Water retention properties of two deep Tertiary clay formations within the context of radioactive waste disposal

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Abstract: Belgium investigates the design for disposal of its 'High-Level Radioactive Waste' in two deep clay formations; the Boom clay at Mol, considered the reference host formation, and Ypresian clay at Kallo as the alternative one. The water retention properties of these deep low-porosity formations have been investigated for two main reasons. High suctions develop as a consequence of sample retrieval (at depths between 223 and 350 m), which affect the hydro-mechanical response of these materials, especially at low stress levels. In addition, water retention properties have also been studied to better assess possible desaturation effects due to venting of the disposal facility galleries. After a description of their main properties at intact state and their microstructural features (pore size distributions), the water retention properties of both clays covering a wide suction range and using different complementary techniques are presented and discussed.

Keywords: deep clay formation; water retention properties; microstructure

1 INTRODUCTION

Belgium investigates the design for disposal of its 'High-Level Radioactive Waste' in two deep clay formations. Boom clay is considered the reference host formation and the objective of an extensive research programme. In order to study this clay, located between 160 and 270 m deep, an Underground Research Laboratory at 230 m depth was constructed near Mol (Belgium). On the other hand, Ypresian clay is the alternative host formation that is beginning to be studied to investigate its thermo-hydro-mechanical properties. Two different sites are being considered as potential deposits: Doel and Kallo (near Antwerp, Belgium), where this clay is located between 300 and 450 m deep.

Particular attention has been focused on the water retention properties of these deep low-porosity formations for two main reasons. On one hand, high suctions develop as a consequence of sample retrieval, which affect the hydro-mechanical response of these materials (important swelling have been observed on liquid contact at low stress levels despite being completely saturated). On the other hand and from an operational viewpoint, water retention properties have also been studied to better assess possible desaturation effects due to venting of the disposal facility galleries.

This paper describes the main properties of Boom and Ypresian clays at intact state and their microstructural features obtained by mercury intrusion porosimetry. These clays display quite contrasting microstructural properties, which have important consequences on their hydraulic properties. Specifically, the paper presents the water retention properties, which were obtained using complementary techniques (vapour equilibrium method, chilled-mirror and transistor psychrometers) and discussed within the light of their different microstructural features.

The paper also discusses the different air-entry values obtained when expressing the retention properties in terms of degree of saturation and water content.

2 TESTED MATERIALS AND EXPERIMENTAL PROGRAMME

2.1 *Boom and Ypresian clays*

The Boom clay is the youngest sea sediment, which was deposited during the Rupelian stage (Northwest European Tertiary Basin), 36 to 30 million years ago (Horseman *et al*., 1987). Its mineralogy mainly consists of clay minerals (average 55%) dominated by illite (20-30%), kaolinite (20-30%) and smectite (10 %). The non-clayey fraction is dominated by quartz (25 %) and feldspar (Coll, 2005; ONDRAF/NIRAS, 2005).

The Ypresian clay is also constituted by marine sediments deposited during the Ypresian stage (Eocene series, 54 to 49 million years ago) as reported by ONDRAF/NIRAS (2005). It presents a clay fraction of 45%. Smectite is the dominant phase (25%), followed by illite (15%) and a small amount of kaolinite and chlorite. Non-clayey fraction is constituted predominantly by quartz (45%) and feldspar (5%).

The studied block and borehole samples of Boom and Ypresian clays were retrieved at depths of 223 m and 350 m, respectively. The location of the retrieval sites is shown in Figure 1.

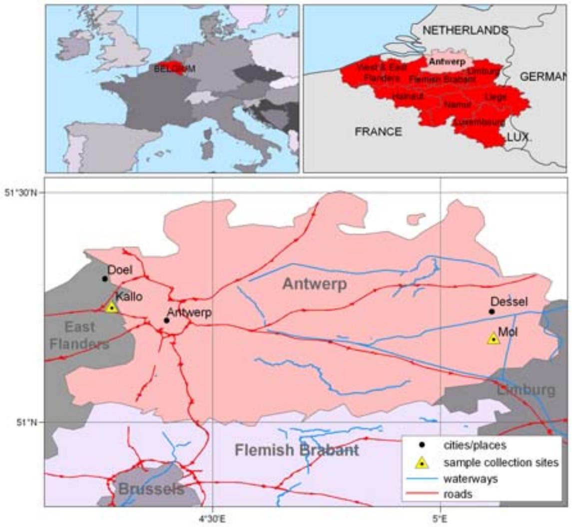

Figure 1. Location of sample retrieval sites.

