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Laboratory determination of migration of Eu(III) in compacted bentonite–sand mixtures as buffer/backfill material for high-level waste disposal

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HIGHLIGHTS

- The migration progress of Eu(III) in compacted bentonite–sand mixtures was researched.
- The hydraulic conductivity of cominpacted bentonite–sand mixtures was measured.
- The migration length of Eu(III) in buffer/backfill material after a certain period of time was forecasted.

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ABSTRACT

For the safety assessment of geological disposal of high-level radioactive waste (HLW), the migration of Eu(III) through compacted bentonite–sand mixtures was measured under expected repository conditions. Under the evaluated conditions, advection and dispersion is the dominant migration mechanism. The role of sorption on the retardation of migration was also evaluated. The hydraulic conductivities of compacted bentonite–sand mixtures were $K=2.07\times10^{-10}$–5.23×10^{-10} cm/s, The sorption and diffusion of Eu(III) were examined using a flexible wall permeameter for a solute concentration of 2.0×10^{-5} mol/l. The effective diffusion coefficients and apparent diffusion coefficients of Eu(III) in compacted bentonite–sand mixtures were in the range of 1.62×10^{-12}–4.87×10^{-12} m^2/s, 1.44×10^{-14}–9.41×10^{-14} m^2/s, respectively, which has a very important significance to forecast the relationship between migration length of Eu(III) in buffer/backfill material and time and provide a reference for the design of buffer/backfill material for HLW disposal in China.

1. Introduction

Compacted bentonite–sand mixtures have been proposed as a suitable buffer/backfill material in the HLW repository in China (Wang et al., 2006). A multi-barrier system is utilized for isolating the waste from the biosphere for the repository of HLW disposal. Based on the concept of the multi-barrier system, buffer/backfill material serves as a significant part of engineering barrier, preventing convective water from flowing due to the low permeability of bentonite, and retarding the migration of radionuclides through buffer by sorption to the matrix. Therefore, the hydraulic conductivity and diffusion coefficient of radionuclides on compacted bentonite–sand mixtures play a very important role in assessing the performances of buffer/backfill material.

The sorption and diffusion of radionuclides in clays have been studied in many researches (Torstenfelt and Allard, 1983, Sato et al., 1992, Yu and Neretnieks, 1997, Wang et al., 2001, Dong et al., 2001, Wang et al., 2004, Wang and Tao, 2004, Montes-H et al., 2005, García-Gutiérrez et al., 2011). It has been reported that ion exchange is a dominant sorption mechanism and another sorption mechanism is surface complexation (Yu and Neretnieks, 1997). The sorption behavior is often represented by the distribution coefficient, K_d, a characteristic parameter of the sorption ability of radionuclides in the solid phase. Most of the diffusion tests were conducted by the in-diffusion (Wang et al., 2004), through-diffusion (Lee et al., 1996), back-to-back diffusion (García-Gutiérrez et al., 2011) or reservoir-depletion method (Lever, 1986), and mainly focused on the effect of dry density (Sato et al., 1992, Lee et al., 1996), sand ratio (Iida et al., 2011), solution concentration (Wang et al., 2004), pH (Wang et al., 2004) or temperature (Martín et al., 2000) on the diffusion coefficient. Few studies considering the advection were available. In this paper, the

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flow-through migration study was conducted on the cylindrical specimens with flexible wall permeameter (US Hombolt). The discharged solution from specimen was analyzed periodically, and at the end of test the specimen was segmented to measure the concentration of Eu(III). Modeling the migration progress of Eu (III) in compacted bentonite–sand mixtures using the advection–dispersion equation was performed in this study, and based on this, obtained the migration length of Eu(III) in buffer/backfill material as a function of time, which provides a reference for the design of buffer/backfill material for HLW disposal in China.

2. Materials and methods

2.1. Materials

The constituents of the mixtures evaluated in this test include the processed GMZ bentonite, from Inner Mongolia, and the quartz sand from YongDeng County in GanSu Province, China. The mineralogical, physical and chemical properties of the bentonite are summarized in Table 1. The particle density of quartz sand used in this research is 2.65 g/cm^3 and particle diameter ranges from 0.5 mm to 1.0 mm.

The sand ratio, R_s is defined as the dry mass ratio of the quartz sand to bentonite–sand mixtures. Quartz sand is uniformly added into the bentonite with R_s=0, 10%, 20%, 30%, 40% and 50%. And then a spray-on process is needed to achieve even objective water content. The mixtures are then placed in polyethylene bags, sealed and kept in a humidor with a room temperature (23 °C ± 2 °C) and almost constant relative humidity (70–80%) for self-curing about 60 h (Zhang et al., 2012a, 2012b). Compacted cylindrical specimens with the size of 100 mm in diameter and 20 mm in height are used for the test. The dry densities of specimens in test are determined to be varying from 1.50 g/cm^3 to 1.90 g/cm^3, as Table 2 shows.

