A Study on the Pelletization and Leaching Characterization of Polymer Waste Form Generating After Soil Washing

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1. Introduction

Radioactive substances may contaminate soil of a wide area when a nuclear power plant undergoes an accident or is decommissioned. Soil contamination caused especially by cesium makes it inevitable to decontaminate it to be reused for residential purposes for the residents or as industrial land. As a way to decontaminate the polluted soil in the wide area within the short period of time, there is soil washing. However, after the process of soil washing generates the secondary waste in the form of sludge/slurry [1]. Such a secondary waste can't dispose immediately, thus requiring it to be packaged and treated to make it disposed, which may involve solidification with a proper solidification agent, such as cement and polymer.

This study manufactured the disposal non-conformity simulated waste samples through a preliminary case study of soil washing. The simulated powder samples were produced in the form of powder with fine soil (silt, clay)-Cs-flocculating agent all mixed together. Especially, the samples in powder form were pelletized with a roll compactor to reduce the volume. Lastly, a leaching test was carried out in line with ANS 16.1 proposed by the U.S. Nuclear Regulatory Commission (NRC) to identify the leaching characteristics of the manufactured waste form. Therefore, polymer was used as a solidification agent and a specimen for leaching test (D 50 x H 50mm) was manufactured by incorporating powder and pellets, respectively.

2. Experiments

2.1 Materials

2.1.1 Contaminated soil

Soil samples used in this experiment were taken from an area near a nuclear power plant. Later, they were dried at room temperature and were classified per grain size using a sieve shaker (Daewha Tech, Analysette 3 Pro) to use only soil particles of size smaller than 38 µm. This considered that the characteristic of radioactive cesium that is strongly adsorbed onto fine soil when released to the atmosphere. Later, soil and flocculating agent (J-AF, Jeon Tech Co., Ltd.) was injected in the ratio of 10:1, which then was agitated with 0.1 mmol/L cesium solution. Then, it was dried for 48 hours using a dry oven (Jongro Industrial CO., STD, VTEC-75). Subsequently, the samples were crushed and turned into powder to form them into pellets and their moisture content was regulated to be 7 %. In addition, Co, Cs, and Sr were injected in the form of chloride with a tracer for a leaching test. Table 1 shows the initial concentration of the injected chemical species.

Table 1. Initial concentration of chemical species.

<table>
<thead>
<tr>
<th>Components</th>
<th>Sample No.</th>
<th>Co (mg)</th>
<th>Cs (mg)</th>
<th>Sr (mg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pellets</td>
<td>J-19</td>
<td>482.00</td>
<td>603.44</td>
<td>391.34</td>
</tr>
<tr>
<td></td>
<td>J-20</td>
<td>482.81</td>
<td>604.39</td>
<td>391.95</td>
</tr>
<tr>
<td>Powder</td>
<td>J-px-CG-05</td>
<td>245.56</td>
<td>307.44</td>
<td>199.37</td>
</tr>
<tr>
<td></td>
<td>J-px-CG-06</td>
<td>280.69</td>
<td>351.36</td>
<td>227.89</td>
</tr>
</tbody>
</table>

2.1.3 Polymer

Epoxy resin used in this experiment was made of the main ingredient (YD-128), hardener (G-1034), and diluent (LGE) purchased from Kukdo Chemical. YD-128 as the standard bisphenol-A liquid epoxy resin shows outstanding adhesive force and heat resistance. G-1034 as polyamide resins boasts of exceptional hardening capability at room temperature as well as adhesive force with less stimulation and toxicity. However, when it is used by mixing only the main ingredient and hardener, its viscosity is very high, making it difficult to be mixed with pellets. Therefore, LGE was added to regulate its viscosity. Here, the main ingredient and the hardener were decided to be mixed at the ratio of YD-128:G-1034 = 65:35 phr, recommended by the manufacturer. Also, LGE was added in the amount of 10 wt.% of YD-128.

2.2 Polymer waste form

2.2.1 Manufacturing the Specimens for Leaching Test

The integrity of pellets for each operating condition of the device was confirmed in a preliminary study, whose results were analyzed to come up with the optimal operating conditions [2]. Therefore, manufactured samples for this study was made into pellets with a roll compactor. As shown in Fig. 1, the
optimal operating conditions were as follows: roll speed 1.5 rpm, feeding speed 25 rpm, and hydraulic pressure 28.44 MPa.

