Impact of alkalinity on the swelling behavior of Opalinus Clay

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1 Introduction

In many countries using nuclear energy, deep geological repositories have been considered for the long term disposal of high level or intermediate level and long-lived (*HL – or IL – LL*) radioactive waste (He et al., 2016). Generally, a multi-barrier system is adopted. It consists of engineered barriers made of compacted bentonite-based materials confined by concrete structures and waste packages embedded in a clay host rock that acts as a geological barrier (Andra. 2016; SKB. 2001), due to its low hydraulic conductivity and high sorption capacity for most radionuclides (Andra, 1999; Andra. 2005). The perturbation due to the excavation of disposal galleries, access shaft and ramps, will generate an excavation damaged zone (*EDZ*) in the surrounding host rock, which can act as preferential pathways for the migration of radionuclides (e.g. Zhang. 2013). It is known that the *EDZ* is able to recover its initial properties over time by self-sealing, thanks to the re-saturation by underground water seepage (Zhang. 2011; Zhang. 2016). However, during the operational phase of the repository, the degradation of massive concrete components, will act as a source of alkaline plumes (Fernandez et al. 2009). The underground water *pH*, initially at 7.5, is expected to increase progressively up to 13.5, further affecting the hydro-mechanical properties of the host rock.

2 EXPERIMENTAL RESULTS

An experimental research program was launched to study the alkalinity impact on swelling properties of the Opalinus clay, considered as a potential host rock in Switzerland. Opalinus clay samples, from the lower sandy facies of Mont Terri site, at an initial nearly dry state ($w = 1.1\%$, $S_r = 25\%$), were exposed to synthetic water ($pH = 7.5$) or to a hyper-alkaline solution ($\text{NaOH}, \text{KOH}, \text{Ca}(\text{OH})_2$; $pH = 13.5$) at different water contents ($w = 1.4\%$, $w = 2.7\%$, $w = 3.3\%$, $w = 4.1\%$). For each water content, the sample's total volume was determined using kerdane following the experimental protocol proposed by Monnier et al. (1973) and adapted to Mont Terri claystones (Matray et al., 2007). It was

found that the void ratio, calculated from the sample's total volume, mass and water content, increases with increasing *pH* (from 0.12 at $w = 1.1\%$ to 0.15 and 0.17 at $w = 4.1\%$ for $pH = 7.5$ and $pH = 13.5$, respectively) (Figure 1).

Fig. 1: Evolution of the void ratio versus the water content for Opalinus clay samples exposed to synthetic water ($pH=7.5$) or to the hyper alkaline solution ($pH=13.5$)

Fig. 2: Evolution of the swelling pressure of Opalinus clay samples exposed to synthetic water ($pH=7.5$) or to the hyper alkaline solution ($pH=13.5$)

In parallel, artificially cracked cylindrical samples of Opalinus clay were subjected to a flow of synthetic water and hyper-alkaline solution in a constant volume cell. The radial swelling pressure was recorded using a total pressure sensor. It has been found that the swelling

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pressure slightly increases with increasing pH (from 0.05 MPa for pH = 7.5 to 0.056 MPa for pH = 13.5) (Figure 2).

The two results presented above show a good agreement. Indeed, exposing Opalinus clay to a hyper alkaline solution triggers several mechanism among which the acid-base reactions concentrated at clay particles edges. An increase in the negatively charged groups occurs resulting in higher repulsive forces (Chavali et al. 2017; Walther. 1996), thus higher porosity (higher void ratio) leading to an increase in the measured swelling pressure. In addition, pH increase triggers calcite and quartz dissolution. This phenomenon can explain the observed increase in total porosity. The latter mechanism was investigated separately by immersing Opalinus clay samples in NaOH, Ca(OH)_2 and KOH alkaline solutions at different pHs (8.5, 11.5 and 13.5). It was found that quartz dissolution was governed by exposure time, while calcite dissolution seemed to depend on both exposure time and pH value, due to the carbonation phenomenon.

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