Behaviors of Perfluorocarbon Emulsion during Dissolution of Oxide Layers

Naon Chang 1,2,*, Huijun Won 2,*, Chonghun Jung 2, Seonbyeong Kim 2, Heechul Eun 2 and Yongsoo Kim 1

1 Department of Nuclear Engineering, Hanyang University, 222 Wangsimri-ro, Seongdong-gu, Seoul 04763, Korea; yongskim@hanyang.ac.kr
2 Decommissioning technology research division, Korea Atomic Energy Research Institute, Daedeok-daero 989-111, Yuseong-gu, Daejeon 34057, Korea; nchjung@kaeri.re.kr (C.J.); sbkim@kaeri.re.kr (S.K.); ehc2004@kaeri.re.kr (H.E.)

* Correspondence: cno0622@kaeri.re.kr (N.C.); nhjwon@kaeri.re.kr (H.W.); Tel.: +82-428688604 (N.C.); +82-428682331 (H.W.)

Abstract: This study investigates the dissolution behavior of oxide layers containing radionuclides using perfluorocarbon (PFC) emulsion as a reusable medium. Chemicals such as PFC, anionic surfactant, and H2SO4 are used for preparing the PFC emulsion, and emulsified using an ultrasonication process. The FTIR results show O–H stretching that is formed by the interaction of the carboxyl group of the anionic surfactant with the hydroxyl group of water containing H2SO4, and find that the H2SO4 can be homogeneously dispersed in the PFC–anionic surfactant–H2SO4 emulsion. The dissolution test of the simulated Cr2O3 specimen is conducted using PFC emulsion containing KMnO4. Through the weight losses of specimens and Scanning Electron Microscope-Energy Dispersive X-ray Spectrometer (SEM-EDS) analysis, it is confirmed that the Cr2O3 layer on the SUS304 specimen is easily dissolved using PFC emulsion. During the dissolution of the Cr2O3, it is observed that the dispersed H2SO4–KMnO4 became unstable and separated from PFC emulsion. Based on these results, the behavior of the PFC emulsion during the dissolution of the oxide layer is explained.

Keywords: perfluorocarbon; emulsion; dissolution; oxide layer; decontamination

1. Introduction

Nuclear facilities consist of various metal equipment and metal devices. The metal surfaces can be contaminated with oxide layers containing the radionuclides during the operation of nuclear facilities [1,2]. The contaminated metals increase the amounts of radioactive wastes when they are disposed of without treatment. Therefore, the contaminations on the metal surface are necessary to be removed using various decontamination methods to reduce the amount of radioactive waste generation.

The radionuclides on the metal surface can be removed by dissolving them along with the oxide layer using a chemical solution [2,3]. Although the water-based chemical method is commonly used to dissolve the oxide layer, large amounts of radioactive liquid wastes can be generated. The radioactive liquid wastes are necessary to be treated using UV decomposition or ion exchange resins [3,4]. However, during, the treatment of the radioactive liquid wastes, additional costs are needed, and secondary wastes are generated.

To solve this problem, the methods of using perfluorocarbon (PFC) as a medium for removing the contaminants from the metal surface were studied by various researchers. PFC is the liquid used for cleaning the device, manufacturing the semiconductor, and cooling the electronics [5,6]. PFC is composed of carbon and fluorine, and the chemical formula of PFC is CxFy. The boiling point, latent heat, and specific heat of the PFC are generally lower than water [7]. The density of the PFC is commonly higher than water. For instance, the density of PF-5058, which is one of the PFC, is 1.75 g/mL [8]. In addition, the PFC has a high radiation resistance [9]. The PFC can be usefully applied to remove the radionuclides from the metal surface due to this feature.
Kaiser studied the removal of the loosely-attached radioactive particles from the various surface by applying the ultrasonication in the bath filled with PFC and surfactant. It was confirmed that 78.0–99.7% of contaminants were removed from the surfaces regardless of the type of specimens. Won studied the removal of the contaminants from the metal surface by spraying the PFC and surfactant mixture. As a result, the radioactive particles were removed from the metal surface because the anionic surfactant reduced the adsorption force between the contaminants and metal. These two methods were the physical method for the removal of the contaminants loosely sticking on the metal surface, thus it cannot be applied to the dissolution of the oxide layer with radioactive contaminant fixed on the metal surface. This is because the PFC medium does not mix with the decontamination agent, reacting with the oxide layer. PFC cannot form a homogenous solution after mixing with the polar molecules. To resolve this problem, it is necessary to mix the PFC with the decontamination agent homogeneously.

