Erosion, Generation, and Stability of Calcium Bentonite Colloids at the Bentonite/Granite Interface

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1. Introduction

Bentonite has been considered as a candidate buffer material in the underground repository for the disposal of high-level radioactive waste because of its low permeability, high sorption capacity, self sealing characteristics, and durability in nature [1]. The water uptake from the rock and the associated swelling causes a penetration of the bentonite into open fractures, the front of the advancing clay being characteristically very soft. Knowledge of the potential extent of colloid generation is required to estimate the required stability for disposal borehole buffer material and to estimate the potential influence of bentonite colloid on radionuclide transport in deep geological media. It has been recently shown that bentonite colloids can be generated at the host rock/buffer interface of such repositories [2,3]. Bentonite colloids may constitute an additional mechanism for radionuclide migration toward the geosphere and biosphere, provided they are stable. In this study, therefore, the potential for generation of bentonite colloids caused by the groundwater erosion and the resulting bentonite colloids was studies experimentally for a Korean Ca-bentonite under repository relevant conditions.

2. Experimental

2.1. Experimental system

Figure 1 shows the experimental system of BEGES (Bentonite Erosion and Colloid Generation Experimental System) originally developed by KAERI (Korea Atomic Energy Research Institute).

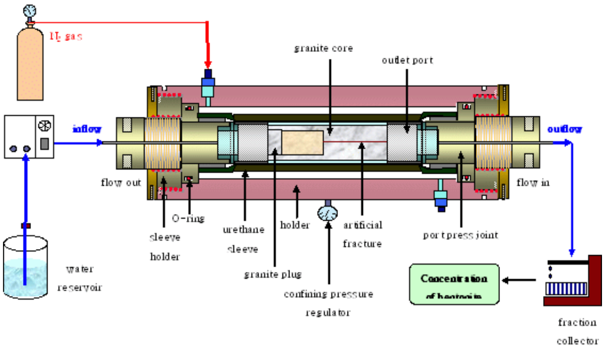

Figure 1. Schematic diagram for the experimental system

As shown in Figure 1, the confining pressure was applied by highly pure (999.99%) nitrogen gas at about 3 MPa and this pressure will hold the granite core tightly not to flow the solution through the perimeter of the granite core. The granite core used in the study was sampled from a domestic granite quarry site located at Dukjeong-myun, Gyonggi-do, Korea. The bentonite used in this study was a bentonite called as 'Kyungju bentonite'. The cation exchange capacity (CEC) of bentonite is 47.7 meq/100 g. Ca^{2+} is a predominant exchangeable cation and the bentonite was identified to be a Ca-bentonite [1].

2.2. Measurement

We used the multi-angle sizing option on the ZetaPlus (BI-MAS, Brookhaven Instruments Corporation) as a dynamic light scattering (DLS) instrument to determine the size and concentration of the bentonite colloids. The accuracy of the instrument is ± 1% to 2% with mondisperse samples and repeatability is ± 1% to 2% with dust free samples. The laser used in 15mW solid state laser. We also calibrated the system with standard latex colloids with diameter of 100 nm. Samples of bentonite and bentonite colloids of known concentration were used to obtain proper calibration curves of count rate vs. concentration [3,4]. The count rate (expressed as counts per seconds, kcps) is in principal determined by the number of particles in the scattered volume. A minimum of 100 particles is required, which corresponds to a count rate above 10 kcps for the PCS [4]. The count rate was proportional to the colloid concentration, however in some concentration ranges.

3. Results and Discussion

3.1. Erosion and generation of bentonite colloids

Results showed that bentonite colloids can be generated at the bentonite/granite interface and mobilized by the water flow although the intrusion of bentonite into fracture by swelling pressure was observed to be small. The higher the flow rate, the higher the concentration of particles eluted and generated in water. Thus different processes of mobilization of theses colloids from the compacted bentonite block have been identified. The erosion of the clay surface due to the groundwater flow together with intrusion processes is the main mechanism that can introduce clay colloids in the fracture of the granite.

