

Competing effects of volume change and water uptake on the water retention behaviour of a compacted MX-80 bentonite/sand mixture

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ABSTRACT

Bentonite-based materials have been studied as potential barriers for the geological disposal of radioactive waste. In this context, the hydro-mechanical behaviour of the engineered barrier is first characterized by free swelling conditions (as a consequence of the progressive filling of technological gaps) followed by constant volume conditions. This paper presents an experimental study conducted in order to characterize the water retention behaviour of a compacted MX-80 bentonite/sand mixture. The water retention properties upon wetting were investigated under both free swelling and constant volume conditions. In the high suction range, the water content was not influenced by the imposed volume constraints. On the contrary, swelling significantly affected the water retention behaviour at low suctions, and the quantity of water stored was higher under free swelling conditions than it was under prevented swelling. In this case, competing effects between bentonite swelling and water uptake did not lead to an increase of the degree of saturation upon wetting, as it was observed for samples wetted under constant volume conditions. The influence of the very strong hydro-mechanical coupling is further discussed.

Keywords: compacted bentonite/sand mixture; water retention properties; swelling; hydro-mechanical coupling; radioactive waste disposal.

1. INTRODUCTION

In the context of deep geological repositories for nuclear waste, particular attention has been paid to the behaviour of bentonite-based materials in relation to their use as engineered barriers ([Pusch 1992](#); [Komine and Ogata 1994](#); [Dixon et al. 1996](#); [Delage et al. 1998](#); [Collin et al. 2002](#); [Lloret et al. 2003](#); [Romero et al. 2005](#); [Imbert and Villar 2006](#); [Tang et al. 2008a](#); [Baille et al. 2010](#); [Dupray et al. 2011](#); [Villar et al. 2012](#); [Liu et al. 2014](#); among others). The aim is to create a zone of low permeability that is able to limit water flow around the excavated galleries, and thereby delay the release of radionuclides to the biosphere. Therefore bentonite-based materials have generally been selected for their high swelling capacity, their low permeability and their important radionuclides retardation capacities.

Depending on the country, different concepts of disposal, with different sealing materials, have been envisaged (see [Sellin and Leupin 2013](#) for a review). In the French CIGEO concept, bentonite-based materials will be used as backfill materials in access galleries and shafts of the deep geological repository, as well as at the disposal cells dedicated to intermediate level waste ([Labalette et al. 2013](#)).

Initially partially saturated, the bentonite barrier will experience hydration from the saturated host rock. However, placing a buffer within a gallery involves unavoidable technological gaps between the engineered barrier and the host rock. During the saturation process, the periphery of the engineered barrier will therefore first swell under free conditions. During this stage, technological gaps will be progressively filled by bentonite and the effective density of the buffer will decrease. Contact between the bentonite barrier and the host rock will be progressively reached and a swelling pressure will start to develop against the gallery wall. The second stage of the

hydration process is thus characterized by constant volume conditions ([Pusch 1992](#); [Dixon et al. 2002](#); [Rothfuchs et al. 2007](#); [Sellin and Leupin 2013](#)).

In order to study the behaviour of bentonite barriers under *in situ* conditions, the ANDRA has launched, in its underground research laboratory of Bure (France), a series of experiments called PGZ2. Among others, one aim of these *in situ* tests is to characterize the water saturation of bentonite seals under natural conditions ([De La Vaissière 2013](#)). In this experience, the objective is to reach a swelling pressure of 7 MPa, accounting for 12% of technological gaps. The *in situ* minor stress in the underground laboratory being close to 7.5 MPa, the imposed swelling pressure should ensure a good contact between the bentonite buffer and the host rock, while avoiding any risk of fracturing the geological formation ([De La Vaissière et al. 2015](#)).

Because of the evolution of the volume constraints during the hydration process of the bentonite buffer, a good characterization of the material under both free and restricted swelling is required. To date, few experimental studies have investigated both volumetric and water retention behaviours of compacted bentonite with the aim of obtaining suction – degree of saturation relationships ([Delage et al. 1998](#); [Dueck 2008](#); [Ajdari et al. 2013](#)). In this paper, the water retention properties of a compacted bentonite/sand mixture are investigated along wetting paths, under both constant volume and free swelling conditions. New experimental data on the behaviour of the material under both constant volume and free swelling conditions are obtained and should provide a better comprehension of the coupled hydro-mechanical phenomena involved in *in situ* experiments. In particular, attention is given to the competing effects of volume change and water uptake which are observed during hydration under free swelling conditions.