Table 1 summarises the basic characterisation and the main volumetric and gravimetric properties of the intact materials, which are lightly overconsolidated and saturated stiff clays. As observed in the table, Ypresian clay displays a larger liquid limit, which is consistent with the important smectite content. Another important aspect to highlight, which can be also associated with this higher smectite content, is the larger void ratio of Ypresian clay, despite being formed at a larger depth.

Table 1 also indicates an important initial total suction for the retrieved quasi-saturated soils. In the case of Boom clay, the *in situ* condition at the sampling depth is characterised by a vertical total stress σ_{vi}=4.50 MPa and a water pressure u_{wi}=2.25 MPa (vertical effective stress σ'_{vi}=2.25 MPa). From oedometer tests on undisturbed material (Lima, 2011), it is possible to determine a preconsolidation vertical effective stress σ'^{max}_{v}=5.2 MPa at the end of the deposition process that includes diagenesis and creep effects, resulting in an overconsolidation ratio *OCR*=2.3. Based on a drained friction angle of the natural material ϕ'=20° (Lima, 2011), the earth pressure coefficient at rest K_0 can be estimated as (Della Vecchia *et al*., 2010):

$$K_0^{SC} = (1 - \sin\phi')\sqrt{OCR} \approx 1 \tag{1}$$

The mean effective stress at the *in situ* condition is:

$$p'_i = \frac{1}{3}\left(1 + 2K_0^{SC}\right)\sigma'_{vi} = 2.25MPa \tag{2}$$

On sample retrieval, total mean stress change is:

$$\Delta p = -p_i = -p'_i - u_{wi} \tag{3}$$

Assuming a constant mean effective stress ($\Delta p'$=0), the pore water pressure change on sample retrieval can be estimated as

$$\Delta u_w = \Delta p = -4.5\,MPa \tag{4}$$

$$u_{wf} = u_{wi} + \Delta u_w = -2.25\,MPa \tag{5}$$

where u_{wf} is the final pore water pressure after retrieval, which in principle should be similar to the initial suction measured under laboratory conditions.

In the case of Ypresian clay and following the same reasoning, the vertical stress and pore water pressure under *in situ* conditions are σ_{vi}=7.8 MPa, u_{wi}=4 MPa, σ'_{vi}=3.8 MPa. A vertical effective preconsolidation stress σ'^{max}_{v}=5.50 MPa was determined from oedometer tests (Piña, 2011), which results in an overconsolidation ratio *OCR*=1.4. An earth pressure coefficient at rest $K_0^{SC}\approx 0.85$ was estimated considering ϕ'=16º (Nguyen, 2011) that allowed determining the mean effective stress at the *in situ* condition p'_i=3.4 MPa. The pore water pressure change after sample retrieval can be estimated in Δu_w=-7.4 MPa, which results in a final pore water pressure u_{wf}=-3.4 MPa.

The final pore water pressures induced on stress relief should be compared with the initial suctions reported in Table 1. In the case of Boom clay, a similar initial total suction was measured under laboratory conditions using psychrometers. For Ypresian clay, despite being retrieved at a larger depth, the measured initial total suction is somewhat lower (1.9 MPa). This fact is a consequence of the lower air-entry value of this clay, which will be discussed in the next section. This means that for Ypresian clay the final water pressure that was calculated based on stress relief considerations (u_{wf}=-3.4 MPa), cannot be sustained under saturated conditions (refer to Table 1, in which a somewhat lower degree of saturation is detected for the retrieved condition).

Table 1. Main properties of Boom and Ypresian clays.

