Development of New Loop System for Simulating Sludge Deposited on the Steam Generator Tubes and Comparative Evaluation with Real Sludge Deposits

Soon-Hyeok Jeon*, Hee-Sang Shim, Ji-Min Lee, Do Haeng Hur

Safety Materials Technology Development Division, Korea Atomic Energy Research Institute *Corresponding author: junsoon@kaeri.re.kr

1. Introduction

Magnetite originated from the secondary coolant systems that feed water, condensate, and the drain line is accumulated on the surfaces of steam generator (SG) tubes, the top of tube sheets in secondary coolant systems in the pressurized water reactors (PWRs) of nuclear power plants [1,2].

SG tube sludge deposits could decrease the heat exchange capability of an SG either by decreasing the heat transfer efficiency or by interfering with the SG hydrodynamics. In addition, aggressive chemical impurities, such as chloride and sulfate ions, and lead, can concentrate in porous sludge deposits [3–5] and thus accelerate the corrosion of SG tubes, such as intergranular attack, pitting and stress corrosion cracking (SCC) [5].

The occurrence probability for these problems is greatly affected by the chemical and physical properties and quantity of the SG tube sludge. To investigate the sludge deposition behavior of SG tube, a proper method to deposit sludge on the surface of SG tube should be established. However, there has been no recirculation loop system for sludge deposition on the SG tube in the secondary water of PWRs. Hence, we newly developed the recirculation loop system for simulating sludge deposited on the Alloy 690TT steam generator tubes. This paper introduces a new loop system to simulate the porous magnetite sludge. This development of loop system for sludge deposition is a new approach to simulate the porous magnetite deposited on the SG tubes in nuclear power plants.

After the sludge deposition tests, the simulated sludge deposits were analyzed by using a focused ion beam (FIB)-scanning electron microscope (SEM) to observe closely the cross-section of deposits. The particle morphology, chemical composition, and layer thickness of the deposits were analyzed using FIB-SEM attached with an energy-dispersive X-ray spectrometer (EDS). In addition, the flake samples collected from an SG of a real nuclear power plant during sludge lancing were also analyzed for comparison to the sludge deposits simulated by using new recirculation loop system.

2. Methods

2.1 Specimen Production and Test Solution

Alloy 690TT SG tube was chosen as the test tube. The dimensions of the test tubes were outer diameter (OD) of 19.05 mm, inner diameter (ID) of 17.00 mm and length of 500 mm.

For comparison with the simulated magnetite accumulated on the Alloy 690TT tube by using new loop system, flake samples were collected from the SG tubes of a real nuclear power plant during sludge lancing.

The secondary water solution of pH 9.0 at 25 °C was used in this study. The pH of the test solution was adjusted using ETA, which is an organic chemical agent widely used to control the pH of secondary water in PWRs. The precursors of Fe ions for sludge deposition were prepared using Fe-acetate. The test solution containing 260 ppm Fe by weight was stored in an injection tank for injecting into the test section.

2.2 Secondary loop system

Sludge deposition tests were performed using a circulating loop, which is shown schematically in Fig. 1. The circulating loop system consisted of the following main components: a secondary water solution tank, Fe ion source tank, high pressure pump (HP pump), preheater for solution inlet temperature control, metering pumps for Fe ion injection and back pressure regulator (BPR) for pressure control, heat exchanger, a test section equipped with a SG tube.



Fig. 1. Schematic diagram of secondary loop system for the sludge deposition test.

The simulated secondary water was stored in the solution tank with a capacity of 100 L and recirculated using the HP pump, pre-heater, test section, and heat exchanger. Dissolved oxygen was controlled at <5 ppb. The flow rate adjacent to the SG tube in the test section was continuously controlled at 260 ml/min. After these conditions were set, we injected the Fe ions into the test section through the metering injection pump with a flow

rate of 1 ml/min from the injection tank. The precursor solution was diluted in the simulated secondary water stream and its final concentration was calculated as 1 ppm Fe in the test section. Each deposition test was performed for 14 days.

2.3 Analysis for sludge deposits

Fig. 2 shows the SG tube specimen before and after the deposition test. After the test was finished, the specimen was dried at 60 $^{\circ}$ C for about 3 hours and then cut with a low speed cutter for analysis.

The simulated sludge deposits and flake samples were analyzed by using a FIB-SEM to observe closely the cross-section of deposits. The particle morphology, chemical composition, and layer thickness of the deposits were analyzed using FIB-SEM attached with an EDS.



Fig. 2. Commercial SG tube (Alloy 690TT) used in this study (a) before and (b) after the deposition test.

3. Results and Discussion

3.1 Particle morphology of simulated sludge deposits and flake samples

Fig. 3 shows SEM images of the particle morphology of simulated sludge deposits by using new loop system and flake samples collected from the real operating nuclear power plant. For two samples, relatively round particles of sizes in the range of 30-800 nm with numerous small pores were observed. The round morphologies and particle size of both samples were almost same.



Fig. 3. SEM micrographs of the surface of sludge deposits at pH 9.0 and flake samples collected from an operating nuclear power plant: (a) simulated deposits and (b) flake samples.