2. MATERIAL AND METHODS

2.1. Material and sample preparation

The material used in the investigation is a mixture of Wyoming MX-80 bentonite (commercial name Gelclay WH2) and quartz sand (commercial name THX1000) with a respective proportion of 7/3 in dry mass. The MX-80 bentonite contains 92% of montmorillonite and other minerals including quartz and feldspars (Tang et al. 2008b). The solid unit weight γ_{sB} of MX-80 bentonite is 27.3 kN/m³ and that of the quartz sand γ_{sS} is 26 kN/m³. Accordingly, the equivalent solid unit weight of the bentonite/sand mixture γ_{sM} is defined as

$$\gamma_{sM} = \left(\frac{f_B}{\gamma_{sB}} + \frac{1 - f_B}{\gamma_{sS}} \right)^{-1} \quad (1)$$

where f_B is the mass fraction of bentonite (equal to 0.7), giving an equivalent solid unit weight of the bentonite/sand mixture equal to 26.9 kN/m³.

In order to facilitate a homogeneous mixture and compaction, the grain size distributions (obtained by dry sieving) are similar for both materials and are in the range of 0.2 to 2 mm.

The initial gravimetric water content of the bentonite/sand mixture (mass of water over solid mass) was 7.08%. Compacted samples were prepared by uniaxial static compaction (57 mm in diameter, 12 mm in height) to a target dry unit weight of 19.6 kN/m³. This dry unit weight was selected in order to reach a swelling pressure of about 7 MPa in the PGZ2 *in situ* experiments performed by the ANDRA (De La Vaissière 2013). Accounting for 12% of technological gaps (Figure 1), the final dry unit weight of the buffer under *in situ* conditions is estimated to 17.5 kN/m³. The

vertical net stress required to reach the target dry unit weight of 19.6 kN/m³ was about 80 MPa.

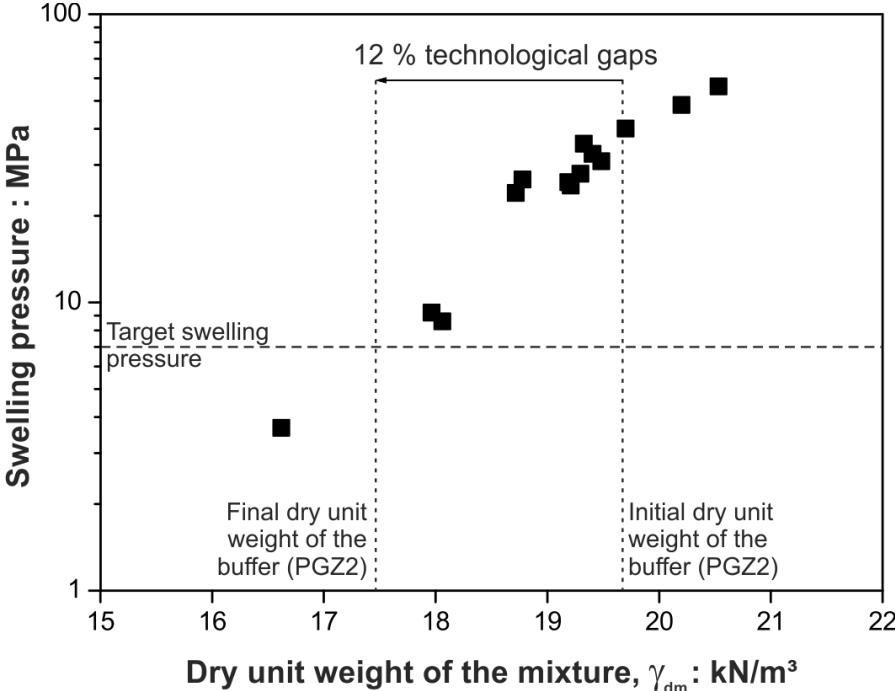


Figure 1. Swelling pressure of the compacted MX-80 bentonite/sand mixture.

