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Chemical Decontamination for Self-Disposal of Metal Wastes

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1. Introduction

The demand for large scale metal waste treatment is increasing due to the replacement of steam generators (SGs) and reactor heads (RHs) according to an increase in operational age of nuclear power plants (NPPs) in Korea. Two SGs of Kori-1 NPP were already replaced in 1998. Recently six SGs of Uljin-1&2 NPPs were replaced over 2011 and 2012. It is expected that the replacement of SGs and RHs from other NPPS in Korea increases continuously[1]. Therefore, there are needs in metal waste volume reduction and self-disposal technology for enhancing the safety and economy in the management of large scale metal wastes such as retired SGs and RHs.

In order to remove radionuclide contamination from the surface of materials, the variety of decontamination technologies have been developed in many countries. Chemical decontamination processes were originally designed to remove the radioactive deposit in order to reduce the radiation exposure to workers while operating the nuclear facilities. Diluted chemical solutions have been used to avoid damaging the system materials. On the other hand, in cases of the decontamination processes for decommissioning or recycling large components, it is greatly important to remove radioactive contamination completely by using a more aggressive solution[2].

In this study, the effectiveness of metal surface decontamination using inorganic acid solutions such as HF/HNO_3 and HF/H_2SO_4 was investigated to evaluate the applicability of these processes to the decontamination of retired SGs.

2. Methods and Results

2.1 Materials and Methods

The 304 stainless steel (STS 304) and Inconel 600 specimen of 20mm×20mm×2mm were used as the surrogate metal wastes. The oxide covered specimens were prepared in an EDTA-hydrazine mixture solution (0.05M-0.05ppm) in an autoclave at pressures ranging from 48~53 kg/cm^2 and at 270℃ for 72 hours.

The decontamination solution was made up of hydrofluoric acid (HF, Duksan Co.), nitric acid (HNO_3, Duksan Co.), sodium fluoride (NaF, Sigma-Aldrich Co.) and sodium nitrate ($NaNO_3$, Sigma-Aldrich Co.). The concentrations of the solutions were 0.5M (HF and NaF) and 0.5M (HNO_3 and $NaNO_3$).

Decontamination tests were carried out in the bottle placed in a thermostatic water bath. The temperatures of 100mL inorganic acid mixture solutions in the bottle were varied in the range of 25 and 90℃ and the stirrer speed was 120 rpm. The range of dissolution time was from 0.5 to 4.0 hours. The weight loss of the specimen before and after the dissolution tests were measured for evaluating the decontamination efficiency.

The morphology of the oxide film on the metal specimens was examined by a SEM (JSM-6300, JEOL LTD Co.). The element composition of the specimen surface was analyzed by an XRD (Rigaku Co.).

Decontamination tests were also performed using STS 304 specimen contaminated artificially with a Co-60 isotope and SG heat transfer tube specimen of Inconel material taken from the nuclear facilities.

2.3 Test results

Fig. 1 shows the dissolution behavior of STS 304 and Inconel 600 specimen in HF/HNO_3 mixture solution at 90℃. A big difference in the dissolution rate between STS 304 and Inconel 600 specimen can be shown. This result evokes concern about an excessive dissolution of Inconel 600 tubes during decontamination of SG resulting large amount of secondary waste.

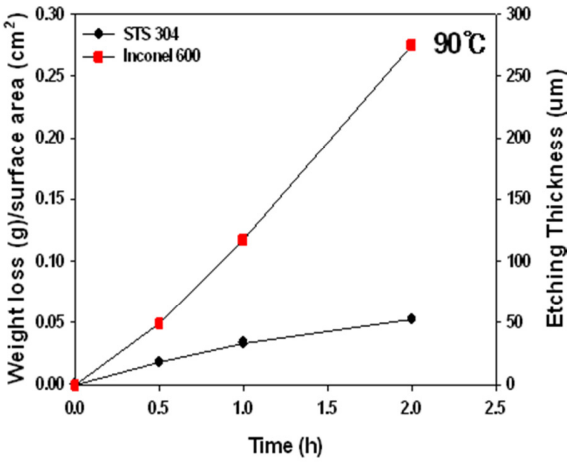

Fig. 1. Dissolution of STS 304 and Inconel 600 specimen in HF/HNO_3 process at 90℃.

Fig. 2 shows the dissolution behavior of STS 304 and Inconel 600 specimen in $HF/NaNO_3$ mixture solution at 90℃. The dissolution rate of Inconel 600 specimen could be controlled by substitution of sodium nitrate for nitric acid. As a result, the dissolution rate of STS 304 and Inconel 600 specimen was very similar.

