

Mineralogical and chemical characteristics of the bentonite in the A2 test parcel of the LOT field experiments at Äspö HRL, Sweden

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ARTICLE INFO

Article history:

Available online 11 October 2011

Keywords:

Bentonite buffer
Cation exchange
CEC
Heater experiment
Hydrothermal
Montmorillonite

ABSTRACT

The Long Term Test of Buffer Material (LOT) project at the Äspö Hard Rock Laboratory, Sweden, is a series of medium-scale field experiments focused on validating models and hypotheses concerning long term processes in the bentonite buffer of a repository for high-level radioactive waste. The test parcels emplaced in crystalline bedrock consist of blocks of compacted MX80 bentonite embedding a Cu-tube equipped with a heater to simulate the heat generation from radionuclide decay. The A2 test parcel had been subjected to elevated temperature (up to 130 °C) and hydration by a Na–Ca–Cl type groundwater for almost 6 years when it was retrieved to be analysed. The analyses included determinations of chemical composition, cation exchange capacity (CEC), exchangeable cations and mineralogy. Both the bulk bentonite and dialysed, homo-ionic Na-clay (<2 µm and <0.2 µm fractions) were analysed when relevant.

Sulphate was redistributed in the heated part of the buffer under the thermal and hydration gradients that prevailed during the test period. Anhydrite accumulated in the warmer parts, whereas gypsum was dissolved in the peripheral parts of the buffer where water was supplied. Carbonate dissolution increased with temperature in the warmest parts, whereas chloride behaved conservatively in all blocks. Cu was incorporated in the bentonite matrix at the surface of the Cu-tube indicating some corrosion, which may be explained by reactions in an early stage of the test when trapped oxygen existed in the system. Along with the dissolution/precipitation reactions the porewater composition changed, which resulted in replacement of exchangeable sodium by calcium and magnesium in the warmest zone. Also Mg in the clay (<2 µm and <0.2 µm fractions) displays a clear gradient with peak values at the heater. Because several of the alternative sinks for Mg were eliminated in the sample preparation prior to the chemical analysis (purified clay fractions, removal of carbonates, Na-saturation) the smectite is suggested a candidate sink for Mg. Parallel with the increase in Mg, a loss in Si is indicated and CEC tends to increase in the clay that had been heated at 130 °C. A loss in tetrahedral Si that is balanced by Al, and replacement of Al by Mg in the octahedral sheet would imply that the layer charge of the smectite increased, which would be consistent with the higher CEC values of these samples. The changes in CEC are, however, close to the analytical resolution of the CEC method and no effect of the changes in chemical composition can be detected in the XRD-characteristics of the clay. Therefore, supplementary high-resolution analyses are required to verify whether the structure of montmorillonite has altered in the test period.

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1. Introduction

In the Swedish KBS-3 concept for repositories for nuclear waste, the spent fuel will be stored in copper canisters embedded in a bentonite buffer and emplaced in vertical boreholes in crystalline bedrock at a depth of ~500 m. The main functions of the buffer are to serve as mechanical support for the canister and reduce the ground-water flow across the deposition hole. A buffer of compacted bentonite is considered the best choice to fulfill these

requirements, thanks to the desirable hydro-mechanical and chemical properties. Since these properties are directly related to the crystal chemistry of the predominant mineral montmorillonite, the stability of montmorillonite when subjected to elevated temperatures and hydration by groundwater of variable composition is of vital importance for the long-term performance of the buffer.

Quantitatively, the most important heat-induced reaction in smectitic clays is the progressive conversion of smectite to illite via mixed-layer illite/smectite. Illitization of smectites in early-diagenetic (<~140 °C), geological environments has been extensively studied during the past 60 years, because of its relevance as “geothermometer” in explorations for hydrocarbons (e.g. Weaver, 1989; Pollastro, 1993; Meunier and Velde, 2004). Besides the large

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number of studies on illitization in natural geological settings, numerous hydrothermal laboratory experiments have been made but there is still no general consensus on whether the conversion of smectite to illite is primarily controlled by pressure, temperature, potassium activity, pH, silica activity or other factors. (e.g. Weaver, 1959, 1989; Perry and Hower, 1970; Hower et al., 1976; Eberl et al., 1993; Abercrombie et al., 1994). During the past decade a growing number of field tests at different scales, and of different design and durations have been performed in order to study the effects of simulated repository-like conditions on bentonite clays. These experiments have demonstrated mainly thermo-hydraulic processes and water and solute transports under the thermal and hydration gradients that will prevail during the initial stage of the operation of a repository (Karnland et al., 2000; Plötze et al., 2007; Villar and Lloret, 2007).

