV vs. a reference electrode (a pure Ni wire with 1 mm diameter) in a 40 wt% NaOH solution containing only 10,000 ppm PbO at 315 °C.

After an entire immersion test, the specimens were chemically etched with a solution of 2% bromine + 98% methanol, and then they were examined by a scanning electron microscopy (SEM, JEOL JSM-6360) equipped with an energy dispersive spectrometer (EDS, Oxford-7582).

3. Results and Discussion

Fig. 1 gives the time record of the ECN measured from the stressed C-ring specimen in the leaded caustic solution at 315 °C for 110 h by the EN technique in the PCCN mode. There were abrupt changes in the ECN after immersion for 132×10^3 s (points B, C, D and E in

Fig. 1), which have generally been observed during localized corrosions such as pitting corrosion, crevice corrosion, intergranular corrosion and SCC. As compared with the ECN at point A, the ECN revealed repetitive current rises followed by fast decay with certain time interval in the time record at points B and C (see the insert in Fig. 1). This means that discrete events of a localized corrosion are occurring from 132×10^3 to 158×10^3 s during the immersion in the leaded caustic solution. On the other hand, from 185×10^3 to 240×10^3 s, the ECN revealed current increases followed by slower decay with longer time interval in the time record at points D and E, indicating the occurrence of a localized corrosion event inducing larger charge passage than that at points B and C.

From the SEM analysis of the apex of the C-ring, it was found that a surface oxide film was locally broken down and also some small cracks were initiated in micron-scale along grain boundary after 42 h immersion in the solution, and one of those small cracks was propagated in an intergranular (IG) mode after 100 h immersion. With the aid of the microscopic analysis, it is easily anticipated that the current rises followed by steeper decay with shorter time interval in the time record of the ECN at points B and C in Fig. 1 are mainly due to the initiation of SCC, whereas the current increases followed by slower decay with longer time interval in the time record at points D and E in Fig. 1 are attributable to its propagation.

Figs. 2(a) and (b) present the plots of the power spectral density (PSD) vs. the frequency calculated from each time record of the ECN by a fast Fourier transform (FFT) algorithm at points A, B and C, and D, E and F, respectively. In Fig. 2(a), it is clearly seen that the PSD of the ECN obtained at points B and C where the initiation of SCC is expected to occur, is higher at whole frequency ranges than that obtained at point A where the general corrosion (the formation of the passive oxide film) occurs on the C-ring surface. Similar behaviors were found in Fig. 2(b), that is, the PSD of the ECN obtained at points D and E where the propagation of SCC is supposed to occur, is higher than that obtained at point F, except the remarkable increase of the PSD at a low frequency limit. The increases of the PSD especially at a low frequency limit in Figs. 2(a) and (b) strongly indicates the increase in the number of localized corrosion events, that is, the initiation and the propagation of Pb-assisted SCC, respectively, as previously reported[1, 2]. Since the type of corrosion cannot be reliably distinguished on the basis of the roll-

off slope [3], it will not be considered as indicator of localized corrosions in this work.

The EN technique was also applied to investigate the SCC initiation and propagation of Alloy 690 SG tubing material. Spectral analysis of the ECN and microscopic analysis will be also performed and discussed in detail.

4. Conclusion

From the microscopic and the EN analyses of nickel-based Alloy 600 and 690 SG tube materials in the leaded caustic solution environment at high temperature, it is strongly suggested that the repetitive current rises followed by steeper decay with shorter time interval in the time record of the ECN measured in the PCCN mode are mainly due to the initiation of SCC, whereas the current increases followed by slower decay with longer time interval are attributable to its propagation. From the spectral analysis of the ECN, the PSD increased more remarkably at low frequency limit for the propagation of SCC as compared to that for the initiation of SCC.

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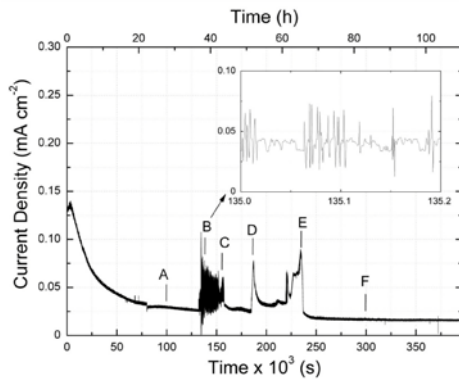


Fig. 1. Time records of ECN measured in PCCN mode from the stressed C-ring specimens in the solution containing PbO.

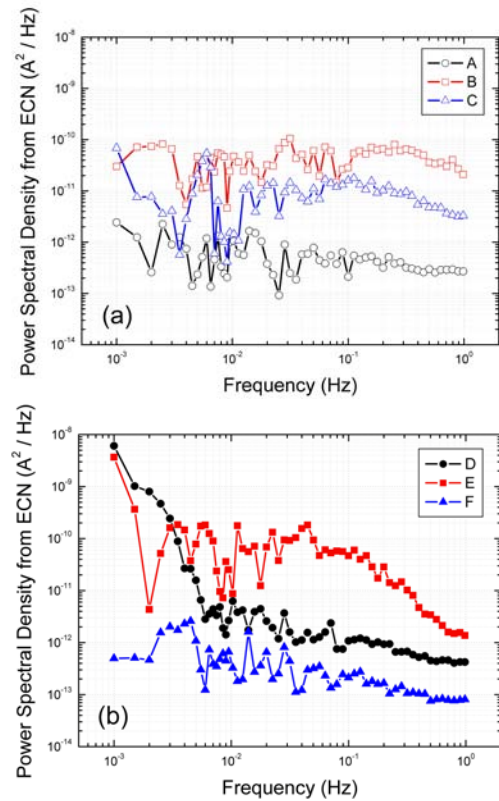


Fig. 2. Plots of the PSD vs. the frequency calculated from each time record of the ECN by the FFT algorithm at points (a) A, B and C, and (b) D, E and F.