2.2. Experimental methods and testing procedure

Water retention curves of the compacted samples were determined under both constant volume and free swelling conditions by using the vapour equilibrium technique. After compaction, the samples were carefully removed from the compaction mould and either transferred to a constant volume cell (having the same diameter as the compaction cell) for the measurement of the water retention curve under isochoric conditions, or placed in a glass Petri dish for the determination of the sorption curve under free swelling conditions. Constant volume cells similar to those used by Villar (2002, 2007) were specially designed for the measurement of the water retention curve under isochoric conditions (Figure 2). Each cell includes a

titanium containment ring and two stainless steel porous discs put on both faces of the compacted sample and allowing vapour exchanges. Two perforated flanges hold the entire assembly.

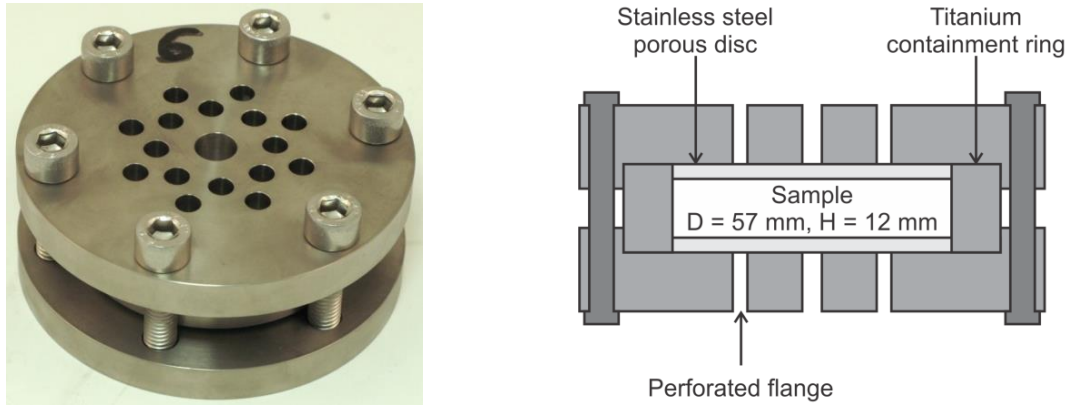


Figure 2. Constant volume cell for water retention curve determination.

All samples were placed in desiccators in which the relative humidity was controlled by using different saturated saline solutions (Table 1). The total suction s imposed to a sample is computed according to Kelvin law

$$s = -\frac{\gamma_w RT}{g M_w} \ln(RH) \quad (2)$$

where γ_w is the water specific weight, R is the ideal gas constant, T is the temperature, M_w is the water molar mass, g is the gravity acceleration and RH is the imposed relative humidity. For the purpose of the experiment, the desiccators were placed within an environmentally controlled room with a temperature of 20°C (+/- 0.2°C).

Salt	Relative humidity (%)	Suction (MPa)
MgCl ₂	33.03	150
K ₂ CO ₃	43.16	113
NaBr	59.14	71
KI	69.86	48
NaCl	75.41	38
KCl	85.13	22
BaCl ₂	90.69	13
K ₂ SO ₄	97.59	3

Table 1. Solutions used and corresponding relative humidity and suction, at 20°C.

Each sample was stabilized under a single relative humidity. In a standard fashion, mechanical and hydraulic equilibria were assumed to occur simultaneously. Equilibrium was detected experimentally by weighing and measuring the sample on a regular basis. Stabilization was assumed when the change in water content with time was such that $\frac{\Delta w}{\Delta t} < (5.10^{-4}) \% / day$. Once this criterion met, the test was stopped. At the end of the test, the mass and the volume of the samples were measured. An important issue is the determination of the volume of samples under free swelling conditions. In the present study, the heights and diameters of the samples were measured with a precision caliper, similarly to [Delage et al. \(1998\)](#), [Olchitzky \(2002\)](#), [Villar \(2002\)](#) and [Tang et al. \(2008a\)](#), among others. Finally, the gravimetric water content was determined by oven drying at 105°C during 24 hours.

3. EXPERIMENTAL RESULTS

Starting from as-compacted conditions, the time required to reach equilibrium was of the order of 30 days for the unconfined samples and 50 days for the confined ones. A longer equalization period was indeed needed for the confined samples due to the limited exchange surface area. On the other hand, the imposed suction appeared to have little impact on the hydration kinetics.

The water retention curves obtained under constant volume and free swelling conditions are shown in [Figure 3](#). The samples submitted to suctions equal or below 113 MPa experienced wetting while the two samples subjected to a suction atmosphere of 150 MPa underwent drying and shrinkage. Therefore the constant volume conditions could not be maintained under this suction.

