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Chemical impact on the hydro-mechanical behaviour of high-density FEBEX bentonite

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ABSTRACT

The effect of the salinity of the saturating fluid on the hydro-mechanical properties of the FEBEX compacted bentonite was investigated by means of swelling, compressibility and permeability tests in which deionised water and solutions of different concentrations and compositions were used as saturating fluids. The solutions were chosen to simulate natural and extreme conditions in a high-level radioactive waste repository excavated in crystalline or clay host rocks. The swelling capacity of the bentonite decreases with the increase in salinity of the pore water, although this difference becomes less patent for high vertical loads and high densities and when the salinity of the solution is very low. The samples saturated with solutions containing high concentration of ions are also less deformable and consolidate more rapidly than the samples saturated with low-salinity solutions. The hydraulic conductivity of the highly compacted saturated FEBEX bentonite increases when high-salinity permeants are used, especially for low densities and when the stress level is low. The influence of the composition of the solution on the hydro-mechanical properties of the bentonite was also checked.

1. Introduction

The investigation reported in this paper was carried out in the context of research projects related to the disposal of high-level radioactive wastes (HLW) in deep geological repositories. A system of natural and artificial barriers will be constructed to isolate the wastes from the host rock and the biosphere. The host rocks that are most widely considered, especially in Europe, are granites, clays and salts. As part of the engineered barrier of a HLW repository, bentonite – a clay material consisting mainly of smectite – will be used as sealing material in most disposal concepts and will be placed around the waste containers in contact with the host rock. Bentonite was chosen as sealing material because of its low permeability, swelling capacity and retention properties, among other features. Its role is to delay the arrival of groundwater to the waste canister by the combined effect of low permeability and swelling capacity, the latter favouring the sealing of preferential pathways; to protect the canister against mechanical damage; and to retard the migration of radionuclides, once the canister fails, by retention processes. In a deep geological repository the type, salinity and geochemical composition of the water reaching the bentonite barrier will depend on the type of host rock, the interaction with other elements of the repository (e.g. concrete) and the modifications induced by the bentonite itself.

It is known that the behaviour of clayey materials is strongly dependent on the physico-chemical interactions between clay particles and pore fluid chemistry. Smectites are phyllosilicates made up of piled laminae with exchangeable cations and layers of water between them. The piling of clay laminae forms primary particles. The thickness of the primary particles varies depending on the hydration state. When two particles are located close to each other, their electric double layers (DDL) interact, inducing a repulsion whose magnitude is partly conditioned by the chemistry of the pore water. According to the DDL theories, the thickness of the DDL decreases as the saline concentration of the water in the pores increases. Consequently, for a given porosity, the effective pore size of the clay would increase with increasing concentration of the solution, with the corresponding increase in permeability. In turn, the swelling capacity of the clay increases with the thickness of the DDL.

Since the most important geotechnical properties to be taken into account to asses the correct performance of the bentonite barrier of a HLW repository are its hydraulic conductivity, swelling ability and compressibility, this paper presents results of experimental studies on the effect of changes in pore water composition on the hydro-mechanical properties of a sealing material statically compacted to high-density, the Spanish FEBEX bentonite. The research was carried out in the framework of FEBEX (Full-scale

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Engineered Barriers Experiment in Crystalline Host Rock), a project for the study of the near field for a HLW repository in crystalline rock according to the Spanish concept: the waste canisters are placed horizontally in drifts and surrounded by a clay barrier constructed from highly compacted bentonite blocks (ENRESA, 2000, 2006a). The tests were performed in two laboratories, at UPC (Universitat Politècnica de Catalunya) and CIEMAT (Centro de Investigaciones Energéticas, Mediambientales y Tecnológicas), and a wide range of salinities, as well as several representative dry densities, were tested. The range of salinities expected for the groundwater of granitic and clay host rocks were broadly covered, by taking into account not only the initial salinities of the groundwater, but also the progressive incorporation of ionic species as the groundwater migrates through the barrier and the possible concentration increases. Fig. 1 illustrates the local increase in the salinity of the pore water of the barrier caused by the processes associated to water advection: dissolution of soluble species of the bentonite in the more hydrated areas, transport with the water front, and local increases in concentration as the hydration front moves. The Figure shows the analyses performed in samples taken after dismantling of the FEBEX *in situ* test at the Grimsel Test Site (Switzerland). This test simulated the conditions of the bentonite barrier of a HLW repository at real scale and under natural conditions. The gallery was excavated in granite and the waste container was mimicked by a heater whose surface temperature was kept at 100 °C. The test was dismantled after five years operation (ENRESA, 2006a,b).

With respect to the groundwater salinities, for the crystalline rocks from the Forsmark area of the Fennoscandian shield, Arcos et al. (2006) report average salinities of 0.9% (0.28 M) and extreme values of 7.3% (2.1 M), whereas the Spanish reference water for the clay host rock concept (Turrero and Peña, 2003) has a salinity of 1.1% (0.24 M).

