Decontamination Process Proposal for Used Steam Generator Tubes by Using Molten Salt Technologies

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1. Introduction

In Korea, a lot of used steam generators were produced due to replacement and permanent shutdown of nuclear power plant. Within 10 years, additional 10 nuclear power plants will be expired and many of steam generators will be replaced by new steam generators because of their high ratio of plugged tubes. Used steam generators are stored in nuclear power plants site and wait for decontamination of them. Among the decontamination of steam generators, the most serious problem is decontamination of heat tubes cause of its complex shape.

Cause of CRUD and uranium, fission products which were generated from damaged fuel, primary coolant contains radioactive nuclides although secondary coolant do not contain any radioactive nuclides. At the primary surface of heat tubes in steam generator, radioactive CRUDs are deposited by rapid temperature decrease of coolant. Thus, most of radioactivity in heat tubes are deposited at primary surface and secondary surface is relatively free from radioactivity.

At the primary surface, a lot of micro cracks exists which were generated by stress corrosion cracking. Floating radioactive CRUDs are deposited in micro cracks and these hidden radioactive CRUDs in micro cracks disturb conventional chemical and physical decontamination processes. Studsvik cooperation, which do decontamination in the main business disposals heat tubes in forms of low level waste after melting although other parts such as outer sheet, steam drum and chamber are recycled after decontamination process.

Heat tubes are made by nickel-chromium-iron based alloy and according to radioactivity stock in steam generator data of Kori unit 1, Hanbit unit 3, 4 and Ontario Power Generation(OPG), radioactivity of main elements of heat tubes (Ni, Cr, Fe) are lower than clearance level. Thus, if decontamination process for heat tubes is developed, economic advantages in decrease of disposal costs and direct recycle of metals will be secured.

In this abstract, electrochemical decontamination process for heat tubes by using molten salt technology is proposed. In this process, 4 stages are suggested; Manufacture of simulated heat tube specimen, Ni-based alloy extraction, Molten salt purifying process, and Nuclide concentration monitoring process. Among these 4 stages, first and second stages are analyzed in this paper and experimental plans are shown.

2. Experimental and results

In this section, experimental setups of each stage of decontamination process are described.

2.1. Manufacture of simulated heat tube specimen

According to previous research, radioactive nuclides in heat tubes are divided with major radioactive nuclides whose radioactivity exceed 10 times than clearance level and minor radioactive nuclides whose radioactivity do not exceed 10 times than clearance level, but exceed clearance level after 20 years cooldown. Major radioactive nuclides in heat tubes are Co-60, Cs-134, Cs-137 and actinides and minor radioactive nuclides are C-14, Eu-154. In heat tubes, micro cracks whose width and depth are less than 10 μm and 100 μm.

To check the corrosion behavior of base material, 1x3x5 mm size of Inconel-600 plate was inserted in 10 g of LiCl-KCl-5 wt% $NiCl_2$ electrolyte. Temperature was 500 ℃ and 48 hours after inserted, specimen was ejected. However corroded depth was less than 10 μm.

To make micro cracks CTOD(Crack Tip Opening Displacement) testing was adopted. With ASTM E1290-08, micro cracks whose geometrical shape is similar with natural cracks can be obtained. After generating micro cracks, Co deposition is proceeded by periodic pulse reverse plating technique in 0.5 M $CoSo_4$ water solution at 70 ℃.

2.2. Ni-based alloy extraction

Electrochemical behaviors of main element of Inconel-600(Ni, Cr, Fe) and major radioactive element(Co) were analyzed by cyclic voltammetry. As shown in Figure 1, Ni is the most reductive element among the whole element. Co is reductive than Cr and Fe. Calculated diffusion coefficients of these elements were Ni(1.415 x 10^{-5} cm^2/s), Cr(7.498 x 10^{-6} cm^2/s), Fe(1.370 x 10^{-5} cm^2/s), Co(1.306 x 10^{-5} cm^2/s) and these are similar values with previous study.