Property	Boom clay	Ypresian clay
Density, ρ (Mg/m^3)	around 2.05	1.97 to 2.02
Dry density, ρ_d (Mg/m^3)	1.65 to 1.67	1.55 to 1.60
Gravimetric water content, *w (%)*	around 25	26 to 26.6
Initial total suction, Ψ *(MPa)*	2.0-2.5	1.9
Density of soil solids, ρ_s (Mg/m^3)	2.67	2.76
Void ratio, *e*	0.60 to 0.62	0.72 to 0.78
Porosity, *n*	0.37 to 0.38	0.42 to 0.44
Degree of saturation, *Sr (%)*	around 100	94 to 98
Liquid limit, w_L *(%)*	56	142 to 158
Plastic limit, w_p *(%)*	29	26
Plasticity index, *PI (%)*	27	116 to 132
Vertical water permeability, $k_{w\,v}$ *(m/s)*	2.4 x 10^{-12}	6.6 x 10^{-12}

These high initial total suctions have important consequences on the hydro-mechanical response of these materials when subjected to stress levels below the *in situ* mean effective stress. Consider as an example Figure 2, which shows a stress-controlled isotropic loading –far below the *in situ* mean stress– on Boom clay under drained conditions. As observed in the figure, the clay underwent an important expansion despite being isotropically loaded and starting from an initial saturated state (Table 1). This expansion is a clear consequence of suction reduction effects due to water contact at low stress levels. To minimise these effects, the materials were always loaded to a total mean stress equivalent to the geostatic condition and only then put in contact with water under atmospheric pressure to restore *in situ* effective stress.

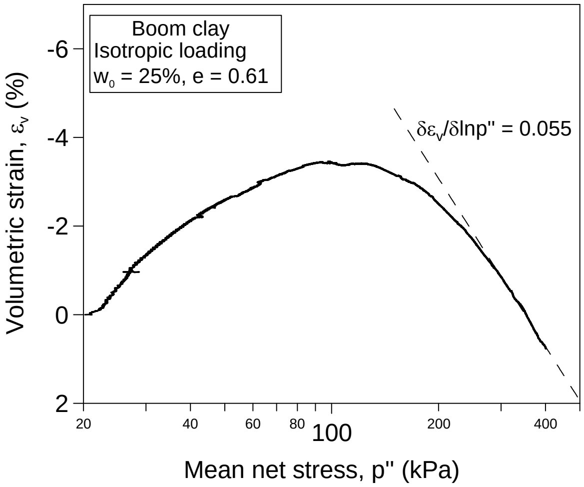

Figure 2. Isotropic loading on retrieved Boom clay under drained conditions. Sample put in contact with water at low stress levels.

Mercury intrusion porosimetry tests were performed using an 'AutoPore IV 9500 – Micromeritics Instrument Corp.' porosimeter to characterise the porosity network on freeze-dried samples –to preserve the pore structure–. Figure 3 presents the pore size distribution PSD curves of both clays. In the case of Boom clay, the plot shows one dominant pore mode at around 90 nm, as expected for a matrix type microstructure. On the contrary, Ypresian clay presents two dominant pore modes, one at 700 nm and another at 80 nm. This structural feature that has been consistently observed at different depths (Piña, 2011) is not typical for a deep clay formation, which has undergone a depositional sequence similar to Boom clay. Besides void ratio effects, this dual porosity has notable effects on the hydraulic response of Ypresian clay. On one hand, larger water permeability values are expected on Ypresian clay as indicated in Table 1, not only as a consequence of its higher void ratio but also due to its macroporosity features. On the other hand, water retention properties will be also affected, as discussed in the next section.