2. Material

The FEBEX bentonite used in this investigation (ENRESA, 2006a) was obtained from the Cortijo de Archidona deposit, Almería, SE Spain. The material presents a content of montmorillonite higher than 90%, the remaining minerals being small amounts of quartz, plagioclase, cristobalite, K-feldspars, trydimite and calcite. The material has a liquid limit of 102 ± 4%, a plastic limit of 53 ± 3%, a

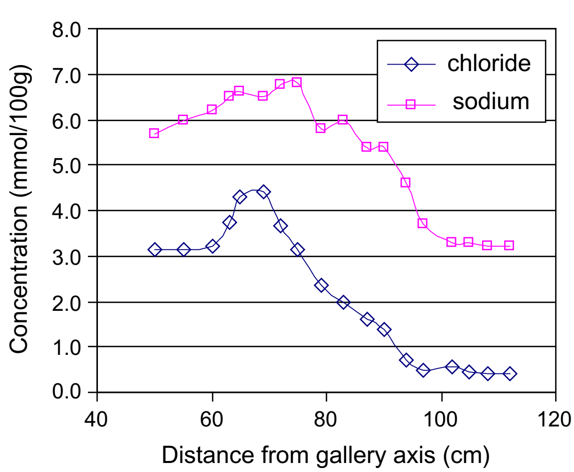

Fig. 1. Chloride and sodium concentration measured in aqueous extracts from samples taken along a vertical section of the bentonite barrier after 5 years of operation of the FEBEX *in situ* test (the heater diameter – placed concentrically with the gallery and simulating the waste container – was 49 cm) (modified from ENRESA, 2006a,b).

specific gravity for the soil particles of 2.70 ± 0.04 and a total specific surface area of 725 m^2/g. The hygroscopic water content in equilibrium with the laboratory atmosphere is 13.7 ± 1.3%. The cation exchange capacity (CEC) varies from 96 to 102 meq/100 g, and the major exchangeable cations are Ca (35–42 meq/100 g), Mg (31–32 meq/100 g), Na (24–27 meq/100 g) and K (2–3 meq/100 g).

The saturated permeability to deionised water (k_w, m/s) of FEBEX bentonite samples that were compacted at different dry densities, is exponentially related to dry density (ρ_d, g/cm^3). The values of permeability to deionised water for dry densities around 1.6 g/cm^3 are in the order of 10^{-14} m/s. A distinction may be made between two different empirical fittings depending on the density interval (Villar, 2002): for dry densities of less than 1.47 g/cm^3:

$$\log k_w = -6.00\rho_d - 4.09 \qquad (1)$$

for dry densities in excess of 1.47 g/cm^3:

$$\log k_w = -2.96\rho_d - 8.57 \qquad (2)$$

The variation in the experimental values with respect to these fittings is smaller for low densities than it is for higher values, with an average – in relative values – of 30%.

The swelling pressure (P_s, MPa) of FEBEX samples compacted with their hygroscopic water content and flooded with deionised water up to saturation can be related to dry density (ρ_d, g/cm^3) through the following equation (Villar, 2002):

$$\ln P_s = 6.77\rho_d - 9.07 \qquad (3)$$

In this case, the difference between the experimental values and the fitting is 25% on average. This dispersion, which is wider for higher dry densities, is due to both the natural variability of bentonite and the measurement method used, which does not allow high degrees of accuracy.

The tests were performed with the FEBEX bentonite compacted to high densities and saturated with deionised water or with solutions at room temperature. The solutions used were classified in low and high-salinity solutions. The first ones represent conditions expected in a HLW repository in granitic rocks, whereas the second ones represent extreme conditions and were chosen because they allow a more systematic study of the effect of salinity.

Two low-salinity solutions were used, termed as saline and granitic (Table 1). The granitic solution simulates the conditions of the outer part of the barrier, and the saline water simulates the situation in the internal part of the barrier, where water is loaded with ionic species from the bentonite, dissolved during water inward movement. The granitic water is a commercial, rather diluted water (0.005 M, 0.02% salinity) from a Spanish granitic massif. The saline water used has a chemical composition similar to that of the bentonite interstitial water (Fernández and Cuevas, 1998), but simplified to include only the major elements. Its salinity is 0.76% (0.22 M), the main ions being chloride and sodium.

NaCl and $CaCl_2$ solutions of different concentrations, always higher than 0.1 M, were used as high-salinity solutions. The osmo-

Table 1
Chemical composition of the low-salinity solutions.

