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学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称 博士（工学） 氏名 Jaka Rachmadetin

学 位 論 文 題 名

Study on the calcium carbonate precipitation and anion diffusion in compacted bentonite for performance assessment of engineered barrier in the geological disposal
(地層処分における人工バリア材の性能評価のための圧縮ベントナイト中のカルサイトの析出および陰イオンの拡散に関する研究)

Abstract

Compacted bentonite is expected to be used as one of engineered barriers in geological disposal of high-level radioactive waste. For the safety assessment of the geological disposal, it is important to understand fate and transport of radionuclides in compacted bentonite in order to predict long term migration behavior of radionuclides at the radioactive waste repository. Although extensive studies have been conducted to improve the knowledge regarding the performance of bentonite as the engineered barrier, the widely accepted model which can predict long-term transport of radionuclides in bentonite has not been established due to its complexities. Therefore, this thesis intends to improve our understanding in this context in terms of three factors which affects the fate and transport of radionuclides in bentonite; the pore water chemistry, the precipitation processes, and the diffusion mechanism of ions in bentonite.

The pore water chemistry of bentonite was studied from the view of activity coefficient of dissolved ions in the pore water. In this study, the mean activity coefficient of Ca^{2+} and CO_3^{2-} ions was obtained from the estimated concentrations of each ion in the pore water in which CaCO_3 precipitation occurred. The CaCO_3 precipitation was enhanced by electromigration method in which Ca^{2+} and $\text{HCO}_3^-/\text{CO}_3^{2-}$ ions were introduced from the opposite sides of the compacted Na-bentonite under electrical potential gradient. A sequential extraction method using radioisotopes as tracer and saturated CaCO_3 solution as extractant was developed to distinctly determine the concentrations in the porewater as free ions and in solid phase in bentonite. The results show that the exchangeable Na^+ ions were progressively replaced by the incoming Ca^{2+} ions. The compacted bentonite sample can be divided into three zones: Ca-bentonite, a mixture of Ca-/Na- bentonite, and Na-bentonite zones, depending on the degree of replacement of the exchangeable Na^+ by Ca^{2+} ions. Precipitates of CaCO_3 were observed both in Ca-bentonite and Ca/Na-bentonite zones. The experimentally determined mean activity coefficients were at least smaller by a factor of three compared to the theoretical approximation calculated using PHREEQC code assuming dilute-solution conditions with no electrostatic interactions between ions and bentonite surface.

The precipitation processes were studied for CaCO_3 which was enhanced to form in bentonite by the same electromigration method at different experimental periods of time. Electromigration experiments with thin layer source were also carried out to obtain the electromigration parameters of Ca^{2+} and $\text{HCO}_3^-/\text{CO}_3^{2-}$ in compacted bentonite. The results showed that the reaction front of CaCO_3 precipitation

under the experimental conditions moved from the anode side towards the cathode side. This advancing reaction front can be attributed to the faster mobility of Ca^{2+} compared to $\text{HCO}_3^-/\text{CO}_3^{2-}$ ions. The CaCO_3 precipitates which were formed during the electromigration were found to have negligible effect on the transport of Ca^{2+} and $\text{HCO}_3^-/\text{CO}_3^{2-}$ ions.

The anions diffusion mechanism was studied for $^{35}\text{SO}_4^{2-}$ ions in Na-bentonite at different dry densities and NaCl concentrations using back-to-back diffusion method. The results showed that the E_a values were smaller than that in free water in bentonite at dry density of 1 Mg m^{-3} with NaCl concentration up to 0.3 M. When the NaCl concentrations increased above 0.3 M, the E_a value increased and be equal to that in free water. This indicates that diffusion in porewater is predominant for SO_4^{2-} ions in compacted bentonite at dry density of 1 Mg m^{-3} with NaCl concentration higher than 0.3 M. However, lower E_a values than that in free water at NaCl concentration lower than 0.3 M cannot be explained by the diffusion in porewater, suggesting the presence of different diffusion path than porewater for anions in compacted bentonite.

The results of this study have important sequences in defining solubility, chemical reaction and mass transport in bentonite which can improve the safety assessment of geological disposal of high-level radioactive waste.