Elucidation of Mechanism of Cesium Immobilization in Contaminated Soil and Development of Practical Technology for Reducing the Volume of Radioactive Waste

Principal Investigator: Kenji Tamura
Institution: National Institute for Materials Science, Tsukuba City, Ibaraki, JAPAN
Tel: 029-860-4667 / Fax:029-860-4667
E-mail: TAMURA.Kenji@nims.go.jp
Cooperated by: Elucidation of Cesium Immobilization Mechanism (The University of Tokyo and Hosei University), Development of Practical Technology for Reducing the Volume of Radioactive Waste and Post-Treatment Measures for the Environment (Ehime University and Center of Analytical Quality Control for Environment)

[Abstract]

Key Words: Radioactive Cesium, Contaminated Soil, Decontamination, Volume Reduction, Weathered Biotite, Hydroxyaluminum-Interlayered Vermiculite (HIV), Pollucite, Hydroxyapatite, Recycle

The soils collected in Fukushima were evaluated by high temperature XRD measurements. The peak at 2θ = 6.3° (d_{001}= 1.4 nm) became broad on increasing temperature and converged to around 9° at 400°C and above. It was suggested that the soils contained hydroxyaluminum-interlayered vermiculite (HIV). Based on the results, a model soil was prepared by intercalating phlogopite with hydroxyaluminum ions. On adsorbing Cs^{+} ions by the model soil, the adsorption mechanisms were investigated in detail. For comparison, a structural model representing extremely stable adsorption sites was constructed for radioactive cesium (RCs) by the MD simulations based on experimental data[1]. Combining the experimental and theoretical results, it is postulated that there exist multiple RC adsorption sites near hydroxyaluminum in addition to the ion exchange sites. In order to desorb RCs from such adsorption sites, a two-step treatment method was developed: (step 1) to treat thermally a soil with molten salt and (step 2) to wash it with a low-concentrated acid aqueous solution[2]. As the practical procedure, radioactively contaminated soils were treated with molten salts such as $CaCl_2$ at nealy above 800°C (step 1). Thereafter the treated soil dispersed in an aqueous solution containing 1M H_2SO_4 was irradiated with microwaves at 800 W for 3 minutes (step 2). After filtration, the radioactivity concentration of the solid precipitate was reduced to 8000 Bq / kg or less. Then, in order to immobilize the desorbed RCs, the following process was developed. The acid-treated aqueous solution was passed through a mordenite (MOR) column. The pollucite (POL) was prepared by hydrothermally treating the MOR at 200°C under alkaline conditions. The POL thus synthesized was composed of a 24-hedron crystal of about 20 μm and showed high RCs selectivity. Nearly all RCs of contaminated soil (100,000 Bq/kg, 20 g) could be transferred to 0.6 g of POL. The radioactivity concentration of POL was concentrated to 1800,000 Bq/kg. Considering the maximum Cs content of POL, highly concentrating of RCs is possible. In this project, we also considered the reuse of purified soil for potting soil, slag, zeolite, etc.

[References]

1) T. Kogure: Clay Minerals, **55**, 3, 203-218 (2020), Visualization of clay minerals at the atomic scale.
2) K. Tamura, H. Yamashita, T. Kogure, M. Morita, A. Yamagishi, and H. Sato: Clay Science, **25**, 7-11 (2021), Removal of cesium ions from radioactively contaminated soils using microwave treatment.