The specimens for the leaching test were manufactured as follows: 1) fill the container with pellets and inject polymers into the voids between pellets to make them solidified, and 2) mix powder and polymers to make them into waste form to be used as a control group. Here, the same solidification agent and the mixture ratio were adopted and the quantity of the pellets injected into the waste form was confined to 60 wt.% (J-19, J-20) on average. And, in case of powder waste forms, the quantity of powder injected was set at 55 wt.% (J(p)-CG-05), and 60 wt.% (J(p)-CG-06) respectively. Later, they were solidified inside a thermostat that was maintained at the temperature of 60°C, and upon their solidification, the waste forms were removed from the cylindrical mold. Then, the upper plane of the waste forms were made in parallel to the lower plane and surface-treated to make sure that the pellets were not exposed to the outside. The size of a manufactured waste form was D 50 x H 50mm and shown in Fig. 2.

2.3 Leaching test

A container adopted for the leaching test was made of PTFE (Poly Tetra Fluoro Ethylene), as shown in Fig. 3. And, the specimens were dipped in a leachate for 90 days in line with ANS 16.1 recommended by NRC. Demineralized water sold in Korea was used as the leachate, which was replaced every cumulative time, and the replaced leachate was used to measure pH, Electrical Conductivity (EC), and the nuclide concentration.

2.3.1 Measuring the pH & electrical conductivity

pH and electrical conductivity of the leachate replaced at an interval of cumulative time were measured using a pH/Conductivity meter (Orion Star A215, USA). The measurements were put in graphs as indicated in Fig. 5 and 6. As shown in Fig. 5, the pH concentration of the waste forms dropped significantly in an initial stage of the test, but later went on an upward trend. The pH concentration of the powder waste forms were quite high around 90 days, but all specimens were found to be within the range of 6 to 8. Furthermore, Fig. 6 demonstrates that electrical conductivity of the pellet waste form showed modest changes compared to that of the powder counterpart. Especially, that of the powder waste form (J(p)-CG-05, J(p)-CG-06) gradually increased, which was estimated to be attributed to the dissolution of some of the ions in large quantities from the surface of the specimens depending on the quantity of injected wastes (55 wt.%, 60 wt.%).
2.3.2 Evaluation of leachability index

An inductively coupled plasma system (X2, I-CAPQ, Attom, Neptune) was used to measure the nuclide concentration of each chemical species in the leachate. The measurements were used as basis to calculate the leachability index of Co, Cs, and Sr, and the cumulative leachability index of each specimen was listed up in Table 2. As indicated in Table 2, the leachability index of Co and Cs of the pellet waste form hovered over approx. three times of the acceptance criteria of disposal sites (≥LX: 6), and that of Sr was approx. two times higher. Especially, the leachability index of J-20 waste form turned out to be most outstanding with Co: 17.76, Cs: 17.74, Sr: 14.07, as shown in Fig. 7.

Table 2. Average leachability index of chemical species for each specimens.

<table>
<thead>
<tr>
<th>Components</th>
<th>Sample No</th>
<th>Leachability index</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Co</td>
</tr>
<tr>
<td>Pellets</td>
<td>J-19</td>
<td>17.76</td>
</tr>
<tr>
<td></td>
<td>J-20</td>
<td>17.76</td>
</tr>
<tr>
<td>Powder</td>
<td>J(P)-CG-05</td>
<td>15.30</td>
</tr>
<tr>
<td></td>
<td>J(P)-CG-06</td>
<td>14.59</td>
</tr>
</tbody>
</table>

Fig. 7. Leachability index according to leaching time for J-20.

3. Conclusions

This study manufactured contaminated fine soil, generated after the soil washing process, as simulated waste samples. And came up with a way to solidify them, using polymers as a solidification agent for their disposal. Here, the powder samples were formed into highly-compressed pellets through roll compaction. Especially, waste forms were created using the two different methods: those manufactured by incorporating pellets, and those made by mixing powder and polymers to be used as a control group. Therefore, a leaching test was carried out to find out about the leaching characteristics of the manufactured waste forms.

As a result, the pH concentration of all waste forms was found to range from 6 to 8 depending on the cumulative time. Additionally, electrical conductivity of the pellet waste forms (J-19, J-20) was measured at between 1.4 and 8.8 µS·cm⁻¹. However, the powder waste forms (J(P)-CG-06) showed a relatively higher electrical conductivity of up to 95 µS·cm⁻¹ compared to the pellet waste forms. It was confirmed that all of the average leachability index for each specimen were satisfied, and the average leachability index of pellet waste form far exceeded the Waste Acceptance Criteria (WAC) value than powder waste form. Especially, the leachability index of the pellet waste form (J-20) was deemed most outstanding with Co: 17.76, Cs: 17.74, and Sr: 14.07. Therefore, the filling rate of the wastes in the container was enhanced by pelletizing the powder, which led to the conclusion that the leach resistance of the waste form made based on the said method was remarkably better than the existing one of mixing wastes and a solidification agent.

If this is supplemented through further research in the future, this technology is judged to be a technology that can efficiently treat fine contaminated soil generated after the soil washing process.

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