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Effects of compression on porous texture of clay powder: Application to uranium diffusion

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A B S T R A C T

This paper presents the results of tests conducted on the compaction properties of clay. Swelling clays of Algeria are being examined as components of the buffer material. Knowledge of the porous texture in clay samples is a useful element in the microstructural characterization of such materials. The prepared materials were characterized by X-ray diffraction, Differential Thermal and Thermo Gravimetric Analysis, Laser Granulometry, Scanning Electron Microscopy, mercury porosimetry and nitrogen adsorption/desorption to obtain information about their structure and surface texture. The cationic exchange of clay minerals, in particular smectites, influences the cation retention and diffusion processes. These processes influence the migration of contaminants from waste disposal sites through clay barrier. In the present work we studied the effect of cation exchange and compaction on the textural properties of clay. Pellets of the Na-saturated and bulk clay samples were prepared by compression of the powder samples using uniaxial stress at different pressures in order to obtain different densities. The pressures of 31–127 MPa are applied for the production of cylindrical samples with diameter of 20 mm and heights of 15–30 mm. The effect of compaction of clay on the diffusion behavior of uranium was studied for the safety assessment of the radioactive waste. The diffusion process of uranium in compacted clay has been modeled by second Fick's law taking into account the effect of sorption and considering the non-steady state. The diffusion coefficients and profiles concentration values of uranium were calculated by computational method using numerical program based on Newton–Raphson algorithm.

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1. Introduction

Clay materials are often used as barriers to the migration of contaminants from waste disposal sites (Barnett et al., 2000; Bourg et al., 2003; Abdel Rahman et al., 2007; García-Gutiérrez et al., 2008; Maes et al., 2008; Bai et al., 2009). Compacted clays are being considered in many countries as a backfill material in high-level radioactive waste disposal concepts. The clay presents a very high sorption capability for radionuclides, low permeability, long term stability and a very small hydraulic conductivity. Several studies (Muurinen, 1990; Miyahara et al., 1991; Sato et al., 1992; Lee et al., 1996; Molera et al., 2003;

García-Gutiérrez et al., 2004; Melkior et al., 2009) have shown that the characteristics of engineered barrier of clay can be related to the textural properties of the buffer material for high-level nuclear waste repositories. Therefore, it is necessary to characterize the void space of the porous solids. Radionuclide migration to the biosphere will be retarded by the engineered barrier. The concept of a deep geological high-level waste repository includes an engineered clay barrier around the canister.

There are two approaches currently available for the determination of textural properties in porous solids, the mercury intrusion porosimetry method and the capillary condensation

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method (Webb and Orr, 1997; Gaboreau et al., 2011). The characteristic parameters are the surface area, porosity, pore-size distribution and pore connectivity. These characteristic parameters are often used for correlating various aspects of materials performance with textural properties. In addition, Altin et al. (1999) reported that specific surface area of clay depends also on pH of the clay suspension. Ye et al. (2011) have examined the effects of temperature on the soil-water retention curves and properties of bound water during the compacting of bentonite. They have found that the water retention capacity decreases as temperature increases, and the influence of temperature depends on suction. In addition, the value of the hydraulic conductivity becomes constant as hydration progresses, under confined conditions the hydraulic conductivity increases as temperature increases.

The retardation of radionuclides in engineered clay barrier is affected by various properties of compacted clay, such as porosity, dry density (Sato et al., 1992), pore size distribution, the kind of exchangeable cations (Kozaki et al., 2005), initial grain size (Kozaki et al., 1999; Sato and Miyamoto, 2001) and temperature (Kozaki et al., 1996, 1997, 1999). With respect to radioactive waste disposal, diffusion in compacted clay materials has attracted many studies for radionuclides (Torstenfelt et al., 1982; Muurinen et al., 1985; Oscarson et al., 1992; Baik et al., 2003; García-Gutiérrez et al., 2004). Cho et al. (1999) reported that the hydraulic conductivities of high density mixtures of clay and crushed granite were increased up to about one order of magnitude with the increase of temperature from 20 °C to 80 °C. To explain these high values of hydraulic conductivity, the degeneration of adsorbed water on the clay pore wall resulting in the increase of effective porosity was suggested. Ouyang and Daemen (1992) also investigated the hydraulic conductivities of both pure clay and clay crushed rock mixture at a temperature of up to 90 °C, but the results do not show great grains size of porosity. The porosity measured at 90 °C is only about two times higher than those at 20 °C.