Element (mmol/L)	Granitic	Saline
Cl^-	0.37	100
SO_4^{2-}	0.15	15
HCO_3^-	2.36	
Mg^{2+}	0.39	15
Ca^{2+}	1.12	10
Na^+	0.48	80
K^+	0.026	
pH	8.3	7
M	0.005	0.22
Salinity (%)	0.02	0.76

Table 2
Osmotic suction of some of the solutions used as measured by psychrometric techniques (in MPa).

NaCl 0.5 M	NaCl 2.0 M	NaCl 5.5 M	$CaCl_2$ 0.5 M	$CaCl_2$ 2.0 M	$CaCl_2$ 5.5 M
2.2	10	33	3.2	18	54

tic pressures corresponding to some of the solutions were measured using psychrometric techniques (Romero, 1999); they are indicated in Table 2.

The low-salinity solutions were used in the tests performed at CIEMAT laboratories (Madrid, Spain), whereas the high-salinity solutions were used in the tests performed at UPC (Barcelona, Spain).

3. Methods

3.1. Swelling pressure

3.1.1. Tests with low-salinity solutions

The swelling pressure tests performed at CIEMAT were carried out in oedometer frames and conventional oedometer cells, in which the surface of the sample was reduced in order to counteract the high forces expected. The initial height of the specimens was 12.0 mm and their cross-section was 998 or 1140 mm^2. The samples were confined in rings preventing them from deforming laterally, and between two porous stones at the upper and lower surfaces.

The samples were prepared by means of uniaxial compaction of the clay directly in the oedometer rings. Specimens of different initial dry densities were obtained by varying the compaction pressure. In all the cases the clay was used with its hygroscopic water content.

The sample contained in the oedometer ring is placed inside the oedometer cell and the lower porous stone is covered with the granitic water whose chemical composition is shown in Table 1, so that the sample begins to saturate from the bottom upwards, allowing the air in the pores to escape. The displacement recorded by a dial gauge as the sample saturates is observed, and swelling of the sample is prevented by the application of loads. Ideally the reading of the dial gauge shall not drift excessively from the initial value (within an uncertainty of ±0.005 mm), thus preventing both the swelling and the consolidation of the sample. The test is considered to be completed when, under a constant vertical load, no strain within the resolution of the gauge is observed for at least 24 h. The duration of the tests was dependent on the dry density of the samples, with an average experimental duration of 4 days.

Once the sample is removed from the oedometer, its water content is checked by oven-drying at 110 °C during 24 h.

3.1.2. Tests with high-salinity solutions

The effect of water salinity on swelling pressure was investigated at UPC by measuring the swelling pressure of bentonite that was saturated under isochoric conditions using deionised water and solutions of NaCl (0.1, 2.5, 5.5, 6.3 M) and $CaCl_2$ (4.2 M). Initially, the bentonite was compacted at a dry density of 1.65 g/cm^3 at hygroscopic conditions.

Some tests were performed in a conventional oedometer, in which the lever arm was fixed to prevent the swelling of the clay and the swelling force developed upon flooding was measured by means of a load cell (Lloret et al., 2003). Other tests were performed in the small isochoric cells shown in Fig. 2 (Hoffmann et al., 2007). In this case, an overpressure of 0.2 MPa was applied to the water input to reduce the duration of the tests and a calibrated strain gauge was used to measure the swelling pressure. In all the tests, the thickness of the specimens was 10.5 mm and the diameter was 50.0 mm.

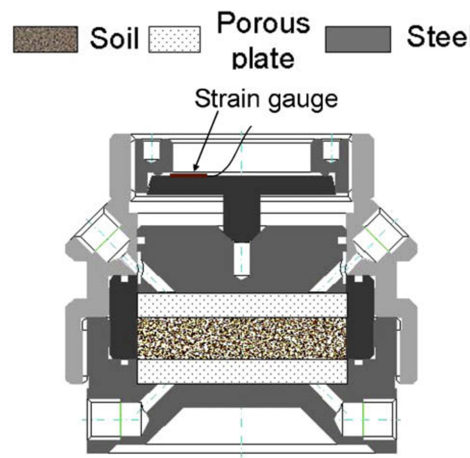

Fig. 2. Schematic representation of the isochoric cell designed by CIMNE to determine swelling pressure.

3.2. Swelling capacity and compressibility

The influence of the salinity of the saturating solutions on the swelling capacity of the FEBEX bentonite was checked by means of swelling under load tests performed in standard oedometers, whereas the compressibility was analysed by means of consolidation tests performed after the swelling stage of some of the swelling under load tests performed with high-salinity solutions.