At high suctions (above around 10 MPa), the water content appears to be relatively independent of the volume constraints. On the contrary, in the lower suction range, bentonite swelling affects the water storage capacity of the material. The water content reached under free swelling conditions at suction of 3 MPa is much higher than it is under prevented swelling (25.3% compared to 14.8%). The experimental results under constant volume conditions enable to identify the air-entry value of the compacted mixture around 10 MPa.

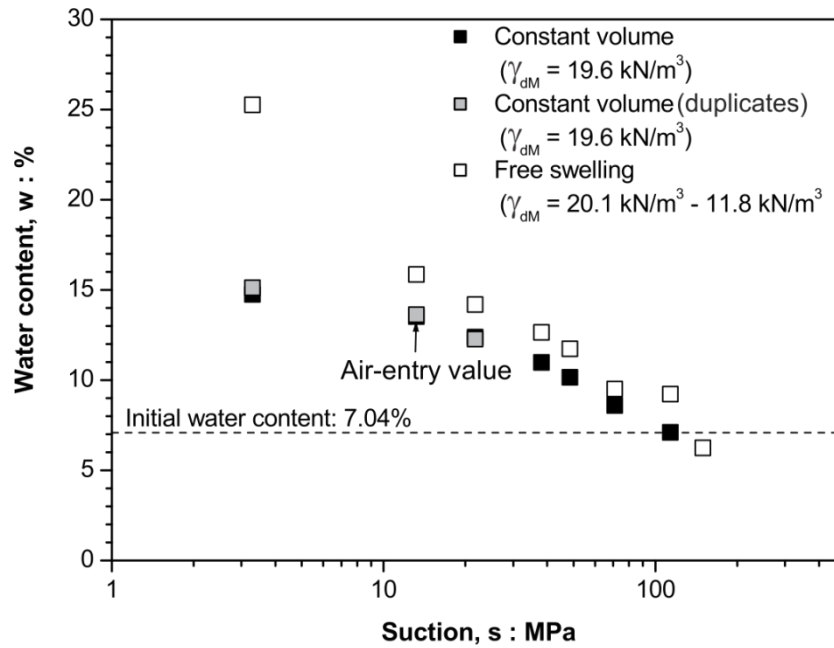


Figure 3. Water retention curves of compacted MX-80 bentonite/sand mixture determined under constant volume and free swelling conditions. Each point corresponds to a different sample.

For the samples under free swelling conditions, the total volumetric, axial and radial strains developed upon suction imposition are presented in Figure 4. An important anisotropy is observed, the axial strain being twice the radial one. This anisotropy is attributed to the uniaxial compaction method used for the preparation of the samples which induces a preferential orientation of the clay particles in the horizontal plane (Denis 1991; Hicher et al. 2000; Tang and Cui 2010; Saba et al. 2014).

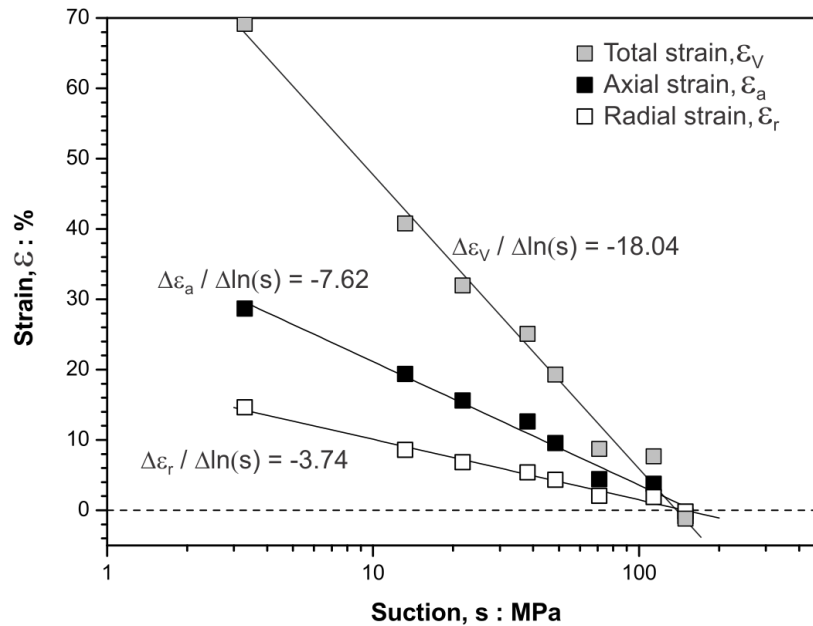


Figure 4. Relationship between the total, axial and radial strains and suction for the unconfined samples.