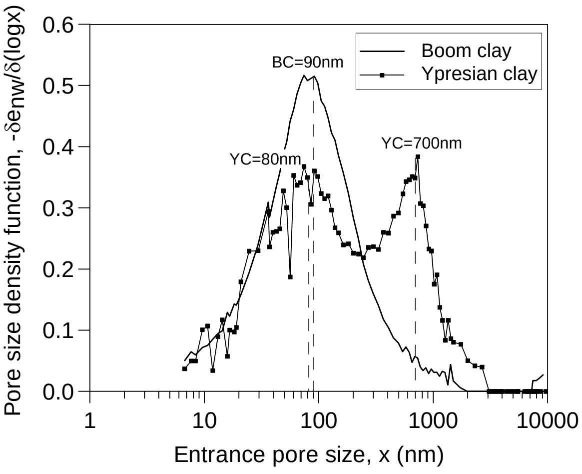

Figure 3. Pore size distribution curves of Boom and Ypresian clays.

2.2 *Experimental programme*

Laboratory tests were carried out on undisturbed material, which were trimmed (15 mm in diameter and 12 mm high) from retrieved Boom clay blocks and Ypresian clay borehole samples.

Water retention properties under unstressed conditions were first determined using chilled-mirror dew-point (WP4, Decagon Devices Inc, USA) and transistor (SMI, Australia) psychrometers. Details on the working principle of both psychrometers, as well as the different calibrations carried out, have been extensively described in Cardoso *et al*. (2007). The tests using WP4 were carried out following a multi-stage procedure, in which the same sample was first subjected to a main drying path and then to a wetting one. Only water content changes were measured along these multi-stage paths to avoid solid mass loss. During drying, samples were let to evaporate for one hour at controlled relative humidity (around 40%) and then allowed for equilibrating for one day under hermetic conditions and before taking the readings with the psychrometer. The total suction measuring time was around 10 minutes and water contents were determined using the initial and final masses (the sample holder is not completely tight to vapour losses). Wetting paths were performed by adding small drops to the sample. An equalisation period of one day under hermetic conditions was afterwards performed and before the determination of the total suction.

Boom clay results were complemented with vapour equilibrium technique using samples installed in hermetic jars. Partially saturated aqueous solutions of NaCl were used to apply different relative humidity values (Romero, 1999) below a total suction of 38 MPa. In the upper range, a saturated solution of $NaBr.2H_2O$ was also used to apply a total suction of 75 MPa (Delage *et al*., 1998; Romero, 2001). Multi-stage drying and subsequent wetting paths at the following steps 5, 10, 20, 38 and 75 MPa were carried out. At specific intervals of the equalisation process the mass of the sample was registered.

3 EXPERIMENTAL RESULTS

Figure 4 presents the time evolution of the changes in soil mass for Boom clay along the different wetting steps using vapour transfer under pure diffusion. As observed in the figure, longer equalisation periods are required for the lower total suction changes. Equalisation is reached after three weeks for the total suction step 10 to 5 MPa, whereas it is completed in less than one week for the total suction step 75 to 38 MPa. Similar results were reported by Merchán *et al*. (2011) on reconstituted Boom clay samples. These authors contrasted their experimental results using numerical simulations to study these longer periods required, in which a consistent trend was also obtained in the simulated results. Total suction equalisation depends on the vapour density (or mass fraction of vapour in gas) gradient, the exchange properties between the evaporating surface and the chamber atmosphere, and the material cross sectional area available for vapour to flow –volume of air to soil total volume, which is larger at the driest condition–.

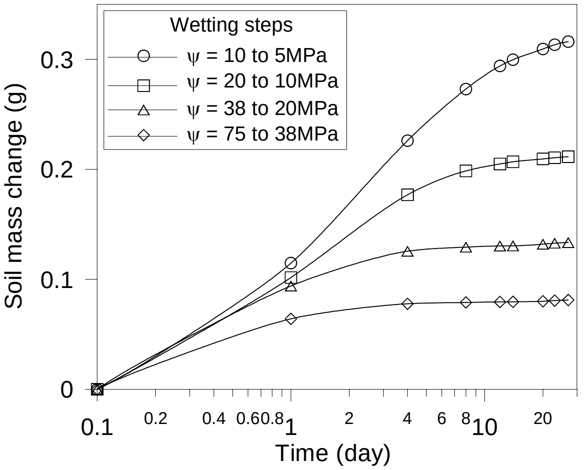

Figure 4. Time evolution of changes in Boom clay mass along wetting steps.