In order to homogeneously mix the decontamination agent with the PFC, the emulsification can be applied. The emulsification of the PFC and chemicals has been studied by various researchers. Song studied the cancer therapy method using PFC and hydrophobic TaOx nanoparticles emulsified by an amphiphilic polymer. As a result, the tumor oxygenation and radiotherapy were enhanced by the O₂ saturation reaction of PFC emulsion. Flögel developed the in vivo visualization of inflammatory processes by magnetic resonance imaging using biochemically inert PFC nanoemulsions. From the above studies, it was found that the PFC emulsion was utilized as a useful medium in the medical field. It was also confirmed that emulsification was effective in mixing the chemicals homogeneously. Therefore, it is expected that the emulsification method can be applied to mix the PFC medium and decontamination agent. In addition, the decontamination agent dispersed in the PFC emulsion can be separated from the emulsion when the emulsion becomes unstable over time. In this case, the used decontamination agent will be located on the PFC because the density of the PFC is higher than the decontamination agent. For this reason, it is expected that the PFC medium can be easily reused after scooping the decontamination agent and the radionuclides dissolved in the decontamination agent.

In this study, an innovative chemical dissolution method of the oxide layer, P-BED (perfluorocarbon-based emulsion for oxide layer dissolution) composed of recoverable PFC, anionic surfactant, H₂SO₄, was designed. The objective of this study is to effectively remove the contaminants and the oxide layers from the metal surface using the P-BED medium and to obtain the behavior of the P-BED during dissolution. The chemical structures and stability of the P-BED were analyzed by FTIR. The dissolution performance of the P-BED containing KMnO₄ was evaluated by dissolution test for the simulated Cr₂O₃ layers on the SUS304 specimen.

2. Results and Discussion
2.1. Characteristics of the P-BED

Figure 1 shows the FTIR spectra of pure PFC and the P-BED, consisted of PFC–anionic surfactant–H₂SO₄ emulsion, under three different concentrations of H₂SO₄. As represented in Figure 1, two peaks appeared at 2880 and 2975 cm⁻¹ in the pure PFC spectrum and also appeared in the spectra of the emulsions. The two peaks were due to the unique characteristic of PFC. In the three types of emulsions, a sharp peak at 3550 cm⁻¹ increased as the concentration of H₂SO₄ increased from 0.18 to 0.74 M. Hintze identified the O-H stretching spectra near 3600 cm⁻¹ in the vapor-phase sulfuric acid. Therefore, a sharp peak at 3550 cm⁻¹, when the concentrations of H₂SO₄ were 0.37 and 0.74 M, were considered to be due to H₂SO₄. In addition, a broad peak at the range of 3000–3700 cm⁻¹ appeared increased when the H₂SO₄ concentration decreased from 0.74 to 0.18 M. It was reported that the broad band around 3400 cm⁻¹ is associated with the O-H stretching modes of H₂O. Based on this result, we could infer that the interaction of each component in PFC emulsions with the hydroxyl peak of water containing H₂SO₄. After emulsification, the carboxyl groups of the hydrophilic head of anionic surfactant (Krytox
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157FSM) interact with H$_2$O, including H$_2$SO$_4$, and the hydrophobic tail interacts with PFC [17,18]. Therefore, the broad peak in the range of 3000–3700 cm$^{-1}$ could be attributed to the interaction of different bonds between the hydroxyl group of H$_2$O and the anionic surfactant and H$_2$SO$_4$ when the H$_2$SO$_4$ concentration was 0.18 M. Because of these results, it can be determined that the P-BED becomes stable because the bond between PFC, anionic surfactant, and H$_2$O containing H$_2$SO$_4$ increases when H$_2$SO$_4$ concentration decreases from 0.74 to 0.18 M.

