

Cesium Removal From Hydrobiotite by Cationic Polyelectrolyte and Hydrogen Peroxide

Ilgook Kim*, Bo Hyun Kim, Chan Woo Park, In-Ho Yoon, Hee-Man Yang, and Kune-Woo Lee
Decommissioning Technology Research Division, Korea Atomic Energy Research Institute,
111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Republic of Korea
*ilgook@kaeri.re.kr

1. Introduction

Following the accident at Fukushima nuclear power plant in 2011, a considerable amount of research has focused on removing radioactive cesium (Cs) from soil and reducing radioactivity in contaminated area. In particular, Cs ions interact strongly and selectively with 2:1 phyllosilicate clay such as smectite, vermiculite, and illite [1]. Cs is generally adsorbed in the interlayer of these clay minerals, and irreversible adsorption takes places at the frayed edge site (FES), which is highly stable and hardly removable by conventional soil treatment methods. Among the clay minerals, hydrobiotite, interstratified layers of vermiculite and biotite, is produced during weathering of biotite to vermiculite. Vermiculitic clay is known to contain more cesium-specific adsorption sites than illitic clay and the amount of FES can increase as weathering of micaceous minerals proceeds [2].

In this study, therefore, we investigated the behavior of Cs desorption from hydrobiotite by Polyethyleneimine (PEI) as a cationic polyelectrolyte and hydrogen peroxide.

2. Experimental

To prepare Cs-sorbed clay, hydrobiotite was dispersed in an aqueous CsCl solution, and the mixtures were subjected to a horizontal shaker for one week at 25°C. The Cs-sorbed clay was collected by centrifugation at 7000 rpm followed by drying at 40°C. The concentration of Cs in the solution was measured with inductively coupled plasma mass spectrometry (ICP-MS).

For desorption test, Cs-HBT was mixed with DTAB and PEI solution in deionized water and hydrogen peroxide solution with cations. The clay was separated and washed twice with deionized water by centrifugation and was dried at 40°C. The desorbed amount of Cs was analyzed by measuring the Cs concentration in the aqueous phase by ICP-MS.

3. Results & discussion

To analyze mineralogical characteristics of hydrobiotite, the interlayer distance of the clay was investigated by X-ray diffraction (XRD) analysis. Fig. 1 represents the XRD pattern of the dried clay, and a strong peak of hydrobiotite was observed at $2\theta=7.4^\circ$ ($d=12.0 \text{ \AA}$), which is interstratified clay minerals of vermiculite and biotite.

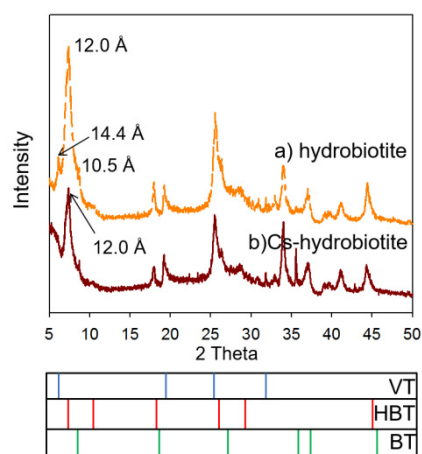


Fig. 1. XRD patterns of (a) raw hydrobiotite and (b) hydrobiotite loaded with Cs at 0.029 mmol/g clay.

To verify the effect of cationic agent on Cs desorption from HBT, the adsorption of desorption agent on clay and Cs desorption were investigated as

a function of desorption agent concentration ranging from 0.16 mmol/g clay to 0.23 mmol/g clay for 24 h. And the pH of the PEI solution was also varied from 3 to 12 (Fig. 2).

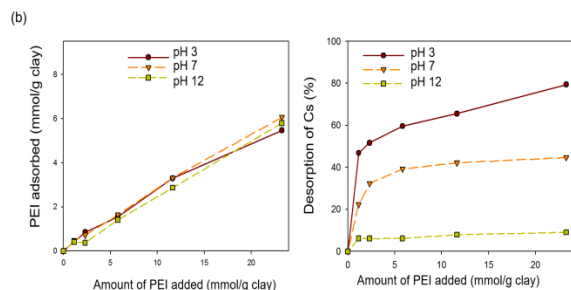


Fig. 2. Adsorption of PEI on Cs-HBT and Cs desorption with various amount of PEI at desired pH for one day.

On the other hand, in order to effect of H_2O_2 on Cs desorption, we tested H_2O_2 solution with cations (NH_4^+ , Mg^{2+} ,) to desorb Cs from HBT. As listed in Table 1, the percentage of the Cs desorbed from the clay tended to increase as the cations added. Also, 35% H_2O_2 solution with 50 mM Mg^{2+} showed the highest Cs removal efficiency. These results indicated that the expansion of the clay interlayer resulting from the inclusion of oxygen from H_2O_2 and the cation exchange by Mg^{2+} ions had a synergetic effect on the Cs desorption.

Table 1. Comparison of Cs removal efficiency by H_2O_2 .

	35% H_2O_2	35% H_2O_2 / 50 mM NH_4^+	35% H_2O_2 / 50mM Mg^{2+}	35% H_2O_2 / 50 mM NH_4^+ + 50mM Mg^{2+}
Cs-HBT	44.6%	46.4%	72.2%	72.8%

4. CONCLUSIONS

We investigated adsorption of Cs ions on hydrobiotite, and Cs desorption by ion exchange with cationic polyelectrolyte and hydrogen peroxide with cations. The Cs desorption efficiencies achieved from two methods were above 75% on hydrobiotite. This is because cesium ions in the interlayers were replaced by desorption agent by ion-exchange. This

comparative Cs desorption study proved that polyelectrolyte-based desorption and hydrogen peroxide methods will have great potential for remediation of Cs-contaminated soil.

REFERENCES

- [1] Motokawa R., Endo, H., Yokoyama, S., Nishitsuji, S., Kobayashi, T., Suzuki, S., and Yaita, T., "Collective Structural Changes in Vermiculite Clay Suspensions Induced by Cesium Ions." *Scientific Reports*, 4, 6585 (2014).
- [2] Kim, B., Park C.W., Yang, H.M., Seo, B.K., Lee, B.S., Lee, K.W., Park S.J., "Comparison of Cs desorption from hydrobiotite by cationic polyelectrolyte and cationic surfactant." *Colloids and Surface A*, 522, 382-388 (2017).