3.2.1. Tests with low-salinity solutions

The swelling capacity of the bentonite was determined by CIEMAT on samples compacted at dry densities of 1.50, 1.60 and 1.70 g/cm^3 with its hygroscopic water content (around 14%), saturated in oedometers under different vertical loads. The material was compacted inside the cell ring using static uniaxial compaction. The average vertical stresses applied to obtain specimens of 36–38 mm diameter and 12 mm height were 13, 19 and 37 MPa for the nominal dry densities of 1.5, 1.6 and 1.7 g/cm^3, respectively. Once in the oedometer, vertical pressures of 0.1, 0.5, 1.0, 1.5, 2.0 or 3.0 MPa were applied to the samples. Immediately afterwards, the samples were flooded at atmospheric pressure from the bottom porous plate. Deionised, saline and granitic waters (Table 1) were used as saturation liquid. The swelling strain experienced by the specimens upon saturation was recorded as a function of time until stabilisation. The water content of the specimens was determined at the end of the tests.

3.2.2. Tests with high-salinity solutions

The high-salinity tests were performed at UPC with samples statically compacted to a dry density of 1.65 Mg/m^3 at their hygroscopic water content (13.7%). Compaction was carried out in the oedometer ring at a vertical displacement rate of 0.2 mm/min and at a maximum vertical stress of 23.0 MPa. Specimens were 10.5 mm high and 50 mm in diameter. Once in the oedometers, the samples were gradually loaded at constant water content up to the vertical stress of the flooding stages (0.02, 0.5 or 2.0 MPa). Since the load applied was lower than the compaction stress, the samples remained in over-consolidated state. Afterwards, the specimens were soaked under load using either distilled water or NaCl and $CaCl_2$ solutions with concentrations of 0.5, 2.0 and 5.5 M. In the soaking stages, the time necessary to reach the stabilization of vertical displacements was about 5 days, but the whole stage was extended to two weeks.

To analyse the compressibility of the bentonite in some of the tests, after the soaking stage, the saturated bentonite was gradually loaded up to 2.0 MPa under oedometer conditions. The unloading stage was performed in two steps. The followed stress paths are presented in Fig. 3.

The evolution of the sample deformation over time was interpreted using a non-linear curve-fitting algorithm, in order to determine the parameters used in the consolidation analysis by following Terzaghi's theory (Romero, 1999). The dial displacement of the oedometer d under an effective stress increment can be theoretically expressed as

$$d = d_0 + \frac{2h}{E_m}\delta(\sigma_v - u_w)\overline{U}(t, C_v) + 2hC_\alpha \log(t/t_{90}) \quad (4)$$

where d_0 is the initial compression, mainly due to the equipment deformability, $2h$ the mean thickness of the specimen, E_m the drained constrained modulus of elasticity, $\overline{U}(t,C_v)$ the average degree of consolidation, which is a function of time t and of the coefficient of consolidation C_v (Lambe and Whitman, 1979; Das, 1983), C_α the coefficient of secondary consolidation and t_{90} the time required for 90% of average consolidation.

3.3. Permeability

3.3.1. Tests with low-salinity solutions

The method for determination of the saturated hydraulic conductivity consisted on the measurement, as a function of time, of the water volume that passes through the saturated specimen, while a constant hydraulic gradient was maintained between top and bottom (Villar and Lloret, 2001). The specimen was confined in a cylindrical rigid cell that prevented any change of the clay volume. The swelling of the saturated clay against the cell wall guaranteed a perfect contact between clay and cell, consequently avoiding a preferential pathway. The cells were made of stainless steel and had an inner section of 1963 mm^2 and a height of 25 mm. Porous stones were placed at the top and bottom of the specimen, which was saturated by injecting water through the porous stones at a pressure of 0.6 MPa. Once the sample saturated, the injection pressure in the lower part of the cell was increased, and the water outflow through the upper outlet of the cell was measured with an automatic volume change device. The permeability coefficient was calculated applying directly the Darcy's law. Hydraulic gradients between 3600 and 24,400 were applied for the determinations with granitic water, and between 400 and 18,000 for the determinations with saline water, depending on the dry density of the clay. It was demonstrated that, for this range of hydraulic gradients, the hydraulic conductivity value obtained is independent of the hydraulic gradient applied (Villar, 2002).

The determinations were performed using deionised or granitic water as saturating fluids and permeants (Table 1).

3.3.2. Tests with high-salinity solutions

The hydraulic conductivity of the bentonite when high-salinity solutions are used was obtained indirectly from the backanalysis of the vertical strain evolution measured during the compression stages performed in oedometer conditions after the saturation of the bentonite, following the methodology described in Section 3.2.2. Once the modulus of elasticity and the consolidation coefficient are known, the permeability can be calculated, since it is related to both (Lambe and Whitman, 1979).