Figure 1. FTIR spectra of the pure perfluorocarbon (PFC) and the P-BED (perfluorocarbon-based emulsion for oxide layer dissolution) consisted of PFC–anionic surfactant–H$_2$SO$_4$.

The phase change of the P-BED with the times was observed to investigate the dispersion stability of the P-BED. The photographs of P-BED are shown in Figure 2. Flocculation was not observed within 25 h when the concentrations of H$_2$SO$_4$ in the emulsion were 0.18 M and 0.37 M. On the other hand, the flocculation was observed in the P-BED after 5 h when the concentration of H$_2$SO$_4$ in the P-BED was 0.74 M. It is considered that the flocculation is happened when the emulsion becomes unstable [19]. Therefore, 0.74 M of H$_2$SO$_4$ was not suitable for the condition of P-BED.

2.2. Dissolution Characteristics of the Oxide Layer Using P-BED

Before the dissolution test, the simulated oxide layer on the SUS304 specimen was prepared. The surface morphologies of the SUS304 specimen, before and after the high-temperature oxidation, are shown in Figure 3. Figure 3 also shows the crystalline particles formed on the specimen surface after the oxidation. The particles were an octahedral shape, with a diameter of between 100–400 nm, and were aligned along the linear grooves that were formed during grinding. From the Energy Dispersive X-ray Spectroscopy (EDS) result, the oxide layer mainly composed of Cr$_2$O$_3$, and the content of oxygen of the oxide layer was 61.33%.
Chromium oxide dissolves in an acidic condition, as shown in Equation (1). At 45 °C, the dissolution reaction was calculated by chemical equilibrium calculation program (HSC Chemistry 6.0), and it was confirmed that the reaction proceeds spontaneously. The reaction

\[
\text{Cr}_2\text{O}_3 + 3\text{H}_2\text{SO}_4 \rightarrow 2\text{Cr}_2\text{O}_7^{2-} + 6\text{H}^+ + 3\text{SO}_4^{2-}
\]

### Table 1: Effect of Concentration of Hydrogen Sulfate Acid on the Phase Change of P-BED

| [\(\text{H}_2\text{SO}_4\)] | Before sonication | After sonication |
|--------------------------|------------------|-----------------|
|                          | 0 hr | 5 hr | 10 hr | 20 hr | 25 hr |
| 0.18 M                   | ![Image](image1) | ![Image](image2) | ![Image](image3) | ![Image](image4) | ![Image](image5) |
| 0.37 M                   | ![Image](image6) | ![Image](image7) | ![Image](image8) | ![Image](image9) | ![Image](image10) |
| 0.74 M                   | ![Image](image11) | ![Image](image12) | ![Image](image13) | ![Image](image14) | ![Image](image15) |

**Figure 2.** The phase change of the P-BED consisted of PFC–anionic surfactant–\(\text{H}_2\text{SO}_4\) over time.

**Figure 3.** FE-SEM images of SUS304 specimen surface: (a) Before oxidation and (b) after oxidation.
(1) proceeds as two successive steps, as shown in Equations (2) and (3) [20]. Especially, reaction (3) is known as the Guyard reaction. At the initial dissolution stage, H\(^+\) ion involves in the dissolution reaction and promotes the dissolution of Cr\(_2\)O\(_3\). They were also calculated by chemical equilibrium calculation program (HSC Chemistry 6.0).

\[
\text{Cr}_2\text{O}_3 + 2\text{MnO}_4^- + \text{H}_2\text{O} \rightarrow 2\text{HCrO}_4^- + 2\text{MnO}_2; \\
(\Delta G = -66.56 \text{kcal, at } 45^\circ\text{C}) \tag{1}
\]

\[
\text{Cr}_2\text{O}_3 + 1.2\text{MnO}_4^- + 0.2\text{H}_2\text{O} + 1.6\text{H}^+ \rightarrow 2\text{HCrO}_4^- + 2\text{Mn}^{2+}; \\
(\Delta G = -39.52 \text{kcal, at } 45^\circ\text{C}) \tag{2}
\]

\[
1.2\text{Mn}^{2+} + 0.8\text{MnO}_4^- + 0.8\text{H}_2\text{O} \rightarrow 2\text{MnO}_2 + 1.6\text{H}^+; \\
(\Delta G = -27.04 \text{kcal, at } 45^\circ\text{C}) \tag{3}
\]

