Development the Technique for Laboratory-Degraded Alloy 600 Steam Generator tubing and Electrochemical Impedance Spectroscopy of Oxide Film on at PWR environment

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1. Introduction

For the integrity management of SG tubes, non-destructive-evaluation performed using eddy-current-test (ECT) is necessary in the assessment. The reliability of ECT evaluation is dependent on the accuracy of ECT for various kinds of defects. For basic calibration and qualification of these techniques, cracked SG tube specimens having mechanical and micro-structural characteristics of intergranular cracks in the field, are needed. To produce libraries of Laboratory-Degraded-SG-tubes (LDT) with intergranular cracks, a radial denting method and direct tension method were explored for generating ID and OD axial cracks and circumferential crack by three-dimensional finite-element-analysis and experimental demonstration, respectively. The technique is proven to be applicable for generating axial cracks with long and shallow geometry as opposed to the semi-circular cracks typically obtained by the internal-pressurization method. In addition, a DCPD method with array probes was developed for accurate monitoring and controlling of crack size and shape. By these methods, long and shallow intergranular axial and circumferential cracks more typical of actual degraded SG tubes were successfully produced.

As a final step, the developed Laboratory-Degraded Alloy 600 Steam Generator tubing specimens were exposed to typical PWR water with 1,200 ppm boron and 2 ppm lithium at a pressure of 15 MPa and temperatures of 320 degree C. Dissolved hydrogen concentration was varied from 0 to 30 cm^3 (STP)/kg to study its effect on the surface oxide film composition of alloy 600 in PWR water environment. As an optional experiment, Alloy 600 SG tubing specimens were exposed to PWR secondary water chemistry environment to understanding the properties of oxide films formed on alloy surface in PWR secondary environment.

This study is focused on the on-line monitoring method on crack generation using DCPD method and the properties of oxide films formed on Alloy 600 in high temperature aqueous environment by performing a series of electrochemical measurements, including electrochemical corrosion potential (ECP) monitoring, electrochemical impedance spectroscopy (EIS).

The properties and depth profiles of Oxide film estimated by EIS will be verified by an analysis using XPS and FE-SEM ultimately.

The objective of this research is to develop a fabrication technology of Laboratory-Degraded SG Tubes which has same geometry and corrosion defect with field-serviced SG tubes, and a technology of precise diagnosis using Laboratory-Degraded SG Tubes.

2. Experiments

On-line monitoring method using DCPD

In this study, DCPD simulation using FEA was performed for improving the reliability of the DCPD method in monitoring the depth and length of cracks. By analyzing changes in the potential drop data of the DCPD FEA simulations of SG tube specimens with a crack or without one, the DCPD method with array probes was expected to demonstrate improvement of crack shape monitoring using the direct current potential drop method. Through the comparison with actual experimental DCPD data, the FEA study results provide basic reference data to verify the measurements for production of cracked SG tube specimen.

Fig.1 shows dcpd signal analysis of 25.4mm ID axial crack. The numbers on this figure represent locations measured DCPD signal. In the lower figure of fig.1, blue rectangular point is FEA simulation results at each crack depth. And red diamond point is DCPD measured signal. At same crack depth, two data has a difference of about 8 V. But as a crack depth is deeper, DCPD signal is higher with similar difference. From these results, we can confirm reliability and adequate application of DCPD. Also, from this comparative study of DCPD method, depth distribution of unknown crack can be assumed, as shown in upper figure of Fig.1.