Figure 5 summarises the water retention results obtained on Boom clay. Generally speaking, a good consistency in the results using different techniques is observed. Nevertheless, vapour equilibrium and transistor psychrometer results systematically occurred below WP4 psychrometer readings. The systematic higher total suctions measured with WP4 psychrometer for given water contents can be explained in terms of the hydraulic paths undergone by the soil during the measurement process. As discussed by Cardoso *et al.* (2007), the sample placed inside the equalisation chamber of the WP4 psychrometer undergoes some drying along the suction measuring period, which can explain these systematic higher values.

Besides hydraulic hysteresis, volume change effects are also affecting the differences observed between drying and wetting paths. The low-suction zone of the retention curve (below 10 MPa) is dependent on void ratio and is consequently sensitive to the stress paths followed (Romero & Vaunat, 2000).

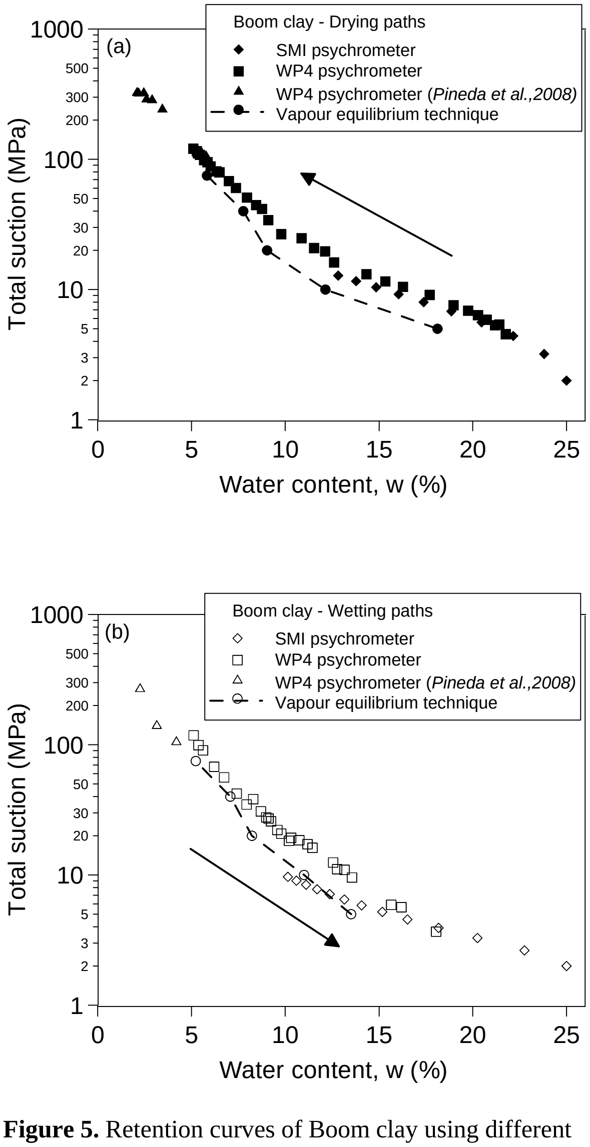

Figure 5. Retention curves of Boom clay using different techniques. (a) Drying paths and (b) wetting paths.

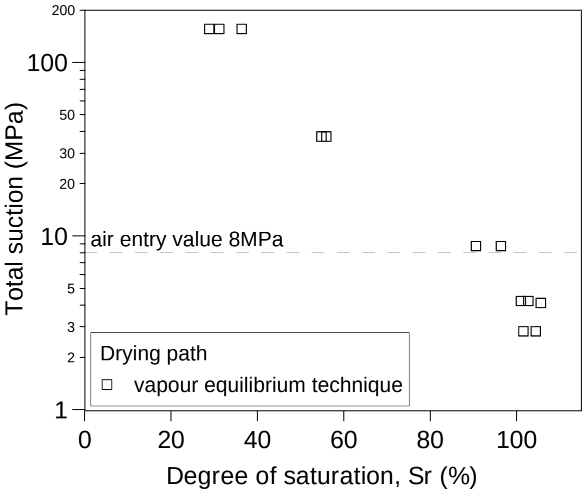

Figure 6. Retention curves of Boom clay in terms of degree of saturation using vapour equilibrium technique (Le *et al*., 2008).