Based on the above dissolution reactions, it was decided to use H\(_2\)SO\(_4\)–KMnO\(_4\) as the decontamination agent and inject it into P-BED to remove the Cr\(_2\)O\(_3\) layer during the dissolution test. The effect of the H\(_2\)SO\(_4\) concentration in the P-BED on the dissolution performance was investigated when the KMnO\(_4\) concentration was constant at 0.13 mM. As shown in Figure 4, the weight loss of simulated oxide layer specimen increased with increasing H\(_2\)SO\(_4\) concentration during the dissolution. This result could be explained by the dissolution reaction represented in Equation (2). In particular, the largest amount of Cr\(_2\)O\(_3\) layer was dissolved when the 0.37 M of H\(_2\)SO\(_4\) was contained in the P-BED among the experimental conditions.

![Figure 4. The weight loss of the specimens during the dissolution test for various concentrations of H\(_2\)SO\(_4\) in the P-BED.](image-url)

The oxide layers that remained on the specimen were observed after 25 h when the H\(_2\)SO\(_4\) concentration was lower, as represented in Figure 5. In addition, more oxide layers were removed with increasing the dissolution time. The SEM result was agreed well with the result of the visual test, as illustrated in Figure 6. The more amounts of crystalline particles, which are the oxides, were maintained after the 25 h when the H\(_2\)SO\(_4\) concentration was lower. In particular, the particles were not observed when the H\(_2\)SO\(_4\) concentration was 0.37 M. From this result, it was also confirmed that the dissolution performance of Cr\(_2\)O\(_3\) layers increased when the H\(_2\)SO\(_4\) concentration in the P-BED was higher. The EDS result of the simulated specimen after dissolution is listed in Table 1. The atomic ratio of oxygen after 25 h of dissolution decreased to 7.96% when the H\(_2\)SO\(_4\)
concentration was 0.37 mM. However, the contents of the oxygen after 25 h of dissolution were 59.44% and 65.96% when the H₂SO₄ concentration was 0.18 and 0 mM, respectively. This was because the H⁺ ion was concerned with the dissolution reaction of Cr₂O₃. Based on these results, it was determined that the optimized H₂SO₄ concentration in the P-BED is 0.37 M for the effective dissolution performance among the experimental conditions. In addition, the above results were compared with the result of the Cr₂O₃ dissolution test using ozone conducted by Maroo [21]. They dissolved the Cr₂O₃ powder into the solution by injecting the ozone at 50 °C, which is the similar temperature condition of this research. As a result, only 20% of the injected Cr₂O₃ were dissolved for 6 h. From this, it was expected that the more amount of Cr₂O₃ layer can be removed within the same dissolution time if P-BED is applied compared with application of ozone.

![Figure 5. The change in the specimen’s surfaces during the dissolution test for various concentrations of H₂SO₄ in the P-BED.](image)

| [Chemicals in the P-BED] | 0 hr | 5 hr | 10 hr | 20 hr | 25 hr |
|--------------------------|------|------|------|-------|-------|
| 0.37M H₂SO₄ + 0.13 mM KMnO₄ | ![Image](image) | ![Image](image) | ![Image](image) | ![Image](image) | ![Image](image) |
| 0.18M H₂SO₄ + 0.13 mM KMnO₄ | ![Image](image) | ![Image](image) | ![Image](image) | ![Image](image) | ![Image](image) |
| 0.00M H₂SO₄ + 0.13 mM KMnO₄ | ![Image](image) | ![Image](image) | ![Image](image) | ![Image](image) | ![Image](image) |

**Table 1.** EDS (at X1000) results of the specimen after a 25 h dissolution test under various concentrations of H₂SO₄ in P-BED.