The design criteria for the KBS-3 repository stipulate that the temperature should never exceed 100 °C at any position in the buffer and according to models of the temperature evolution, the maximum temperature will be below 90 °C (Hökmark and Fälth, 2003). Karnland and Birgersson (2006) estimated the smectite-to-illite conversion under KBS-3 repository conditions to be insignificant, using the kinetic model and constants of Huang et al. (1993) and extreme values for the potassium concentration of the groundwater at Äspö. The Long Term Test of Buffer Material (LOT) project at the Äspö Hard Rock Laboratory is a series of medium-scale field experiments aiming at validating such models and hypotheses concerning long term processes in the bentonite buffer under repository-like conditions. Compared with the expected KBS-3 conditions, the presently analysed A2 test parcel represents adverse conditions with respect to temperature and temperature gradient, which are factors that will accelerate alteration processes. The A2 parcel consisted of blocks (doughnuts) of compacted MX80 bentonite embedding a Cu-tube equipped with a heater to simulate the heat generation from radionuclide decay. The parcel had been subjected to elevated temperature (up to 130 °C) and hydration by a Na–Ca–Cl type groundwater (~ 7000 mg Cl⁻/L) from the granitic host rock for almost 6 years when it was retrieved in 2006 to be analysed. In the present study the chemical and mineralogical characteristics of bentonite blocks from the heated and the cool parts of the A2 parcel are compared with those of unexposed MX80 bentonite. Parallel mineralogical investigations of bentonite from the A2 test parcel have been performed by the laboratories of Posiva (Finland), ANDRA (France), BGR (Germany) and Nagra (Switzerland) and the results are reported in Karnland et al. (2009).

2. Materials and methods

2.1. LOT parcel A2

The layout of the A2 parcel is shown in Fig. 1. Before swelling the blocks of compacted bentonite had a diameter of 28 cm and a height of 10 cm. The bulk density after hydration was 2000 kg/m³. An electrical heater was contained in a copper tube with a diameter of ~ 10 cm and the lower 2 m of the tube was heated. The bentonite parcel was instrumented for monitoring physical conditions during the experiment and Fig. 1 shows the temperature distribution in the parcel at steady-state heat flow conditions, which were reached during the first year of the experiment. The design of the A2 test is described in detail in Karnland et al. (2009).

2.2. Sampling and sample nomenclature

The chemical and mineralogical characteristics of bentonite from two blocks, 09 and 11, from the heated section and one block, 33, from the cool section of the A2 parcel have been compared with

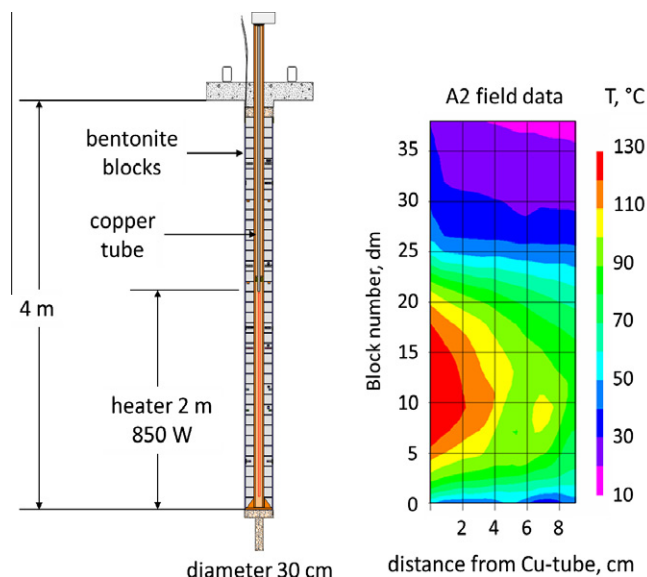


Fig. 1. Lay-out and dimensions of the LOT A2 parcel (left). The temperature distribution in the test parcel at steady state heat-flow conditions (right). Bentonite blocks are numbered from bottom to top. The blocks investigated are 09, 11 and 33.

reference materials consisting of the specific batch of MX80 that was used for fabrication of the individual blocks. The entire volume of blocks 09 and 11 has been heated to temperatures >80 °C, and the innermost 2 cm to ~ 130 °C. Block No. 33 was never exposed to temperatures exceeding 30 °C, apart from the innermost centimetre (Fig. 1). Samples were taken contiguously at five positions along the radius of the blocks. A 1–2-mm thick layer of the contact surfaces bentonite/Cu-tube and bentonite/bedrock was removed before the sampling. The contact between the bentonite and the Cu-tube was sampled separately in the heated blocks by scraping off a 1–2-mm-thick layer from the inner envelope surface of the bentonite block. Individual samples have been labeled with the block number and the radial position, e.g. A2 09 3 means that the sample is from the A2 parcel, block No. 09, radial position 3 (≈ 2 –4 cm from the heater).

2.3. Sample preparation

Both the bulk bentonite and the fraction <2 μm have been analysed when relevant. Silicate analyses were made also on the fraction <0.2 μm . The bulk material was not subject to any pre-treatments prior to the analyses, apart from drying at 60 °C and grinding.

The fine fractions were separated by centrifuged sedimentation after removal of soluble salts by centrifuge-washing with water. Dispersed samples were centrifuged with a centrifugation time/speed calculated by use of the centrifugal form of Stokes' Law to correspond to particle separations at the specified equivalent diameters of 2 μm and 0.2 μm . The supernatants were concentrated by evaporation at 60 °C.

2.4. Silicate chemistry

The chemical composition was determined for the bulk bentonite and for the fractions <2 μm and <0.2 μm by ICP emission spectrometry (AES) at an ISO 9001 certified laboratory, using standard techniques for silicate analysis (fusion with LiBO₂ followed by nitric acid digestion). Total carbon and sulphur were determined by evolved gas analysis by combustion of the samples in a Leco

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