4. Results

4.1. Swelling pressure

4.1.1. Tests with low-salinity solutions

The determination of swelling pressure was performed at CIEMAT on specimens manufactured from the clay with its hygroscopic water content, compacted at various densities and using granitic water to saturate the sample (Villar, 2002). The results obtained are shown in Fig. 4, which also includes the empirical fitting of Eq. (3) obtained for samples saturated with deionised water. Taking into account the dispersion of the data obtained for this parameter, which is 25% on average (see Section 2), it may be concluded that the values obtained using granitic water

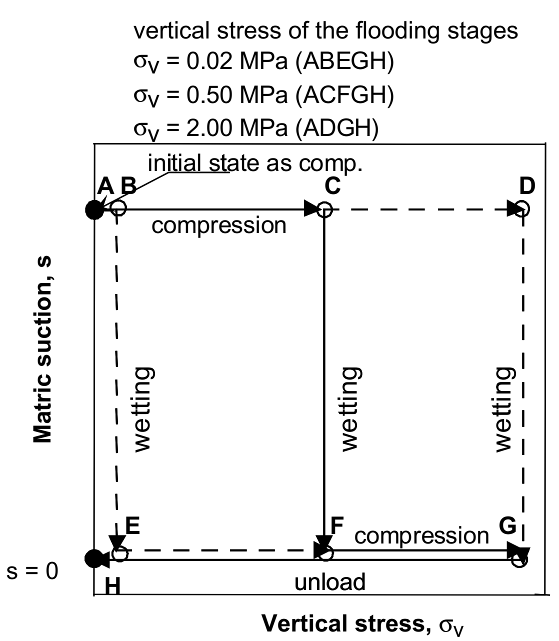

Fig. 3. Stress paths followed under oedometer conditions in the swelling and compressibility tests performed by UPC.

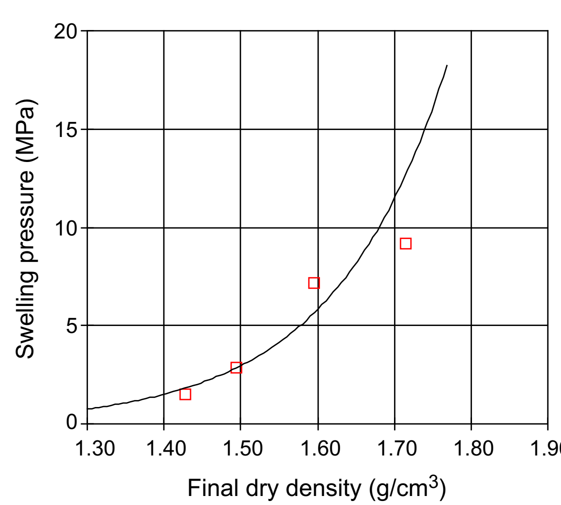

Fig. 4. Swelling pressure obtained for clay saturated with deionised (line, Eq. (3)) and granitic water (symbols).

Table 3
Summary of swelling pressure tests using NaCl and $CaCl_2$ solutions for bentonite compacted at ρ_d 1.65 g/cm^3.

Test	Cell	Solution	Liquid injection pressure (MPa)	Time to reach P_s (days)	Swelling pressure (MPa)
1	Conventional oedometer	Distilled water	0.0	15	4.5
2	Isochoric cell	Distilled water	0.2	7	4.4
3	Isochoric cell	Distilled water	0.2	8	4.4
4	Conventional oedometer	0.1 M NaCl	0.0	13	4.0
5	Conventional oedometer	2.5 M NaCl	0.0	4	2.9
6	Conventional oedometer	5.5 M NaCl	0.0	2	2.6
7	Isochoric cell	6.3 M NaCl	0.2	2	2.4
8	Isochoric cell	4.2 M $CaCl_2$	0.2	5	2.8

do not differ much from those that would have been obtained for the same densities with deionised water.

4.1.2. Tests with high-salinity solutions

The effect of high water salinity on swelling pressure was investigated by measuring the swelling pressure of bentonite that was compacted to a nominal dry density of 1.65 g/cm^3 and saturated under isochoric conditions using solutions of NaCl and $CaCl_2$ of different concentrations. Table 3 shows a summary of the tests performed and the results obtained. Fig. 5 shows the evolution over time of the swelling pressure measured in some tests. It can be seen that an increase in salt concentration reduces the swelling pressure. The reduction of the time necessary to reach the final swelling pressure when the concentration of NaCl increases may be attributed to an increase in the permeability with salinity.

4.2. Swelling capacity

4.2.1. Tests with low-salinity solutions

The swelling tests with low-salinity solutions were performed with samples compacted to nominal dry densities of 1.5, 1.6 and 1.7 g/cm^3 saturated under different vertical loads from 0.1 to 3.0 MPa, using either deionised, saline or granitic water. The strains reached at the end of some of the tests are plotted as a function of the vertical pressure in Fig. 6. The influence of the initial dry density of the bentonite and of the load applied during saturation is clear: under a given vertical load the samples of higher initial dry

