Effect of pH on smectite dissolution rates under alukaline conditions

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Extensive use of cement is envisaged in geological repositories of radioactive waste for encapsulation, backfilling and grouting purposes. Degradation of cement materials in the repositories is considered to produce a high pH pore fluid with a pH 13.0-13.5 initially, and then the pH of fluid progressively decreases to moderately alkaline due to the formation and evolution of subsequent materials (Atkinson, 1985). The pore fluids with high pH have the potential to migrate from the repository and react chemically with the host rock and bentonites employed to enhance repository integrity. These chemical reactions probably affect the rock's and bentonite's capacity to retard the migration of radionuclides. Especially, the smectite (main constituent mineral of bentonite)-cement fluid interaction has been a key research issue in performance assessment of radioactive waste disposal because bentonite presumably lose some of their desirable properties at the early stage of the interaction. It is therefore important to understand the effect of high pH solution on the chemical and physical behavior of smectite, especially dissolution behavior and rate. Stirred-flow-through dissolution experiments were carried out to measure the dissolution rates of the pure Na-montmorillonite (Kunipia-P) under alkaline conditions and to evaluate the effect of pH on the rates. The structural formula of the sample was calculated to be

Na0.39K0.01Ca0.06 (Al1.56Mg0.33Fe0.09Ti0.01) (Si3.87Al0.13)O10(OH)2

The experiments were performed in NaOH-NaCl and KOH-KCl mixed aqueous solutions at 50℃, 0.3 M in ionic strength, 0.2 mL min-1 in flow rate and valuable pH from 8 to 13.4. Our research addresses here the dissolution rates of Na-montmorillonite within the complete pH interval under alkaline conditions. Hereby, possible effects of other variables were eliminated by conducting all experiments at constant of the factors. Congruent dissolution rates of Na-montmorillonite under the alkaline conditions were obtained from steady state reaction data. The smectite dissolution rates increased as function of pH in both NaOH-NaCl and KOH-KCl mixed aqueous solution systems. Obvious pH effect on the dissolution rate was observed in the present study. On the other hand, any effect of interlayer species on the rate was not observed by comparison of the dissolution rates from NaOH-NaCl system with that from KOH-KCl system. The rate lows were calculated by fitting the dissolution rates to the pHs of output solution and obtained the following equations (from Si concentrations):

Rate (mol m-2 s-1) = 10-14.11aH+-0.25 (in NaOH solution)

Rate (mol m-2 s-1) = 10-13.93aH+-0.21 (in KOH solution)