The cation exchange capacity is one of the basic properties of clays and clay minerals. The layers comprising tetrahedral and octahedral sheets are separated by an interlayer containing hydrated exchangeable cations which balance the negative charge of the layer. These ions can be exchanged by other cations and the measure of this characteristic property of a clay mineral is known as its cation exchange capacity (CEC). The surface properties of clay depend on chemical composition, nature of the surface atoms, layer charge and type of exchangeable cation.

Ion sorption on clays and clay minerals is controlled by two different mechanisms, a pH-independent adsorption, usually attributed to cation exchange in the interlayers and resulting from electrostatic interaction between the ions and the permanent charge, and a pH-dependent adsorption, thought to result from surface complexation reactions. For the diffusion process, the cationic exchange is an important interaction mechanism between ions in porewater and solid mineral, especially in highly compacted clay where the volume of porewater is small and the fraction of montmorillonite is large (Bradbury and Baeyens, 2002, 2003).

In the present work we studied the effect of cation exchange and compaction on the porosity and pore size distribution textural properties of clay. The change in pressing pressures of clay samples contributes greatly to the increase of porosity. Porous texture (structure and pore volume) influence considerably the physical and mechanical properties of

a material. We studied changes in pellets properties (porosity, pore size distribution, tortuosity and constrictivity) due to consolidation of powder clay.

Reactive transport of radionuclides in porous media is a complex process affected by many parameters, such as the properties of the diffusing species and the properties of the solid itself (porosity, pore structural properties, tortuosity and dry density) (Bourg et al., 2003; Melkior et al., 2009).

The principal mechanism of mass transport through these materials is molecular diffusion, because the permeability of dense clay-based materials is very low. Therefore, the reactive transport is a diffusion-controlled process retarded by sorption (Kohler et al., 1996; Wang et al., 2005; Aldaba et al., 2010).

Uranium diffusion in porous clay media is always found to be coupled with sorption to the solid phase such processes retard the mobility of U (VI) in the porous sediments. To predict U (VI) diffusive transport, it is necessary to know the diffusion coefficient of the U (VI) species, which is determined by both the tortuous nature of the pore network and by characteristics of the molecular U (VI). Since diffusion coefficients are relatively difficult and time-consuming to measure in compacted clays, it is desirable to have other means of estimating them; hence the need to compare measured and calculated diffusion coefficients. Diffusion is generally classified into two states: non-steady state and steady state (Crank, 1975).

Non-steady state diffusion represents the diffusion coefficient in a transient state, which is called apparent diffusion coefficient (D_a). In performance assessment, D_a is accounted as an important parameter for evaluating nuclide retardation in clay (Eriksen et al., 1999; Bourg et al., 2003). In this work, we conducted diffusion cell experiments using the compacted clay to study U (VI) diffusion when it was coupled with sorptive retardation. We attempt to measure experimentally the diffusion coefficients for U (VI) in compacted clays having a wide range of dry density, and to compare the measured diffusion coefficients with those calculated from a numerical program based on Newton-Raphson algorithm.

2. Experimental

2.1. Materials

A bulk clay from Maghnia (Algeria) was used for the study. Samples were crushed and sieved to give a granular material with particle sizes lower than 200 μm . The average particle size measured by a particle size analyzer (laser granulometer, MALVERN) was 30 μm (Korichi et al., 2009). For the preparation of Na-clay samples, the bulk samples were saturated with Na by treating them with 1M NaCl, washing with distilled water and centrifuging. The samples were then freeze-dried. The powder samples were named B and Na-B for bulk and Na-saturated clay samples, respectively. The chemical composition of the original clay was performed by conventional chemical analysis and determination of elemental concentrations by atomic absorption spectroscopy as follows: SiO₂, 62.62%; Al₂O₃, 17.95%; Fe₂O₃, 2.53%; TiO₂, 0.73%; CaO, 2.86%; MgO, 3.24%; Na₂O, 2.24%; K₂O, 0.48% and loss on ignition at 900 °C, 6.35%. The cation exchange capacity (CEC) of the Maghnia clay was 80 mequiv./100 g. The CEC was determined by adsorption of a copper ethylenediamine complex method (Korichi et al., 2009) using a UV-vis spectrophotometer (GBC-918).

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