平成２４年度　環境科学院　修士論文内容の要旨

Evaluation on fixation of cesium ions in Japanese bentonite clays
日本のベントナイト粘土へのセシウムイオンの固定化現象の評価

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[Introduction] Bentonite has found as the suitable material for radioactive disposal. Smectite, a major constituent of bentonite, is a ubiquitous laminar clay in the environment. Although swelling, large cation-exchange capacity (ca. 1.1 meq/g) in the interlayer and high selectivity for Cs^+ ion sorption were confirmed as the properties of smectite, studies on irreversibility of Cs^+ ions sorption on bentonite clay differ in a wide way (e.g., Iijima et al., 2010). In contrast, that frayed edge sites of layer fix Cs^+ ions in illite (another laminar clay) is an accepted view. So, I studied the fixation reaction of Cs^+ ions in unrefined bentonite samples.

[Experiments] Two Na-bentonites (Yamagata) and one Ca-bentonite (Miyagi), produced by Kunimine Industry, were used without refinement as the samples. (1) Sorption: After 0.3 g of the samples and 30 mL of 5-100 μM CsCl solution with/without 0.5 M of $CaCl_2$, NaCl and KCl (matrix) are shaken for 24 hours at pH= 8.7-9.6, the Cs^+ concentration in the extracts were measured by ICP-MS; (2) Desorption: 0.3 g of the Cs-loaded sample and 30 mL of each extract (the matrix solution, 1 M of oxalic (Ox) acid, NH_4Ox, NH_4Ac, and HNO_3) are shaken for 24 hours, the Cs^+ concentration in the extracts were determined; (3) After the sorption procedure, with initial $[Cs^+]$ = 10 μM, the Cs-loaded samples were incubated for 1-120 days (aging time) at 25 ˚C or 60 ˚C. Then, the incubated samples were served to the Cs^+ desorption by 1 M NH_4Ac. About the clay fraction separated from the desorption process, the Cs fixation ratio and the interlayer distance were measured by ICP-MS and XRD.

[Result and discussion] The average Cs^+ value of three measurements is indicated over all of the study. (1) Over 89% of Cs^+ ions were absorbed in bentonite in all cases; (2) 1 M NH_4Ac and 1 M HNO_3 solutions showed the highest capability for Cs^+ desorption from the Cs-loaded samples. We can regards Cs^+ contents remained in the residue after 1 M NH_4Ac treatment as fixed (irreversible) Cs^+ ions in the clay; (3) 8-9 μg of Cs^+ in 1 g of bentonites were fixed before aging. After aging for 4 months, 20 μg and 13 μg of Cs were fixed in Ca- and Na-bentonite, respectively. In three samples, the interlayer distance decreases gradually with initial Cs^+ concentration for the sorption procedure, and it also changes with aging time (4 months), which may indicate fixation of Cs^+ ions in the interlayer space. Thus, this study demonstrates that Cs^+ fixation occurs in Japanese bentonites, and the irreversibility of Cs^+ sorption increases with aging. The mechanism and adaptability are still under consideration.