| Element | Atomic Percent (%) |
|---------|--------------------|
| O       | 0.37 M H₂SO₄ + 0.13 mM KMnO₄ | 0.18 M H₂SO₄ + 0.13 mM KMnO₄ | 0.00 M H₂SO₄ + 0.13 mM KMnO₄ |
| O       | 7.96               | 59.44              | 65.96               |
| Cr      | 17.07              | 17.96              | 15.96               |
| Fe      | 67.89              | 22.01              | 17.41               |
| Ni      | 7.08               | 0.58               | 0.66                |
| Total   | 100.00             | 100.00             | 100.00              |
When the same concentration of H$_2$SO$_4$ and the various concentrations of KMnO$_4$ were added in the P-BED, the weight loss of the simulated specimen over the dissolution time is illustrated in Figure 7. The photographs of the simulated specimens during the dissolution test are shown in Figure 8. As shown in Figure 7, the number of removed oxide layers increased as the concentration of KMnO$_4$ increased from 0 mM to 0.13 mM. Because the reaction (2) and (3) increased with increasing the concentration of KMnO$_4$. This result can also be found in the photographs of the specimens in Figure 8. Especially, the oxide layers were fully removed after 25 h of dissolution when the KMnO$_4$ concentration was 0.13 mM. Based on the result, the optimum concentration of KMnO$_4$ in the P-BED for the effective dissolution of the Cr$_2$O$_3$ layer was determined 0.13 mM among the experimental condition.
Figure 8. The change in the specimen’s surfaces during the dissolution test for various concentrations of KMnO$_4$ in the P-BED.

Figure 9 shows the morphology of the simulated specimens after 25 h of dissolution, when the concentrations of H$_2$SO$_4$ was 0.37 mM, and KMnO$_4$ concentrations in the P-BED was 0 to 0.13 mM. The crystalline particles (which are the oxides) on the specimen decreased with increasing the concentration of KMnO$_4$ in the P-BED. EDS results of the simulated specimens after 25 h of dissolution using P-BED was listed in Table 2. The contents of oxygen decreased to 44.17%, 32.97%, and 7.96% after the dissolution when the concentrations of KMnO$_4$ in the P-BED were 0, 0.06, and 0.13 mM, respectively. These results were consistent with the results of weight loss, shown in Figure 7, and photographs of specimens, represented in Figure 8. In addition, a part of the oxide layer was removed even when the KMnO$_4$ was absent in the P-BED, as shown in Figure 9. Based on the result, it was found that a portion of the oxide layers could also be removed by reaction with H$^+$ ions.

Figure 9. SEM images of the specimens after a 25 h dissolution test under various concentrations of KMnO$_4$ in the P-BED.
Table 2. EDS (at X1000) results of the specimen after a 25 h dissolution test under various concentrations of KMnO₄ in P-BED.

| Element | Atomic Percent (%) |
|---------|---------------------|
|         | 0.37 M H₂SO₄ + 0.00 mM KMnO₄ | 0.37 M H₂SO₄ + 0.06 mM KMnO₄ | 0.37 M H₂SO₄ + 0.13 mM KMnO₄ |
| O       | 44.17               | 32.97               | 7.96               |
| Cr      | 16.23               | 9.80                | 17.07              |
| Fe      | 37.98               | 54.09               | 67.89              |
| Ni      | 1.61                | 3.15                | 7.08               |
| Total   | 100.00              | 100.00              | 100.00             |

2.3. P-BED Behaviors during the Dissolution of the Oxide Layer

Figure 10 shows the photographs of the P-BED consist of PFC, anion surfactant, 0.37 M H₂SO₄, and 0.13 mM KMnO₄ before applying the ultrasonication process and during the oxide layer dissolution test. H₂SO₄–KMnO₄ was injected in the P-BED because they were used as the decontamination agent in the PFC medium. The H₂SO₄–KMnO₄ with a purple color was located at the top of the PFC–anionic surfactant–H₂SO₄–KMnO₄ mixed solution before the ultrasonication. The reason is that the density of H₂SO₄–KMnO₄ was lower than that of PFC, 1.75 g/cm³ [6]. According to the pourbaix diagram, the purple color was caused by MnO₄⁻ ions [22]. After the ultrasonication process, H₂SO₄–KMnO₄ was uniformly dispersed in the PFC, and the P-BED changed to a cloudy phase, as shown in Figure 10. After 25 h of dissolution, the flocculation and the separation of some agents were observed from the P-BED. The flocculated and separated agents were the used decontamination agent. The separated agent did not have a purple color, unlike before the dissolution test. Because the purple MnO₄⁻ ions in the agent were consumed during the Cr₂O₃ removal reactions indicated in Equations (1)–(3). The flocculation and separation of the used decontamination agent, however, were not observed when the dissolution of the oxide layer was not conducted, as shown in Figure 2. Through the results, it was confirmed that the P-BED became unstable as MnO₄⁻ ions were consumed during oxide layer dissolution.