Finally, duplicates were realized for the samples wetted to suction values of 3 MPa, 13 MPa and 22 MPa under constant volume conditions. An excellent agreement was found between the measured values, as observed in the Figure 3.

4. DISCUSSION

The observations made from Figure 3 are in accordance with previous studies by Romero et al. (1999) and Romero et al. (2011) on compacted Boom Clay, Loiseau et al. (2002) on compacted Kunigel V1/sand mixture, Villar (2002, 2007) on FEBEX bentonite, Agus et al. (2013) on various Calcigel bentonite/sand mixtures, Seiphoori et al. (2014) on granular MX-80 bentonite and Salager et al. (2013) on a compacted clayey soil, among others. It has generally been explained by the complex double structure of compacted clays and its evolution along hydro-mechanical stress paths (see Dieudonne et al. 2014 and Della Vecchia et al. 2015 for a complete review).

Such a bimodal structure has been observed by using mercury intrusion porosimetry on the studied material by Wang et al. (2013) (Figure 5). Even at high density, two pore size families, presumably corresponding to inter-particle and inter-aggregate pores, are identified.

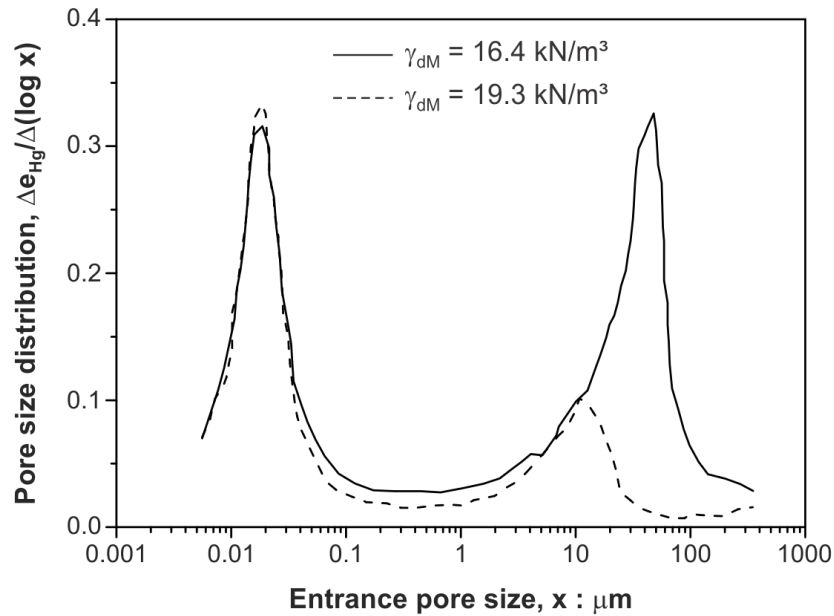


Figure 5. Pore size distribution of the bentonite/sand mixture compacted to dry unit weights of $\gamma_{dM} = 16.4 \text{ kN/m}^3$ and $\gamma_{dM} = 19.3 \text{ kN/m}^3$ and a water content of about 11% (Wang et al. 2013).

In the high suction range, water is believed to be stored essentially by adsorption at the surface of the clay particles. Therefore water uptake is mainly controlled by the physicochemical properties of the clay minerals, in particular smectite, and is independent of the current density. Under confined conditions, highly compacted bentonite can adsorb a limited number of water layers. Cases et al. (1992), Bérend et al. (1995), Yahia-Aissa et al. (2001) and Villar (2007), among others, showed that smectites (particularly those having Ca or Mg as main exchangeable cations) adsorb two water layers for a broad range of relative humidity. More recently, Villar et al.

(2012) observed the development of a third water layer in fully saturated samples of compacted FEBEX and MX-80 bentonites. Under these conditions, hydration leads to a decrease of the inter-aggregate porosity, as a result of aggregate exfoliation. On the other hand, up to four water layers can be adsorbed under free swelling conditions (Saiyouri et al. 1998; Saiyouri et al. 2004). In addition, for smaller suction values, the development of osmotic swelling allows a higher quantity to be stored in the unconfined bentonite samples (Saiyouri et al. 1998; Salles et al. 2009). In this case, hydration is associated to an increase of the intra-aggregate porosity, but also an increase of the total porosity (Figure 4). However further work regarding the evolution of the material structure upon wetting under free swelling conditions is required to better assess the role of the microstructure changes on the overall volumetric evolution.