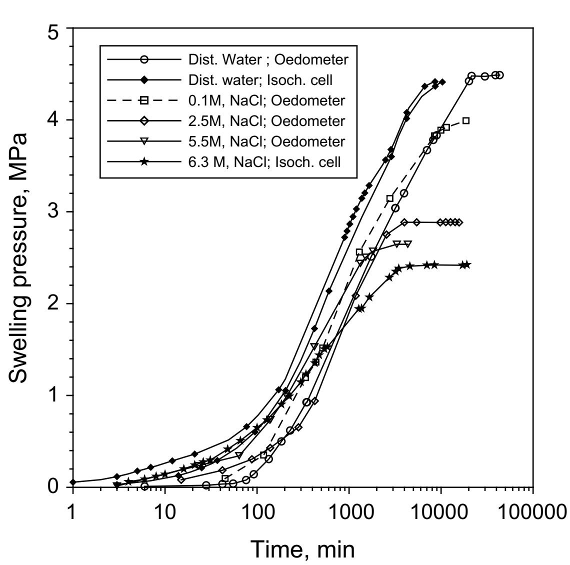

Fig. 5. Evolution of swelling pressures measured using different solutions (ρ_d 1.65 g/cm^3).

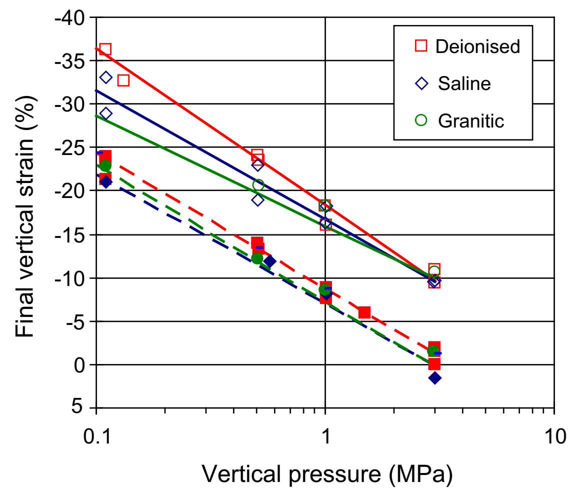

Fig. 6. Final strain of samples saturated under different vertical pressures with different solutions (filled symbols: dry density of 1.50 g/cm^3, open symbols: dry density of 1.70 g/cm^3).

density swell more, whereas swelling is hindered by the application of high vertical loads. The samples with a dry density of 1.5 g/cm^3 and saturated under a vertical load of 3 MPa – which is close to its swelling pressure, according to Eq. (3) – experienced compression when the vertical load was applied and small collapses at the beginning of the saturation. The average duration of the tests, which is related to the time needed for stabilisation of the swelling strain, was 18 days for the tests performed with deionised water, 13 days for the tests performed with granitic water and 11 days for the tests performed with saline water. These values are also a qualitative indication of the permeability of the samples, which would be higher for those saturated with saline water.

Taking into account all the results, the following fittings between final swelling strain (ε, %) and vertical pressure (σ, MPa) as a function of the initial dry density (ρ_d, g/cm^3) were found and are also plotted in the figure (Lloret et al., 2004).

For deionised water:

$$\varepsilon = (5.40\rho_d - 1.32)\ln\sigma + (-48.25\rho_d + 63.69) \quad (5)$$

For saline water:

$$\varepsilon = 6.47\ln\sigma + (-48.71\rho_d + 66.13) \quad (6)$$

For granitic water:

$$\varepsilon = (-6.49\rho_d + 16.53)\ln\sigma + (-43.59\rho_d + 58.14) \quad (7)$$

A slightly higher swelling capacity was observed in samples saturated with deionised water (14% on average for samples saturated with saline water), but there is no distinct behaviour between the samples saturated with the low-salinity solutions.