As observed in Figure 5a and with reference to psychrometer readings on drying, important changes in water content occur when total suction is above 4 MPa, indicating some kind of air-entry value for Boom clay in terms of water content. Le *et al.* (2008) studied the water retention curve of undisturbed Boom clay in terms of degree of saturation. In their study, volume changes were measured along the hydraulic paths, displaying the results plotted in Figure 6. As observed in this figure, relatively high total suctions could be maintained without inducing appreciable desaturation on the material, suggesting a larger air-entry value in terms of degree of saturation. This aspect is a consequence of the shrinkage undergone by the material on drying. Despite losing water, the material also undergoes volume reduction, and a relatively high degree of saturation is still maintained at around 8 MPa (refer to Fig. 6).

Figure 7 shows the water retention curve of Ypresian clay compared to Boom clay obtained from WP4 psychrometer readings. As indicated in Table 1, the initial water content of Ypresian clay is slightly larger, which is also reflected at the starting point –water storage capacity at saturation– of the drying curve (Fig. 7a). Ypresian clay also presents a larger water retention capacity in both wetting and drying paths, as a consequence of the dominant smectite phase of the clay fraction. It also appears that Ypresian clay loses slightly larger water content (from the water storage capacity at saturation) when applying a total suction of 8 MPa (Fig. 7a). As also observed in this figure, at a total suction around 3 MPa, an early desaturation process seems also to occur on Ypresian clay. This desaturation process is associated with the double porosity network detected on Ypresian clay and its effects on air-entry value (Fig. 3). To better interpret the consequences of this double porosity structure, Figure 8 plots the water retention curves estimated from mercury intrusion results and in terms of water content. In this case, the drying paths are assimilated to the non-wetting mercury intrusion (Romero & Simms, 2008). Boom clay displays an air-entry value consistent with the one estimated in terms of water content (Fig. 5a). Nevertheless, Ypresian clay presents a much lower air-entry value compared to Boom clay, as a consequence of the developed macroporosity. The air-entry value for Ypresian clay obtained from MIP and associated with macropores is around 0.4 MPa (early desaturations were also observed at a total suction around 3 MPa, as shown in Fig. 7a).

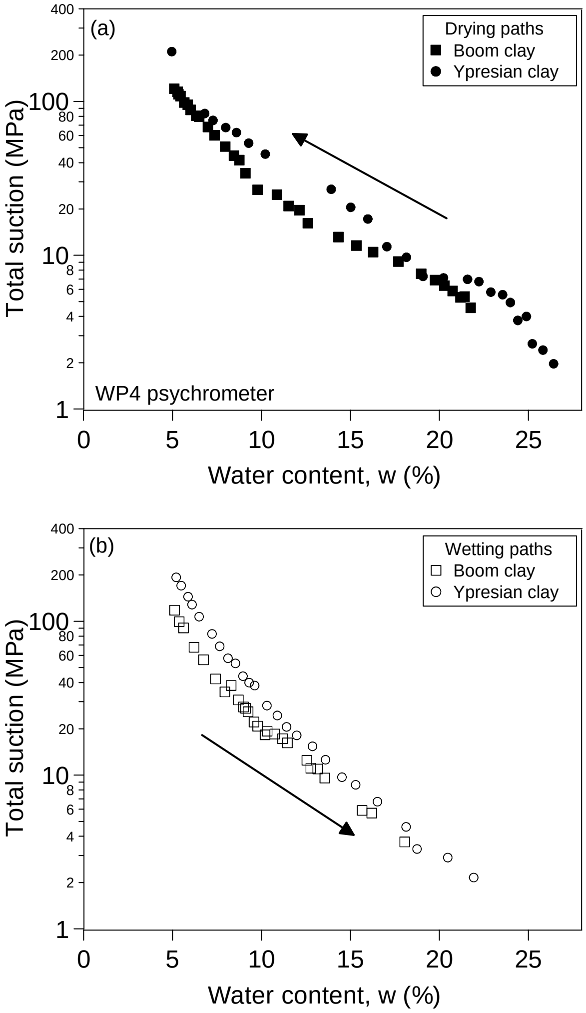

Figure 7. Comparison between water retention curves of Ypresian and Boom clays obtained from WP4 psychrometer. (a) Drying paths and (b) wetting paths.