Knowing the volume of each sample, the water retention curve may be expressed in terms of degree of saturation, which is particularly useful for numerical simulations (Figure 6). As can be observed, the imposed volume constraint significantly impacts the water retention behaviour of compacted bentonite-based materials. For samples wetted under isochoric conditions, hydration led to an increase of the degree of saturation. On the contrary, the decrease of suction did not significantly impact the degree of saturation of the samples wetted under free conditions (in the investigated range of suctions at least).

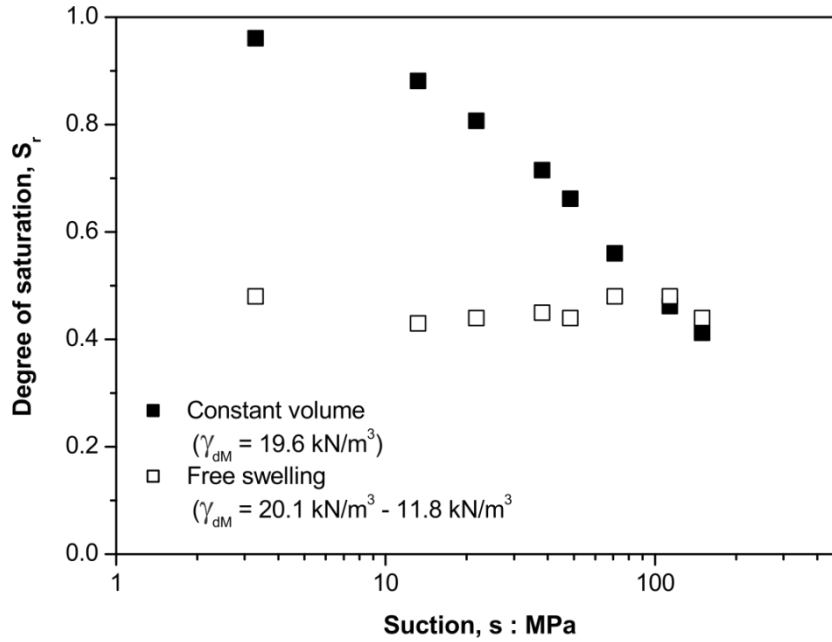


Figure 6. Water retention curves expressed in terms of degree of saturation versus suction and determined under constant volume and free swelling conditions.

This observation is a direct consequence of the important hydro-mechanical coupling in compacted bentonites and can be further explained using the concepts of hydraulic and mechanical wetting (Tarantino 2009). Indeed, the degree of saturation may be expressed as the ratio between the volume of water V_w and the porous volume V_p , or alternatively as the ratio between the water ratio $e_w = (\gamma_{SM}/\gamma_w) w$, and the void ratio e

$$S_r = \frac{V_w}{V_p} = \frac{e_w}{e} \quad (3)$$

This expression evidences the dependence of the degree of saturation on both water ratio and void ratio. Tarantino (2009) introduced the concepts of hydraulic wetting to describe a change in the degree of saturation due to a modified water ratio, and mechanical wetting for mechanical deformation of the porous medium. When bentonite samples are hydrated under constant volume conditions, only the water

ratio is modified and the degree of saturation univocally increases. On the contrary, during hydration under free swelling conditions, swelling strains develop as the material takes water, and both water ratio and void ratio are affected. In this case, hydraulic wetting occurs simultaneously as mechanical drying.

In order to quantify both processes, the evolution of the water ratio upon suction imposition, together with the evolution of the void ratio, are presented in Figure 7a for the free swelling samples. Swelling strains developed upon wetting are particularly important (around 68% when the sample is hydrated to a suction of 3 MPa). In addition, the increase in void ratio upon wetting is more important than the water uptake, i.e. the samples swell more than they take water, hence a relatively constant degree of saturation.

