Structural Change of Non-expandable Mineral Illite by Treatment With Organic Acid (Oxalic Acid) and Extraction of Cesium

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1. Introduction

Illite has a 2:1 structure, with one octahedral sheet and two tetrahedral sheets combined. In addition, the charge balance of potassium between the interlayers causes little swelling. Illite has an FES (Frayed Edge Site) at the end of the interlayer. When this weathering occurs, the potassium may be released and replaced with other cations to desorb Cesium. Oxalic acid is a chelating agent having a carboxyl group, and a carboxyl group is bonded to a metal ion such as silicon or aluminum constituting a crystal of a clay mineral, It has the function of extracting. Therefore, crystals of clay minerals can be destroyed. This reaction can accelerate the release of cesium.

In this study, we investigated the desorption mechanism of cesium and the interlayer changes due to crystal destruction by treating oxalic acid with illite.

2. Main Title

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) as an illite and purchased from the clay minerals society (USA), Oxalic acid dehydrate, sodium chloride, potassium chloride, and calcium hydroxide were 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 a temperature of 80°C.

2.2 Hydrothermal desorption of Cs-Illite

Add oxalic acid 0.015, 0.15, 0.2, 0.5, 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). After adding 1 M KCl and 1 M NaCl to oxalic acid, add the reaction mixture to a 60 ml tube.

The reaction is 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 rate 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

We show Cesium desorption efficiency in illite when treated with 1M concentration of oxalic acid. The reaction was carried out at 80℃ for 1 day and 3 days. As shown in the chart, the desorption efficiency was 94.76% on the first day and 95.58% on the third day.

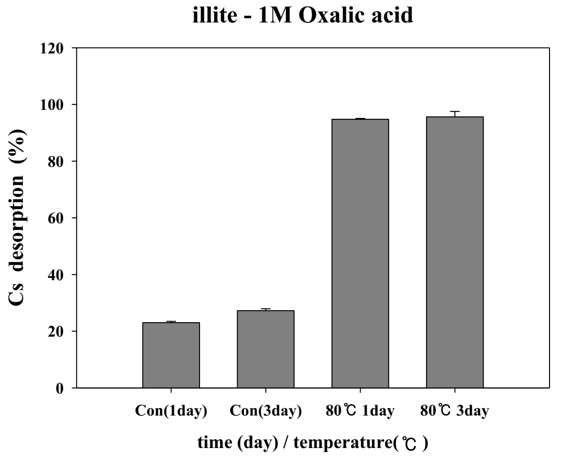

Fig. 1. Cs desorption rate using 1M Oxalic acid desorbents in illite at 80℃ for 3 day.

3.2 The release of Al, Fe, K, Mg affected by the Oxalic acid concentration

This is the result of metal ion elution test for 1M treatment of oxalic acid. It shows the change of elution of metal ion with reaction time.

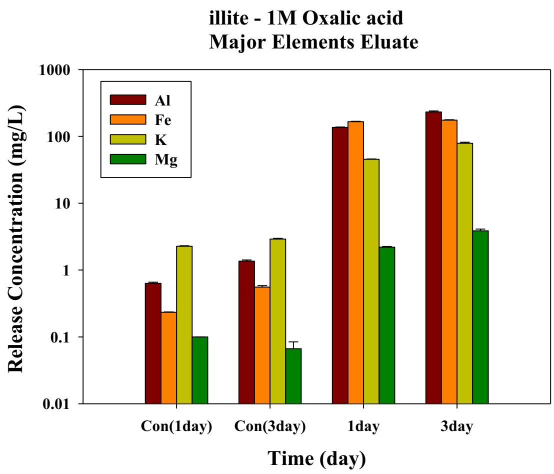

Fig. 2. Extraction of major Clay mineral component of illite by 1M Oxalic acid.

As shown in Fig. 2, when the concentration of oxalic acid was 1M, the elution was performed in the order of Al, Fe, K and Mg, and the amount of elution was greatly increased when compared to the untreated group. However, the change with reaction time did not appear much.

4. Conclusion

In conclusion, Oxalic acis (organic acid) proved to be effective in desorbing Cs from clay. 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.

ACKNOWLEDGEMENT

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