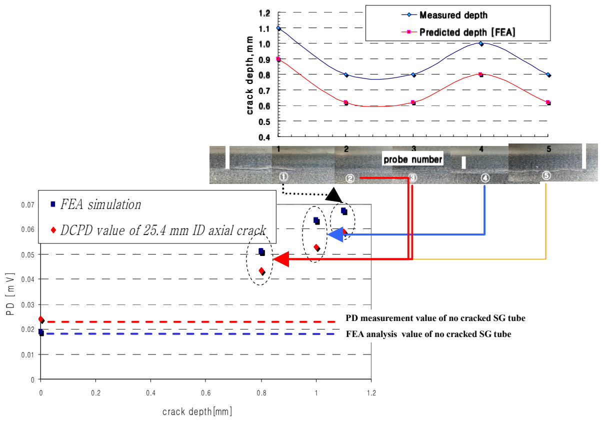

Fig.1 Comparison of measured DCPD signal and predicted DCPD data using FEA of 25.4mm ID axial crack

Electrochemical Impedance Spectroscopy of Oxide Film on at PWR environment

To make more same geometry and corrosion defect with field-serviced SG tubes, as a final step, the developed Laboratory-Degraded Alloy 600 Steam Generator tubing specimens were exposed to typical PWR water with 1,200 ppm boron and 2 ppm lithium at a pressure of 15 MPa and temperatures of 320 degree C.

All experiments were carried out at HTHP condition under continuous dearation with high purity nitrogen in a PWR primary water chemistry, which is composed of 1200 ppm boron and 2 ppm lithium.

The impedance scan frequencies ranged from 10^6 to 10^{-2} and its spectra were collected using system consisting of a Solatron Model 1260 FRA, Solatron 1286 potentiostat, and Zplot software. Impedance spectra were analyzed by Zview. The results of EIS experiment and equilibrium circuit analysis was summarized in Fig.2 and 3.

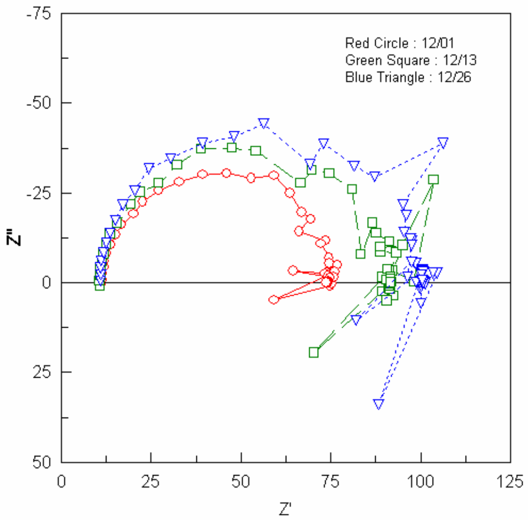

Fig.2 Complex Nonlinear Least Square (CNLS) fitting for high frequency data

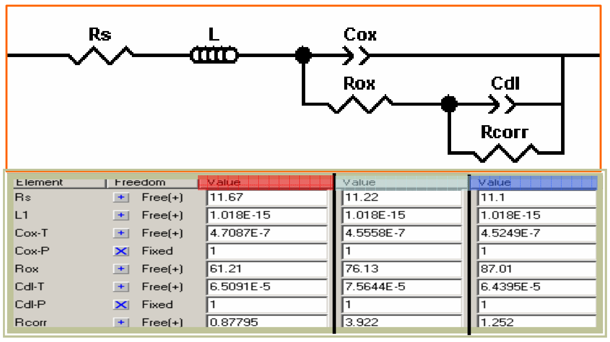

Fig. 3 Equivalent circuit and CNLS fitting result for high frequency data

From these analysis results and eq.1, the oxide film thickness of alloy 600 SG tubing can be assumed as shown in table.1.

$$C = \varepsilon \cdot \varepsilon_0 \cdot \frac{A}{d} \qquad \text{(eq.1)}$$

Where, $C =$ Capacitance [farad]

$A =$ Area [m^2]

$d =$ Oxide thickness [m]

$\varepsilon =$ Dielectric constant

$\varepsilon_0 = 8.854 \times 10^{-12}\ F/m$

Tab. 1 calculated oxide thickness of Alloy 600 SG tubing

	12/01	12/13	12/26
Oxide capacitance	0.47087μF	0.45558μF	0.45249μF
Calculated thickness	0.18803 μm	0.19435 μm	0.19567 μm

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