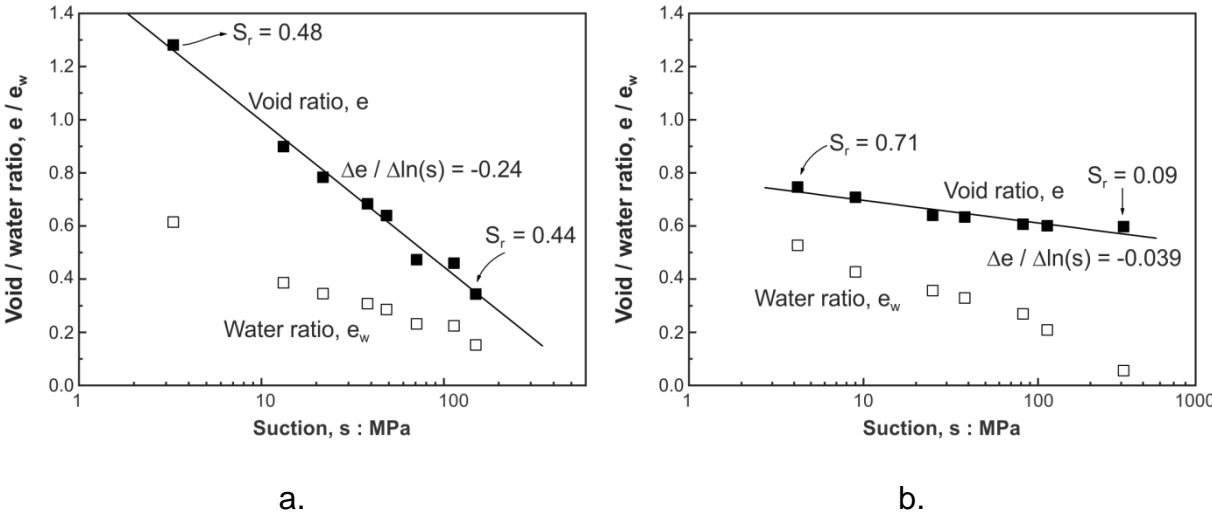


Figure 7. Evolution of void ratio and water ratio with suction under free swelling conditions starting from as-compacted conditions: a. $\gamma_d = 19.6 \text{ kN/m}^3$ (present study) b. $\gamma_d = 16.4 \text{ kN/m}^3$ (Wang et al. 2013).

The same figure can be plotted using the data from Wang et al. (2013). Wang et al. determined the water retention curve of the same mixture but compacted to a lower dry unit weight of 16.4 kN/m^3 (Figure 7b). In the high suction range, the water ratio

follows the same branch as for the investigated mixture, both under constant volume and free swelling conditions. However, as a direct consequence of the lower density of clay particles in the mixture, the material exhibits smaller swelling strains upon wetting. For the sake of comparison, the swelling modulus for changes in suction, $\kappa_s \approx -\Delta e/\Delta(\ln s)$, is around 0.24 for the initial dry unit weight of 19.6 kN/m³ but reduces to 0.039 when the initial dry unit weight decreases to 16.4 kN/m³. As a consequence, the degree of saturation of the mixture studied by [Wang et al. \(2013\)](#) increases upon wetting ([Figure 7b](#)).

5. CONCLUSIONS

Water retention curves under constant volume and free swelling conditions were determined. For high suction values, the wetting branch in terms of water ratio appeared to be void ratio independent. On the contrary, the imposed boundary conditions significantly influenced the water uptake capacity of the bentonite/sand mixture in the lower suction domain. The quantity of water stored under free swelling conditions was much higher than under constant volume.

In terms of degree of saturation, the samples at high initial density wetted under free volume conditions did not exhibit significant changes of degree of saturation. This may be explained by the important competing effects of bentonite swelling and water uptake along wetting paths, as a consequence of strong hydro-mechanical coupling in compacted bentonites. Indeed the high-density samples exhibited important swelling, i.e. mechanical drying when suction decreased.

From a practical point of view, technological gaps between the engineered barrier and the geological formation cannot be avoided in underground structures. These gaps may be observed after the emplacement of the bentonite buffer and affect the water retention behaviour of the barrier (Villar et al. 2005; Mayor and Velasco 2014). Therefore the observed features prove the need for the development of advanced water retention models accounting for the dependency of the water retention properties on the mechanical state of the material, whether it be the current void ratio or the mechanical stress. Classical water retention models (Brooks and Corey 1964; van Genuchten 1980) relating suction to the degree of saturation are not able to represent the observed behaviour. Still they are generally used in numerical modelling and their parameters are determined from constant volume water retention curves. In this case, the predicted degree of saturation of a bentonite plug under *in situ* conditions can be considerably overestimated if technological voids are important and not taken into account in modelling.

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