![Figure 10](image_url)

Figure 10. Photographs showing the formation change of the P-BED composed of PFC, anionic surfactant, 0.37 M H₂SO₄, and 0.13 mM KMnO₄ before ultrasonication and during the dissolution test.

Based on the results, the behaviors of the P-BED during the dissolution of the oxide layer were suggested, as indicated in Figure 11. As shown in Figure 11a, the hydrophilic head of the anionic surfactant, which had the carboxyl group, formed an O–H bond with the water in the H₂SO₄–KMnO₄ after the ultrasonication process. In addition, the P-BED was emulsified as the hydrophobic tail of the anionic surfactant interacted with PFC. As seen in Figure 11b, the number of bonds between the anionic surfactant and the water decreased as the P-BED became gradually unstable over time. However, the flocculation did not occur at this time, and the H₂SO₄–KMnO₄ participated in the dissolution reaction of the oxide layer. The bond between the water and the surfactant decreased further over time, and the P-BED offered more activation sites for dissolution reaction. Therefore, oxides were dissolved in the metal ions. The metal ions coexisted with H⁺, SO₄⁻, and K⁺ ions.
in the decontamination agent of P-BED. Afterwards, the \( \text{H}_2\text{SO}_4-\text{KMnO}_4 \) participated in the dissolution reaction of the oxide layer began to flocculate with each other, as shown in Figure 11c. After finishing the dissolution, the used decontamination agent containing various ions was finally separated, and they were located at the top of the P-BED as represented in Figure 11d. Due to the characteristic of P-BED, the used decontamination agent can be easily removed from the P-BED by scooping method after finishing the oxide layer dissolution. Therefore, the PFC and surfactant mixed solution located at the bottom can be reused for removing the oxide layers after re-injecting the decontamination agent and applying the ultrasonication process.

Figure 11. Schematic diagrams of the P-BED behaviors during oxide layer dissolution; (a) P-BED after ultrasonication process, (b) oxide layer dissolution by reaction with unstable P-BED, (c) \( \text{H}_2\text{SO}_4-\text{KMnO}_4 \) flocculation after participating the oxide layer dissolution reaction, and (d) separation of \( \text{H}_2\text{SO}_4-\text{KMnO}_4 \) after finishing the oxide layer dissolution.

### 3. Materials and Methods

#### 3.1. Materials

P-BED was composed of 93 vol% of PFC (3M, PF-5058, St. Paul, MN, USA), 3 vol% of anionic surfactant (Krytox 157FSM, Dupont, Wilmington, DE, USA) and 4 vol% of a decontamination agent. The total volume of the P-BED is 50 mL, and they were prepared in the bottle with a lid. The decontamination agent consisted of \( \text{H}_2\text{SO}_4 \) (98%, DAEJUNG, Siheung-si, Gyeonggi-do, Korea). When the dissolution test was performed, \( \text{KMnO}_4 \) (>99.3%, DAEJUNG, Siheung-si, Gyeonggi-do, Korea) was additionally injected into the decontamination agent. DI water (Direct-Q® 3 UV, Millipore, Burlington, MA, USA) was used for preparing the decontamination agent. The concentrations of \( \text{H}_2\text{SO}_4 \) and \( \text{KMnO}_4 \) in the P-BED were 0 to 0.74 M and 0 or 0.13 mM, respectively.

#### 3.2. P-BED Preparation

The P-BED was prepared through the process, as represented in Figure 12. Firstly, PFC–surfactant–\( \text{H}_2\text{SO}_4 \) and \( \text{KMnO}_4 \) agent mixed solutions were prepared. The solution was stirred at 500 rpm for 5 min using a magnetic stirrer (HSD 150-03P, MTOPS, Yangju-si, Gyeonggi-do, Korea). After stirring, the ultrasonication process was applied to the mixed solution using ultrasonic (VC-505 Ultrasonic Processor, SONICS, Newtown, CT, USA). The decontamination agent in the PFC was dispersed homogeneously through this process.
The application conditions of ultrasonic were 500 Watt of power, 20 kHz of frequency, and 80% of amplitude. The ultrasonication process was conducted for 3 min.