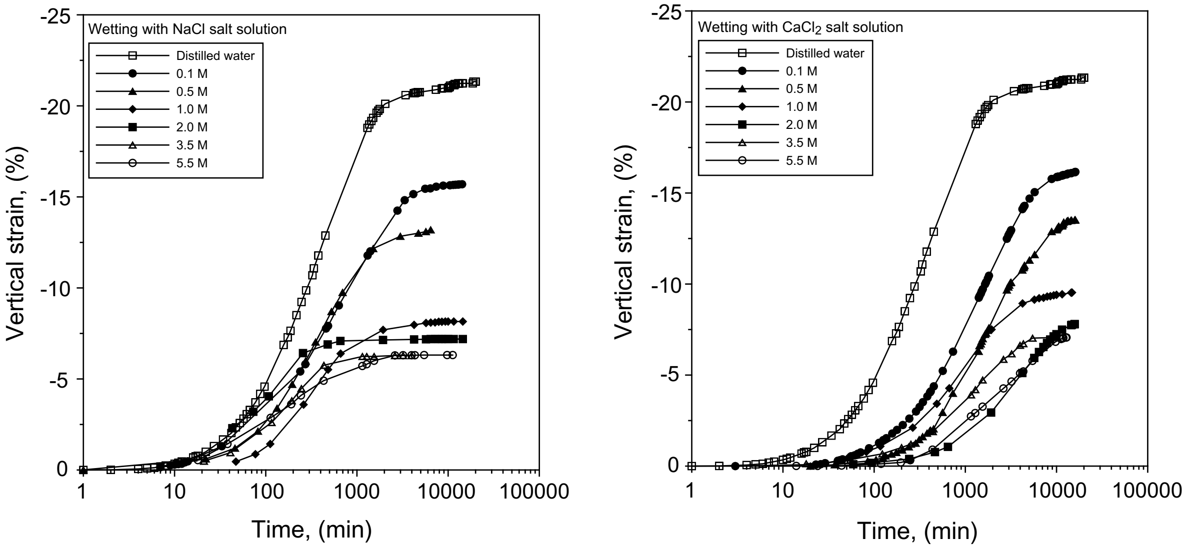

Fig. 7. Evolution of swelling strains during wetting under a vertical load of 0.5 MPa with different solutions (initial ρ_d 1.65 g/cm^3).

4.2.2. Tests with high-salinity solutions

In order to evaluate the effect of high-salinity on swelling strains, swelling tests were performed in conventional oedometers, in which bentonite compacted to a dry density of 1.65 g/cm^3 was soaked under different vertical loads using different types of solutions (Castellanos et al., 2006).

A high variety of salinities was used in the tests performed under a vertical load of 0.5 MPa. The loading up to 0.5 MPa was performed stepwise over two days (steps of 0.02, 0.05, 0.1 and 0.5 MPa) keeping the hygroscopic water content. During this loading stage, the volume changes were very small, since the load applied was lower than the compaction stress (about 23 MPa) and consequently the samples remained in over-consolidated state. The time necessary to reach the stabilisation of vertical displacements during wetting was about 5 days, but this stage was extended to two weeks. The evolution of volumetric strains during wetting is presented in Fig. 7 for vertical stresses of 0.5 MPa. When the vertical stress applied during wetting was 2 MPa, the samples experienced an initial collapse eventually followed by the swelling of the microstructure. Fig. 8 shows the values of the final swelling strains. The values computed with Eqs. (4)–(6) (deionised, saline and granitic water, respectively) have also been included. It can be seen that the swelling capacity decreases significantly with salinity and that the samples wetted with $CaCl_2$ solutions swell slightly more than those wetted with NaCl. Also, the swelling strain value obtained with Eq. (5) for a sample with an initial dry density of 1.65 g/cm^3 saturated with a saline solution of chemical composition equivalent to that of the interstitial water of the bentonite (saline water of Table 1), is somewhat higher than that measured for similar samples saturated with NaCl or $CaCl_2$ solutions of equivalent concentration (0.22 M).

The effect of vertical load on the final swelling strain is shown in Fig. 9. When high vertical stresses were applied (2 MPa), all the samples experienced an initial collapse associated with the rearrangement of the macrostructure, that was recovered afterwards. The swelling capacity decreases significantly with salinity, but the differences in the swelling strains tend to be smaller when the vertical stress increases. In particular, the swelling strains of the bentonite compacted to a dry density of 1.65 g/cm^3 decreased to almost half its initial value when 2 M $CaCl_2$ and NaCl solutions were used as saturating fluids. For higher concentrations the swelling capacity barely changed.

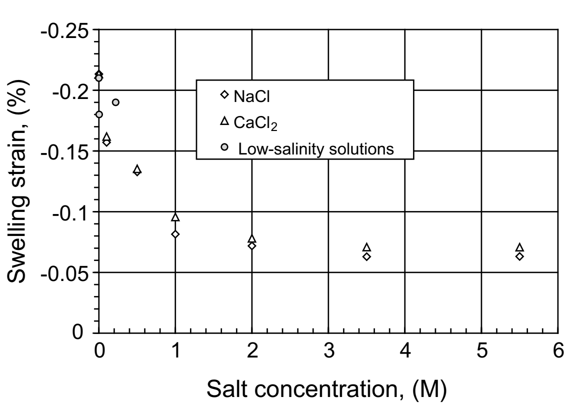

Fig. 8. Variation of swelling strains due to wetting with different solutions under a vertical stress of 0.5 MPa (initial dry density 1.65 g/cm^3) (modified from Lloret and Villar, 2007).

4.3. Compressibility

Once the bentonite was saturated in the swelling tests performed under a vertical load of 0.5 MPa and described in the previous section, it was loaded under oedometric conditions up to 2 MPa, in order to evaluate the bentonite stiffness and permeability. After the saturation stage, the vertical load was increased up to 1 and then 2 MPa. The unloading stage was performed in two steps. The evolution over time of vertical settlements measured after the application on the saturated samples of a vertical load of 2 MPa is shown in Fig. 10 for samples saturated with NaCl solutions. Similar trends were obtained for samples saturated with $CaCl_2$ solutions. Despite the dispersion of the results, it is possible to observe that the samples saturated with solutions containing high concentration of ions are less deformable and consolidate more rapidly than the samples saturated with low-salinity solutions. The higher compressibility of samples saturated with low-salinity solutions could be also a consequence of the higher porosity they reach after saturation. Fig. 11 shows the results of the soaking under load 0.5 MPa and compression stages in terms of final void ratios.