Eu(III) is usually taken as an analog for trivalent actinide ions (Wang et al., 2004). The initial Eu(III) concentration used in this test was 2.0×10^{-5} mol/l.

2.2. Methods

It have be reported that flexible wall permeameter (Fig. 1) is the best suitable equipment for measuring the hydraulic conductivity of the specimen that has a hydraulic conductivity lower than 10^{-7} m/s. When the cylindrical specimen was positioned into the permeameter cell, a confining pressure of 450 kPa was applied to the cell filled with water to push the membrane in contact with the specimen side tightly, and distilled water was used to saturate the specimen with "back-pressure saturation" method wherein the specimen is exposed to water under an applied (positive) back pressure (Bishop and Henkel, 1962) of 150 kPa. After steady-state fluid flow was established through the soil specimen, the fluid (distilled water) in the influent reservoir was changed to the Eu(III) solution at the constant concentrations of 2.0×10^{-5} mol/l. At the same time, a pressure of 200 kPa was initially applied to induce an upward flow in the specimen and the cell was filled with water at a applied confining pressure of 500 kPa. The head in the standpipe was monitored as a function of time, and the hydraulic conductivity of the specimen was calculated from the head data using the falling head method. The discharged solution from the specimen was collected periodically and then filtered using a 0.45 μm filter paper and analyzed for Eu(III) concentration by ICP-MS. No Eu(III) was detected above background in the outflow for the duration of the experiment.

After 300 d, experiment was terminated, and then set-up was disassembled, followed by slicing specimen into thin sections at about 0.50 mm. About 0.30 g soil, taken from each slice which was pestled and homogenized, was then dissolved in 8 ml 10 mol/l HNO_3 and 5 ml 20 mol/l HF with microwave digestion instrument for 45 min and then added 2 ml $HClO_4$ into the digested solution. The solution was heated to dense smoke, then dissolved to clear solution with HNO_3 and volume was adjusted to 50 ml. The amount of Eu(III) in each solution was measured with ICP-MS.

On the basis of Fick's second law, combining with the mass conservation equation for the solute, the one-dimensional advection-dispersion equation was able to modeling the migration of the Eu(III) in this study (Rabideau, 1999):

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x_i}\left(D_{ij}\frac{\partial C}{\partial x_j}\right) - \frac{\partial}{\partial x_i}(v_i C) - \frac{\rho_d}{n}\left(\frac{\partial S}{\partial t}\right)_{srp} - \left(\lambda_a C + \lambda_s \frac{\rho_d}{n} S\right) + \frac{q_s}{n} C_s \quad (1)$$

Table 2
Physical parameters of specimens for the tests.

Parameter	Bentonite–sand mixtures					
Sand ratio (%)	0	10	20	30	40	50
Objective dry density (g/cm^3)	1.70	1.70	1.70	1.70	1.70	1.70
Objective water content (%)	17.00	15.00	13.00	12.00	11.00	10.00

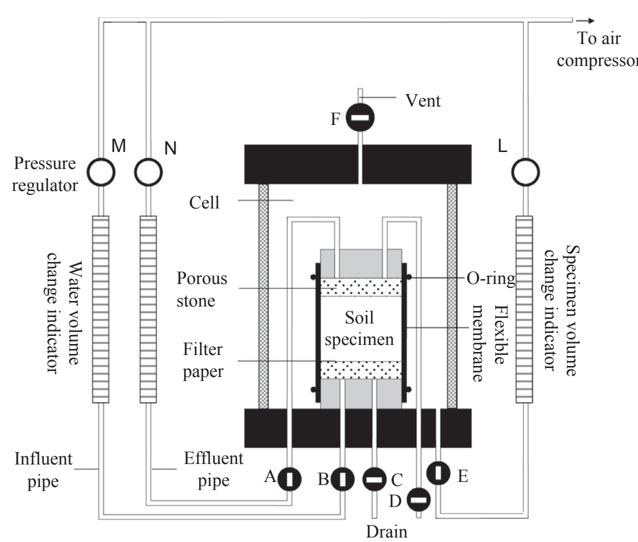

Fig. 1. Sketch of flexible wall permeameter.

Table 1
Physical and chemical properties and mineralogical composition of GMZ bentonite.

Mineralogical composition (wt%)						
Montmorillonite	Quartz	Cristobalite	Feldspar	Kaolinite	Plagioclase	Chalcedony
75.4	11.7	7.3	4.3	0.8	–	–
Physical and chemical properties						
Particle diameter	Specific surface	Air-dried moisture contents	Plastic limit	Liquid limit	Specific gravity	Cation exchange capacity
Main < 2 μm	570 m^2/g	10.53%	32.43%	228.00%	2.71	77.30 meq/100 g