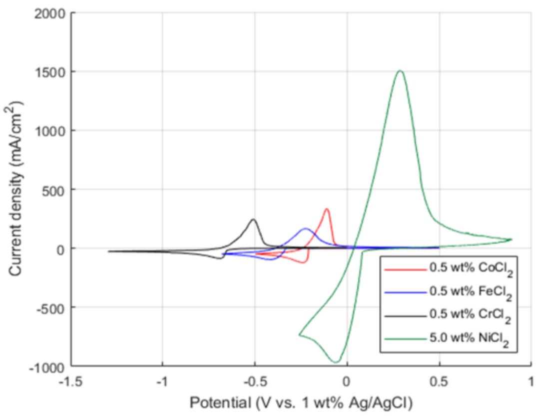

Figure 1. cyclic voltammograms of LiCl-KCl-$NiCl_2$, $CrCl_2$, $FeCl_2$, $CoCl_2$. Temperature: 500 ℃, v: 300 mV/s

In this part, 1 mm thick Inconel-600 plate was used for anode electrode and 2 tungsten rods whose diameter is 3.125 mm was used for counter electrode and LiCl-KCl-5 wt% $NiCl_2$ molten salts was used for electrolyte. Temperature was 500℃. By differing adopted potential of anode, extraction and deposition of Ni in anode were proceeded and Ni deposition at counter electrode was observed. In this experiment, applied potential was -0.20 V, 0.05 V, 0.15 V, 0.20 V, 0.30 V.

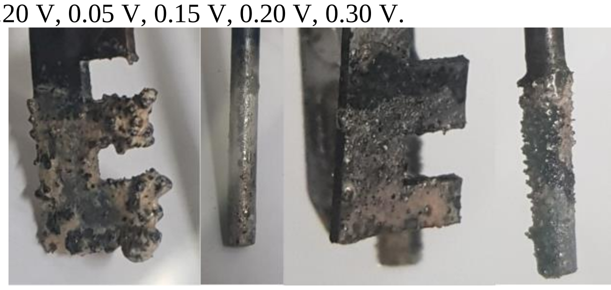

Figure 2. Ejected anode(left) and cathode(right) after experiment. Applied potential: 0.20 V, exchanged charge: 700 C, temperature: 500 ℃

In cases of applied potential is less than 0.10 V, potential of cathode increases and saturates at Cl^-/Cl_2 reaction potential. If the potential is higher than 0.30 V, recovered Ni was obtained by forming dendrite crystal. When the applied potential was 0.20 V, purity of recovered Ni at cathode was 99.7%.

3. Discussion and Future work

In this section, discussions of each experimental results are analyzed.

3.1. Manufacture of simulated heat tube specimen

Since Cs-134 and Cs-137 are major radioactive nuclides, using CsCl and CsF was impossible although they are the most oxidative chloride and fluoride. Thus LiCl-KCl eutectic salts were adopted because they have lower melting point than NaCl-KCl eutectic salts.

Inconel-600 shows very slow corrosion behaviors in environment of LiCl-KCl-5 wt% $NiCl_2$ at 500 ℃. Thus chemical reaction does not guarantee the crack widening or surface-crack polishing effect. In other words, electrochemical reaction is essential to decontaminate heat tubes.

In this experiment, CTOD testing was used to make artificial cracks and periodic pulse reverse plating technique was used to deposit Co. Specimen is similar with used steam generator heat tubes, but Cs and actinides deposition was not included. Thus 2 options can be chosen; first option is intermetallic compound formation and second option is sputtering technique.

Experiments which uses actinides is hard to execute, Mg which has similar redox potential with Pu and Am will be used as surrogate element. By using arc plasma, Cs-Mg-Inconel 600 compounds can be made and insert this specimen in LiCl-KCl-5 wt% $NiCl_2$ molten salts. If the Cs and Mg of the surface region of the alloy extracted into the electrolyte, Cs and actinides also will oxidize and dissolve into electrolyte. If this chemical reaction shows opposite result, then sputtering technique which uses target metal will be adopted.

3.2. Ni-based alloy extraction

According to Figure 1,

Redox potentials of main elements of Inconel-600(Ni, Cr, Fe) and major radioactive elements (Co, Cs, Actinides) are differ at least 0.15 V[Iizuka, xxx].

4. Conclusion

5. Future work

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Equations should be placed on separate lines and numbered:

$$\int_{\alpha}^{\beta}(x-\alpha)^{n}(\beta-x)^{m}dx=\frac{1}{(n+m+1)!}(\beta-\alpha)^{n+m+1} \quad (1)$$

REFERENCES

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