Fig. 4 shows the SEM micrograph of the crosssection of the simulated sludge deposits at pH 9.0 and flake samples after ion-milling by using a FIB. The thickness of the simulated sludge deposits was approximately 15 µm. The flake sample showed that the thickness was about 110 ~ 120 µm. A large number of the micro-pores were observed throughout the simulated deposits and SG flake samples. The number of micropores and pore size increased from the tube side to the water side. These results may be closely related to the boiling behavior such as bubble growth and boiling chimney. When comparing the cross sections of two specimens, the thickness of simulated deposits was much thinner than that of the flake sample. This is because the test time is too small even the deposition test is performed under high concentration of Fe ion source. Thickness of the simulated deposit could be made thicker by increasing the test time.





Fig. 4. SEM micrographs of the cross section of sludge deposits at pH 9.0 value and flake samples: (a) simulated deposit and (b) flake sample

3.2 Chemical composition of simulated sludge deposits and flake samples

The simulated deposits were analyzed by SEM-EDS to identify their chemical composition. Point-EDS analyses were conducted, corresponding to this shown in Fig. 5 (a). The results of point EDS analysis are shown in Fig. 5 (b). Based on the result, the sludge deposits were only consisted of magnetite.



Fig. 5. SEM-EDS analysis of the particle on the surface of SG tube deposits: (a) SEM image and (b) the result of point -EDS.

Fig. 6 shows the EBSD data on the tube side of the SG flakes. As shown in Fig. 6 (a), the analyzed region is containing of the number of the micro-pores. In the 001 inverse pole figure (IPF) orientation map (Fig 6 (b)), a random orientation was observed (predominant orientation color did not appears). Fig. 6 (c) shows the

phase distribution map. Four phases such as magnetite (Fe_3O_4) , trevorite (Ni_2FeO_4) , jacobsite (Mn_2FeO_4) , and Cu were observed and randomly distributed in all the analyzed regions. The detailed phase fractions of the flakes are presented in Table I. These results indicated that the flake samples were mainly composed of magnetite and contained only small amounts of trevorite, jacobsite, and metallic Cu particle.

The magnetite deposit that was produced in this work could not perfectly simulate the flake samples, which included not only magnetite (about 90%) but also various compounds, such as jacobsite (about 5.4%) and trevorite (about 4.4%), metallic copper (about 1%). However, because the flake samples consisted of about 90% magnetite and has a porous structure, the simulated magnetite deposit is sufficiently similar to simulate the flakes on the SG tubes of actual PWRs.



Fig. 6. EBSD data on the tube side of the SG flakes: (a) SEM micrograph, (b) 001 IPF orientation map, and (c) phase distribution map.

Structure	Phase fraction (%)
Magnetite	89.22
Jacobsite	5.36
Trevorite	4.40
Cu particles	1.02

Table I: Phase fraction of the tube side of the SG flakes using EBSD analysis (wt.%)

In the future plan, the deposition tests will be performed to determine the optimum water chemistry conditions like pH values or pH agents for reducing the amount of sludge deposition in secondary water system.

Furthermore, porous magnetite deposits produced by the new loop system can be used for studying the variation of heat transfer according to the thickness of magnetite accumulated on SG tubes (Fig 7 (a)). It can also be used to study the corrosion rate of SG tubes in secondary water containing aggressive ionic species (Fig. 7 (b)).



Fig. 7. Applications to the researches about the integrity of SG by using simulated sludge on SG tubes: (a) heat transfer deterioration and (b) corrosion acceleration owing to the concentration of chemical impurities.

4. Conclusions

The objective of this study was to introduce the development of new loop system for simulating the porous magnetite deposits on SG tubes in PWRs. The simulated magnetite deposits are appropriate to simulate real SG tube flakes because the microstructure and porous morphology are almost same to those of SG flakes. Using the simulated SG tube sludge, various aspects of nuclear research, such as heat transfer characteristics and the corrosion acceleration of porous magnetite deposited on SG tubes could be studied.

Acknowledgement

This work was supported by the National Research Foundation (NRF) grant of the Republic of Korea funded by the Korean government (NRF-2017M2A8A4015159).

REFERENCES

[1] C. Ramesh, N. Murugesan, A.A.M. Prince, S. Velmurugan, S.V. Narasimhan, V. Ganesan, Application of polymer electrolyte based hydrogen sensor to study corrosion of carbon steel in acid medium, Corros. Sci. Vol. 43, p. 1865, 2001.

[2] A.A.M. Prince, S. Velmurugan, S.V. Narasimhan, C. Ramesh, N. Murugesan, R.S. Raghavan, R.J. Gopalan, Dissolution behavior of magnetite film formed over carbon steel in dilute organic acid media, J. Nucl. Mater. Vol. 289, 281, 2001.

[3] J.P.N. Paine, S.A. Hobart, S.G. Sawochka, Predicting steam generator crevice chemistry. In Proceedings of the 5th International Symposium on Environmental Degradation of Materials in Nuclear Power System-Water Reactors, Monterey, CA, USA, 25-29 August, p. 739, 1991.

[4] P.J. Millet, J.M. Fenton, A detailed model of localized concentration processes in porous deposits of SGs. In Proceedings of the 5th International Symposium on Environmental Degradation of Materials in Nuclear Power System-Water Reactors, Monterey, CA, USA, 25-29 August, p. 745, 1991.

[5] T. Sakai, T. Senjuh, K. Aoki, T. Shigemitsu, Y. Kishi, Lead induced stress corrosion cracking of Alloy 600 and 690 in high temperature water. In Proceedings of the 5th International Symposium on Environmental Degradation of Materials in Nuclear Power System-Water Reactors, Monterey, CA, USA, 25-29 August, p. 764, 1991.