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Microstructural evolution and physico-chemical behavior of compacted clayey soil submitted to an alkaline plume

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Abstract: In the French concept of deep nuclear waste repositories, the galleries should be backfilled with excavated argillite after the site has been filled. Some additives like lime could be used to improve the mechanical characteristics of the argillite. After thousands of years, the degradation of the concrete lining of the galleries will generate an alkaline solution (pH value > 12) that will diffuse through the backfill. This study presents the effect of a saturated $Ca(OH)_2$ solution circulation through lime-treated sample at 60 °C for 3, 6 and 12 months, respectively. The effect of such circulation on the lime-treated Manois argillite (MA) was assessed by petrographical examination coupled to image analysis and scanning electron microscopy (SEM) equipped with energy dispersive X-ray (EDX) analyser of soil pieces. The objective of this study is to make the link among the mineralogical transformations, the textural and mechanical changes produced in the compacted clayey soil as a consequence of the alkaline solution circulation.

Key words: argillite; lime treatment; petrographical examination; scanning electron microscopy (SEM); image analysis; microstructure

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1 Introduction

If the extensive use of smectite-rich bentonite as buffer material in radioactive waste disposal is largely due to the suitable properties of smectites, the interaction of highly alkaline pore water coming from the destabilization of the concrete lining may diminish the bentonite's desirable properties (its physico-chemical buffering). This is why the interaction between smectite and highly alkaline solution has become a major consideration in assessing the performance of bentonite in radioactive waste disposal. In recent years, numerous investigations were conducted in either experimental or modelling fields to evaluate the behaviors of clayey barriers in a deep geological repository for high-level radioactive wastes. To achieve this objective, it is necessary to predict mineralogical changes and transformations that are expected to occur in the barrier during the time required for radioactive wastes to reach non-hazardous radioactivity levels (tens of thousands of years to get the natural background). The alkaline solution can react with the bentonite in the proximity of concrete, inducing dissolution and precipitation of a number of phases. Bulk dissolution experiments were used to study the dissolution behavior of smectite under alkaline conditions [1–7]. Previous work on transport-reactivity systems, involving claystones in contact with hyper-alkaline fluids, predicted that the main reaction processes were dominated by ion exchange in a short time, and by dissolution-precipitation of minerals over a long time [8]. Previous studies of the reactivity of montmorillonite in alkaline solutions were focused on the collapse of expandable smectite layers, in particular on the formation of illite or illite/smectite mixed layers. However, formation of mixed layers is an intermediate step in a series of dissolution-precipitation processes. Elements released from bentonite dissolution react to precipitate several secondary phases as crystalline zeolites and amorphous calcium silicate hydrate (CSH) gels. Recrystallization of beidellite and saponite-like clay minerals was also observed [9, 10]. The alteration of the bentonite

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components neutralizes the basic solutions, and it is clearly a function of the pH value of solution, especially when the pH value is above 11 [4, 10, 11]. If the pH value is below 11, bentonite damage is limited [12]. Such modifications as dissolution and formation of some other non-swelling crystalline phases induce changes in porosity and loss of swelling and sorption capacities. These reactions will influence the porosity evolution as well. Nakayama et al. [13] showed an increase in porosity and permeability of compacted bentonite in contact with a highly alkaline solution (NaOH, a pH value of 14) tank due to montmorillonite dissolution. In the experiments performed by Karnland et al. [14], compacted bentonite was exposed to solutions with high pH values to observe changes in bentonite mineralogy and physical properties. It was pointed out that the transport was strongly affected by chemical reactions between hydroxide ions and bentonite minerals. Interaction between bentonite and hydroxide solutions leads to a significant reduction in the swelling pressure in the bentonite. The reduction seems to be due to an instant osmotic effect and a continuous dissolution of silica minerals, leading to a mass loss, and consequently a decrease in the bentonite density. Yamaguchi et al. [15] focused their experimental efforts on predicting long-term variations in hydraulic conductivity of compacted sand-bentonite mixture. Recently, Suzuki et al. [16] mentioned that smectite in bentonite can be dissolved under hyper-alkaline conditions and promotes the damage of the low hydraulic conductivity. However, data about the relationship between changes in mineralogical composition of bentonite and variations in its porosity, hydraulic conductivity or mechanical properties are scarce. The present paper examines the effect of the alkaline plume on mineralogical evolution and microstructural behavior of compacted MA treated with 4% of lime. Indeed, in the French concept, it is planned to introduce additives (bentonite or lime) in the remoulded argillite to backfill the deep galleries to enhance their geotechnical properties. The lime treatment is currently a common technique in earthworks to improve both the implementation and the long-term behavior of the soils. The basis of lime treatment is firstly cation exchange followed by a reaction with the siliceous components of clay to cause stabilization. The first reaction leads to a rapid flocculation of the clay particles by changing its cohesive nature into a friable and granular structure associated with an improvement of its strength. Stabilization is devoted to long-term additional strength development caused by precipitation of CSH and calcium aluminate gels in response to the dissolution of clays minerals in alkaline environment. Hence, the main objective of this work is the assessment of the relationship between mineralogical transformations and microstructural changes produced in the MA as a consequence of the propagation of the alkaline plume, and the assessment of the effect of lime on the argillite microstructure will be detailed. The characterization of soil structure was conducted by petrographic investigations and numerical characterization of the porous network by image analysis [17, 18]. The main difficulty is to observe clay soils in conditions which conserve the structural characteristics of their natural and hydrated states. An accurate method consists in the impregnation of the material by water-acetone-resin exchange [19]. In the present case, this step was modified since the samples were submitted to a freeze-drying procedure. The impregnation method using a resin with added UV photo-luminescence pigment was adopted to numerically characterize the macroporosity of soils on centimetric to decimetric intact samples [20].

This approach is complementary to the evaluation of the mechanical behavior presented by Cuisinier and Masrouri [21] and aims at investigating the geo-chemical processes and their link with the mechanical processes.

2 Materials and methods

The MA represents the upper part of the Callovo-Oxfordian argillite. After homogenization and being crushed in a very fine grain powder, physico-chemical analysis indicates that the MA contains 26%–32% calcite, 22%–27% rough quartz and 41%–49% clays (fraction lower than 2 μm). Clays are mainly illite, kaolinite and interstratified illite-smectite. The specific surface determined with BET is (40.4 ± 1) m^2/g. The quicklime used in that study was made of more than 97% of pure CaO. A lime content of 4% on a dry-weight basis was selected. The argillite was first wetted and left in an airtight container to reach moisture equilibrium for 2 days. Then, MA and lime were thoroughly mixed. After 24 hours, the mixture was ready for the static compaction. The lime-treated samples were sealed and maintained for 90 days before a given circulation test. The static compaction stress is about 470 kPa, the optimum dry