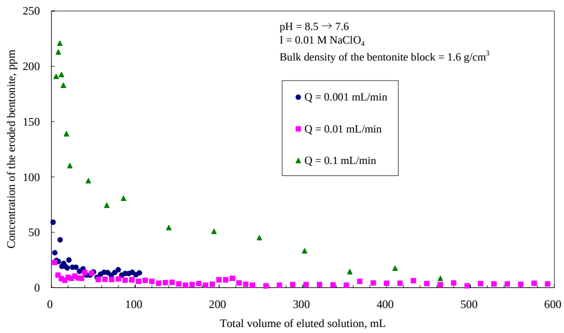

Figure 2. The elution profiles of the generated bentonite colloids

3.2. Stability of the generated bentonite colloids

The potential relevance of these colloids in radionuclide transport was also studied by evaluating their stability in different chemical environments. The coagulation kinetics of natural Ca-bentonite colloids was investigated by means of a time-resolved dynamic light scattering technique, which was recently demonstrated as being a valuable tool for the study of the coagulation kinetics of clays, as a function of pH, ionic strength, and the type of electrolyte cations [4].

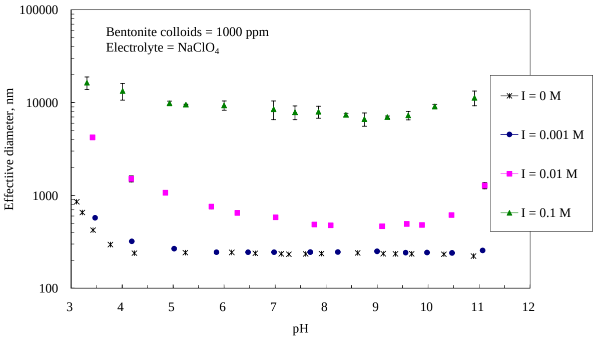

Figure 3. Effects of pH and ionic strength on the effective hydrodynamic diameter of Ca-bentonite colloids at equilibrium after contacting 1 day.

Coagulation rates are often expressed in relative terms as the so-called stability ratio, W, by normalizing them to the rate of fast (diffusion limited) coagulation k^f [5]

$$W = \frac{k^f}{k} = \frac{1}{\alpha}$$

where k is the coagulation rate of the sample under consideration and α is the colloid-colloid attachment efficiency. The fast rate k^f can either be a theoretical value (e.g., Smoluchowski rate) or an experimental value determined for coagulation in the fast regime (i.e., in the absence of repulsive energy barriers). The stability ratio can be determined directly from the initial slopes of coagulation curves as [5]:

$$W = \frac{\left[\left(dr_h / dt \right)_{t \to 0} / C \right]^f}{\left[\left(dr_h / dt \right)_{t \to 0} / C \right]}$$

where superscript f refers to measurements in the regime of fast coagulation. The initial slopes of the resulting coagulation curves $\left(dr_h / dt \right)_{t \to 0}$ were determined by fitting a straight line to the data obtained during the initial stage of coagulation experiment, in which the average radius increases linearly with time.

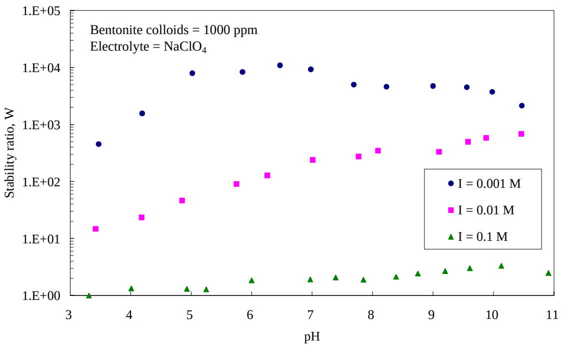

Figure 4. Stability ratio as a function of the pH with varying ionic strength calculated from the coagulation curves

The result in Figure 4 shows that the stability of the bentonite colloids strongly depends upon the pH and ionic strength of the suspensions. It is also shown that the bentonite colloids are unstable at higher ionic strength greater than 0.01 M NaClO$_4$ even at basic pH.

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