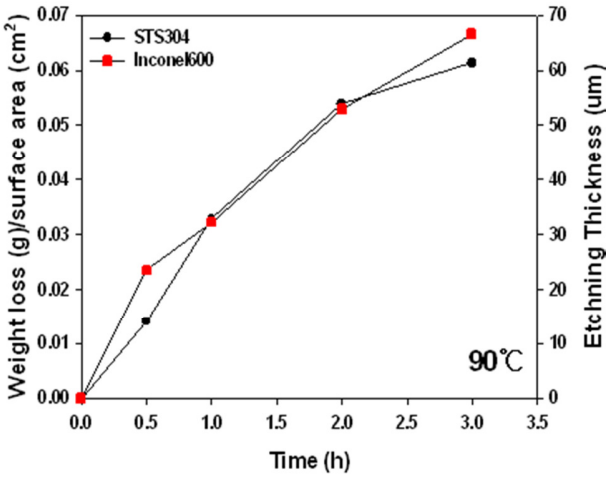

Fig. 2. Dissolution of STS 304 and Inconel 600 specimen in $HF/NaNO_3$ process at 90℃.

Fig. 3 shows the dissolution behavior of oxide covered STS 304 specimen in $HF/NaNO_3$ and NaF/HNO_3 inorganic acid mixture solutions at 90℃. The dissolution behavior of the specimen was similar in the two processes. The oxides come off the surface by pulverization process in which the inorganic acid mixture solution penetrates the oxide layer to attack the base metal[3].

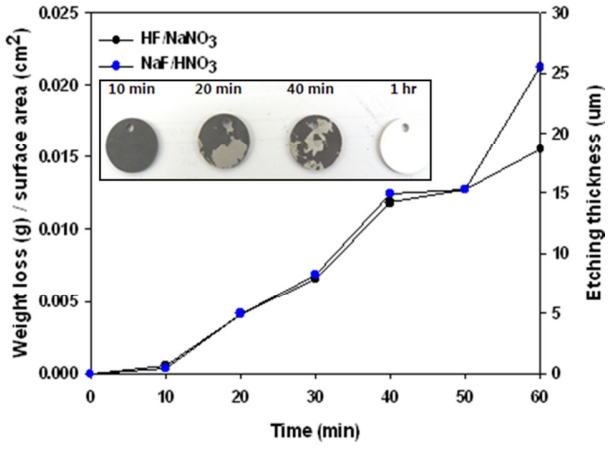

Fig. 3. Dissolution of oxide covered STS 304 specimen in $HF/NaNO_3$ and NaF/HNO_3 processes at 90℃.

Fig. 4 shows the results for decontamination of SG heat transfer tube specimen taken from Younggwang NPP. Prior to the chemical decontamination in $HF/NaNO_3$ and NaF/HNO_3 mixture solution, dry abrasive blasting as decontamination for pre-treatment was carried out. The decontamination factors (DFs) of the SG tube specimen after decontamination for 2 hours were 95 in $HF/NaNO_3$ and 65 in NaF/HNO_3 process.

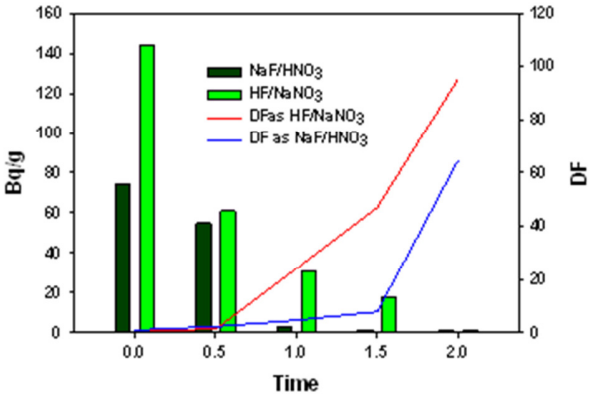

Fig. 4. Decontamination behavior of SG heat transfer tube specimen in $HF/NaNO_3$ and NaF/HNO_3 processes.

3. Conclusion

The chemical decontamination process using inorganic acid containing fluoride was investigated as a metal surface decontamination process for decommissioning and self-disposal of metal wastes. The $HF/NaNO_3$ decontamination process that improves the existing HF/HNO_3 decontamination process has been suggested as a decontamination process for retired SGs consisting of different types of system materials such as stainless steel and Inconel.

Acknowledgement

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