Figure 12. Preparation of P-BED.

3.3. Dissolution Test

The dissolution performances of P-BED containing KMnO₄ were evaluated using the simulated oxide layer of the SUS304 specimen. The specimen was prepared by a high-temperature oxidation process using a muffle furnace (MF-12, Hanyang Science Equipment Co., Nowon-gu, Seoul, Korea). The high-temperature oxidation process was conducted at 800 °C for 32 h. Before the oxidation process, the specimens were treated using 800 grit sizes of sandpaper. The simulated oxide layer mainly consisted of Cr₂O₃. The dissolution test was performed by immersing the simulated oxide layer specimen in the 50 mL of P-BED, while the P-BEDs were stirred at 250 rpm using a magnetic stirrer (MTOPS, HSD150-03P). The P-BEDs were stored in the bottle with the lid during the dissolution test. The temperature of P-BEDs was maintained at 45 °C. To compare the performance, the mass of the specimen was measured at 5, 10, 20, and 25 h, and the appearance change of the specimen was observed during the test. The weight loss was calculated using Equation (4).

\[
\text{Weight loss} = \frac{\text{Initial specimen weight} - \text{specimen weight at time } t}{\text{surface area of specimen}}, \ [g/m^2] \tag{4}
\]

3.4. Analysis

The bonding characteristics of the P-BED were analyzed using FTIR (ABB, MB3000). The P-BED was filtered using a PTFE syringe filter (0.2 µm pore size, Whatman, Maidstone, Kent, UK) before the FTIR analysis. The morphology of the simulated oxide layer specimens before and after the high-temperature oxidation was observed using FE-SEM (Merlin Compact, ZEISS, Oberkochen, Land Baden-Württemberg, Germany). In addition, the composition of the simulated oxide layer specimens was analyzed by EDS (ZEISS, Merlin Compact, ZEISS, Oberkochen, Land Baden-Württemberg, Germany). During the dissolution, the weight change of the simulated oxide layer specimen was measured using the electronic micro-balance (XS204, Mettler Toledo, Columbus, OH, USA).

4. Conclusions

A number of metallic equipment installed in the nuclear facilities are contaminated with radionuclides contained in the oxide layer during the operation. The radionuclides have been generally removed by dissolving the oxide layer using the chemicals in an aqueous solution. It, however, generates a large amount of radioactive liquid waste as a secondary waste after dissolving the oxide layers and contaminants. Therefore, the removal method for dissolution of the oxide layer using reusable chemicals called P-BED is suggested in this study. P-BED consists of PFC, anionic surfactant, H₂SO₄, and an oxidant or the reducing agent. An advantage of P-BED is that the P-BED can be reused the PFC and
surfactant after easily removing the used decontamination agent located at the top of the spent P-BED. This is because the emulsion medium is easily separated over time, and the used decontamination agent can be separated from the P-BED. They can be removed by scooping from the top of the solution because the density of the decontamination agent is lower than PFC. Therefore, the P-BED can be effectively used for decontamination of metal equipment, and it can contribute to reducing the amount of radioactive waste generation.

FTIR result of P-BED revealed that the homogenous dispersion of H$_2$SO$_4$ in the PFC was caused by the interaction between the carboxyl group of anionic surfactants, and the water including H$_2$SO$_4$. The results of the dissolution test showed that the simulated oxide layer specimen was satisfactorily removed by applying the P-BED method. During the dissolution of the oxide layer, the dispersion stability of the P-BED gradually decreased, and the H$_2$SO$_4$–KMnO$_4$ was separated from the PFC. In addition, it was confirmed that the MnO$_4^-$ ions in the P-BED were consumed as participating in the dissolution reaction of Cr$_2$O$_3$. From the results, the behaviors of the P-BED during the dissolution of the oxide layer were illustrated. Based on the research result, it is possible to evaluate the oxide layer removal performance using the reused P-BED in the future.

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