The lower air-entry value of Ypresian clay affects the maximum total suction measured of the retrieved material. The estimated pore water pressure after retrieval is u_{wf}=-3.4 MPa (refer to section 2.1), which is higher (in suction terms) than the predicted air-entry suction and cannot be sustained under saturated conditions. The estimated degree of saturation corresponding to a total suction of 3.4 MPa is around 90% (lower than the degree of saturation reported in Table 1). The initial total suction reported in Table 1 of 1.9 MPa is consistent with the degree of saturation range indicated in Table 1. In the case of Boom clay, since the estimated water pressure after retrieval is -2.25 MPa (lower in suction terms than the estimated air-entry value), then this total suction can be sustained and the fully saturated condition maintained.

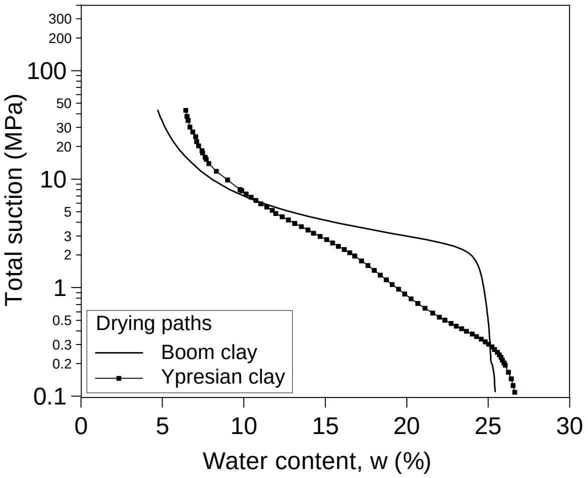

Figure 8. Water retention curves estimate from MIP data, comparison between Boom and Ypresian clays.

4 CONCLUSIONS

Two deep clay formations from Belgium, Boom clay (160 to 270 m deep) and Ypresian clay (300 to 450 m deep), are being investigated as host formations for disposal of high-level radioactive waste. The initial total suction is of importance when interpreting the state of the material after retrieval, which remains under nearly saturated conditions. These high total suctions affect the hydro-mechanical response of the materials, particularly at low stress levels (on water contact, the materials undergo swelling despite starting from nearly saturated state).

One important aspect to highlight is that both clays, which share a common depositional sequence, display quite different microstructural features. Boom clay displays a matrix type microstructure with dominant pore mode at around 90 nm. On the contrary, Ypresian clay presents two dominant pore modes, one at 700 nm and another at 80 nm, which have important consequences on the hydraulic response of the material.

The retention properties have been studied using psychrometer and vapour equilibrium techniques.

The consistent results on Boom clay show an air-entry value around 4 MPa when expressing water retention properties in terms of water content and slightly larger (around 8 MPa) when considering the volumetric variable degree of saturation. The difference between both values has been explained in terms of the shrinkage undergone by the material on drying, in which a relatively high degree of saturation is still maintained at around 8 MPa.

The macroporosity developed on Ypresian clay induces a lower air-entry value, despite being retrieved at a larger depth. The estimated pore water pressure after retrieval (-3.4 MPa) is higher (in suction terms) than the predicted air-entry suction and cannot be sustained under saturated conditions (early desaturations were observed at a total suction around 3 MPa, as shown in Fig. 7a). A total suction of 1.9 MPa is finally measured on Ypresian clay after retrieval (below the early desaturation suction of 3 MPa).

5 ACKNOWLEDGEMENTS

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