Removal of Cesium From the Field Soil Using Chemical Extraction

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1. Introduction

The operation of nuclear facilities can pollute the soil of the facility site by the emitted gas and liquid radioactive materials. Furthermore, decommissioning the nuclear facility requires significant costs for decontamination and disposal of contaminated soil. A trace amount of Cs is well known to be strongly adsorbed by clay minerals. This factor is also one of the reasons for the use of clay as barriers in the disposal of radioactive waste. Cs is specifically adsorbed onto micaceous minerals notably onto the frayed-edge sites on illite, which account for about 1% of their total cation exchange capacity (CEC).

In this study, the sample was collected near the Kori No. 1 nuclear power plant located in Gyesang-gun, Busan, and the composition analysis and desorption experiment using oxalic acid was performed by contaminating non-radiative cesium.

2. Materials and Method

2.1 Materials

The clay used in this study was sieved using a sieve (Mesh Size 20 um, EF-8F020) with particle size (> 20 um) an collected from the soil Kori No. 1 nuclear power plant located in Gyesang-gun, Busan. Oxalic acid dehydrate, NaTPB, Hydrofluoric acid, Ammonium sulfate purchased from Sigma-Aldrich as a desorbent to remove Cs from Cesium chloride and clay. A shaking heating bath (BS-21, JEIO TECH Company, Korea) was used to conduct desorption experiments at 80°C.

2.2 Hydrothermal Desorption of Cs-gori Clay Minerals

We add oxalic acid 0.015, 0.15, 1 and 1.5 mol/L (35 ml) to the illite (0.35 g) contaminated with CsCl in a 60 ml tube (Graduated Bottle Wide neck, PP 60 ml, Azlon®, UK). The reaction was carried out in a shaking heating bath (BS-21, JEIO TECH Company, Korea) for thermal desorption at 80°C for 3 days. After 3 days, the solid / liquid was separated using a centrifuge (Multi-purpose Centrifuge, Combi-514R, Hanil Science Inc., Korea), and only the supernatant was collected and filtered through a polyvinylidene fluoride (PVDF) membrane filter (pore size = 0.45 um) The desorption efficiency of Cs in clay is analyzed by inductively coupled plasma mass spectroscopy (ICP-MS; ELAN DRC II, Perkin-Elmer, USA).

3. Results and Discussion

3.1 Cs Desorption Effect of the Clay on Oxalic Acid and Complex Desorbents

The higher the concentration of oxalic acid desorbent, the increaser the desorption rate. At the
concentration of 1.5 M, the Cs desorption rate was 88.41%. Furthermore, desorption experiments were carried out with various desorbents in a sequential process. It was confirmed that 92% of Cs desorption was achieved with HF and (NH₄)₂SO₄.

Table 1. Cs desorption rate using oxalic acid and complex desorbents in gori-vicinity surface clay at 80 °C for 3 day

<table>
<thead>
<tr>
<th>Clay</th>
<th>Chemical</th>
<th>Concentration (mol/L)</th>
<th>Cs desorption (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>D.W</td>
<td>0.72±0.03</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.015</td>
<td>39.06±0.13</td>
<td></td>
</tr>
<tr>
<td>Gori</td>
<td>Oxalic acid</td>
<td>0.15</td>
<td>69.61±2.25</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>80.46±0.30</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.5</td>
<td>88.41±3.83</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Oxalic acid</td>
<td>HF</td>
<td>92.22±1.90</td>
</tr>
<tr>
<td></td>
<td>1.5 (mol/L)</td>
<td>(NH₄)₂SO₄</td>
<td>92.27±1.90</td>
</tr>
<tr>
<td></td>
<td>NaTPB</td>
<td>90.51±1.90</td>
<td></td>
</tr>
</tbody>
</table>

3.2 Effects of Oxalic Acid and Complex Desorbents on the Structures of Clay Minerals

Fig. 1 shows the interlayer and structural changes of Clay following treatment with oxalic acid and complex adsorbents using XRD. It was confirmed that the reflection intensity of 2theta = 7.22°, 10.15° peak in oxalic acid were significantly weakened because the structure of clay was changed from crystalline to amorphous structure. Also, the leaching of cations by organic acids causes the making them finer, thinner and weakening the mineral.

4. Conclusion

In conclusion, the oxalic acid proved to be effective in desorbing Cs from clay. The oxalic acid binds to the metal ions forming the crystal of the clay through the chelating mechanism and breaks the interlayer. For that reason, it facilitates Cs desorption. Therefore, it can be effectively used to restore soil in a residential area that has been widely contaminated with radioactive radionuclides such as Fukushima nuclear accident.

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REFERENCES