An EXAFS study for characterizing the time-dependent adsorption of cesium on bentonite

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Abstract

Bentonite is considered to be used as buffer material in the final disposal repositories of radioactive waste. For long-lived ^{135}Cs with half-life of 2.3×10^6 y is a key radionuclide in high-level waste, and lots of ^{137}Cs with half-life of 30.2 y existed in low-level waste, the adsorption of Cs on bentonite is a critical issue for evaluating the long-term safety of radioactive waste disposal. In this study, EXAFS technique was used to characterize the time-dependent process from beginning to equilibrium of adsorption. From the results of this study we found the changes, including Cs adsorption sites, Cs-O distances between Cs and oxygen atom, and adsorption species of Cs ion occurred before the reaction reached equilibrium. The fraction of OS complex of Cs adsorbed on bentonite can refer the CN($Cs\text{-}O_{1st}$) /CN ($Cs\text{-}O_{2nd}$) ratio of coordination numbers, and this study found the OS complex that was the major adsorption species of Cs adsorbed on bentonite. Beside the ratio CN($Cs\text{-}O_{1st}$) /CN ($Cs\text{-}O_{2nd}$) give the information of adsorption site, we also discussed the change of $Cs\text{-}O_{1st}$ and $Cs\text{-}O_{2nd}$ bond distance to identify adsorption sites at different time. Comparing the XRD patterns of montmorillonite and bentonite, we found that the interlayer collapsed after Cs adsorbed on montmorillonite, but it expanded after Cs adsorbed on bentonite. From the result of EXAFS fitting, we found the movement of Cs ion from regular interlayer site to expanded interlayer site, which causing the interatomic distance of $Cs\text{-}O_{2nd}$ decreased with the time increased. It was revealed that the adsorption of Cs on bentonite occurred in two steps. The first step includes the rapid uptake of Cs by attachment to the oxygen atoms of the H_2O molecules on regular interlayer sites, especially for OS complex. The second step includes the slower process that dehydrated Cs ion move from the regular interlayer sites to the expanded interlayer sites. In this study, Cs L_3-edge EXAFS spectroscopy was conducted for the Cs adsorbed on bentonite to identify the Cs adsorption sites with time, which is important in evaluating the mobility of Cs in the environment. Those results have benefit to find the process of Cs adsorbed on bentonite, and it could use for the design of the final disposal of spent nuclear fuel.

Keywords

Bentonite, EXAFS, Cs, Adsorption