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Doctoral Thesis Outline

Thesis title

Enhancement of Cesium Removal from Clay Minerals by Hydrothermal Treatment for Volume Reduction and Decontamination of Nuclear Post-accident Soils in Fukushima (福島原発事故で発生した土壌廃棄物の減容・除染のための水熱処理による粘土からのセシウム脱離促進)

Because of the Fukushima nuclear accident in March 2011, relative large amounts of radioactive Cs contaminated soils had been accumulated in Japan. Therefore, decontamination of these soils and their volume reduction become very urgent issues. Herein, Cs reversibility (adsorption and desorption) on typical clay minerals, which are responsible for Cs retention in soil, is systematically investigated in present study by combining sorption in different conditions and desorption with various cationic solutions at varied temperature. It was found that Cs desorption from clay minerals was significantly strengthened under the specific conditions such as hydrothermal treatment with multivalent cations. These results may open new ways to satisfactorily remediate the Cs-contaminated soils by achieving short retention time, low environmental stress, and good efficiency.

Chapter 1: General introduction

The environmental contamination and decontamination activities related to Fukushima Daiichi Nuclear Accident in Japan was overviewed in this chapter. For the Cs polluted soil collected from primary decontamination activities, their general management and treatment were compiled and the technologies for volume reduction of these contaminated soil were compared. To explain the difficulties for volume reduction, the Cs retention and migration behaviors in contaminated soil were reviewed. The unsolved problems for soil remediation were clarified and the objectives and structures of this thesis were finally outlined.

Chapter 2: Adsorption properties of Cs on clay minerals

The properties of Cs adsorption on various clay minerals were detailedly investigated in this chapter. The adsorption isotherms and kinetics under the effect of size fractions and coexisting cations were examined and fitted to different models to clarify the mechanism of Cs adsorption on different binding sites on micaceous clays. The composition, microstructure, lattice parameters and elemental maps of clay particles before and after cesium adsorption were analyzed by various characterizations of XRF, XRD, SEM and EDX to expound the cation exchange process during Cs adsorption and confirm the specific distribution of sorbed Cs.

Chapter 3: Desorption of Cs from vermiculitized biotite by ambient treatment with various cations

In this chapter, desorption behaviors of Cs from typical micaceous clay of vermiculitized biotite (VB) were investigated by exaction with various solutions at room temperature. Prior to Cs desorption, the VB was firstly pretreated by saturation of specific cations and then sorbed Cs to saturation amount. The Cs sorption amounts on these three pretreated VB were measured and distribution of sorbed Cs on different binding sites were estimated. Then desorption of Cs from known binding sites was investigated by semicontinuous extraction of Cs sorbed VB with various solutions, to ascertain abilities of various cations in desorbing Cs from different binding sites. Results suggest Cs retention in collapsed interlayer sites is responsible for its high fixation in micaceous clays while highly concentrated K^+ can slowly substitute the fixed Cs from these collapsed sites at room temperature.

Chapter 4: Desorption of Cs from vermiculitized biotite by hydrothermal treatment with various cations

In this chapter, desorption behaviors of Cs from collapsed interlayers were investigated by exaction of Cs saturated VB with various solutions at higher temperatures. Firstly, seawater was selected due to its free utility and environmental friendly feature. Compared with the long duration time but limited desorption ratio achieved at low temperature, it was clarified that extraction by seawater at subcritical temperature (250 °C) could greatly improve Cs desorption efficiency even at relative short duration time. Furtherly, different roles of containing salts in seawater in Cs desorption were examined. Results suggested that it was the divalent cations (Mg^{2+}/Ca^{2+}) rather than monovalent cations (K^+) that revealed effective substitution of Cs from its collapsed interlayers even at a low extraction concentration (0.01M). Finally, multivalent cations (Al^{3+} , Fe^{3+} or La^{3+}) were selected to extraction the sorbed Cs under the same condition to increase the desorption ratio. It was suggested that desorption of Cs from its collapsed interlayers could be greatly enhanced by hydrothermal treatment through specific cations with suitable cationic properties, such as larger hydrous radius, high hydration energy and high ionic valence.

Chapter 5: Validation of enhanced desorption of Cs from actual contaminated soil

This chapter examined whether the enhanced desorption of Cs could be achieved by hydrothermal treatment of the actual contaminated soil with multivalent cations. As a result, negligible amount of Cs was removed from actual soil by treatment with $0.5 \text{ mol L}^{-1} Mg^{2+}$ at intermediate temperature (60 °C), while nearly complete Cs removal was impressively achieved at higher temperature (250 °C), which clearly validated desorption results as achieved for Cs-VB and definitely reinforced the applicability of current reported method for practical Cs decontamination with soil remediation.

Chapter 6: Conclusions and Recommendations

The important results and findings obtained from Chapter 2 to 5 were summarized.