The parameters of the Terzaghi's 1-D consolidation model C_v and E_m obtained by backanalysis of the temporal evolution of set-

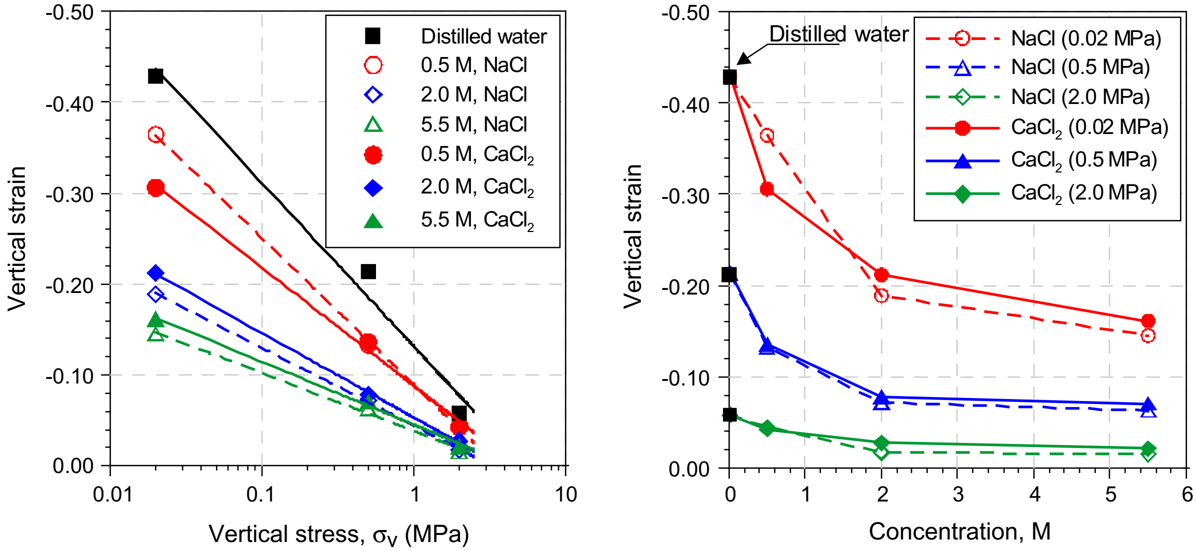

Fig. 9. Final swelling strains of FEBEX bentonite compacted at dry density 1.65 g/cm^3 due to wetting with different solutions under the different vertical stresses indicated in the legend (Castellanos et al., 2006).

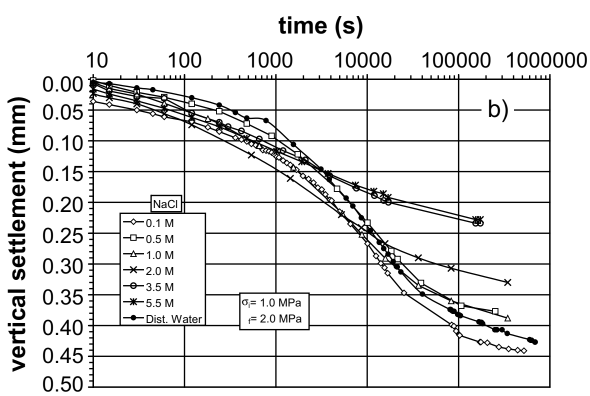

Fig. 10. Evolution of vertical settlements due to loading under oedometer conditions of samples saturated with solutions of NaCl of different concentrations. Loading from 1.0 to 2.0 MPa.

tlements are given in Fig. 12. The increase in the coefficient of consolidation (C_v) and the drained constrained stiffness (E_m) with increasing solute concentrations is clearly observed. This increase cannot be attributed only to solute effects, in fact, the different structure (void ratio) of the samples after saturation plays a significant role in the value of these parameters. No clear variation of C_v with the salinity of the solution was observed.

4.4. Permeability

4.4.1. Tests with low-salinity solutions

The saturated permeability of specimens of different density compacted with hygroscopic water content was determined using granitic or saline water as permeating agent (Villar, 2002). The results obtained are shown in Fig. 13 along with the fittings obtained for deionised water (Eqs. (1) and (2)). No clear trend is observed concerning the variation of the values obtained with granitic water with respect to those obtained with deionised water. The values obtained with saline water are, however, 184% higher on average

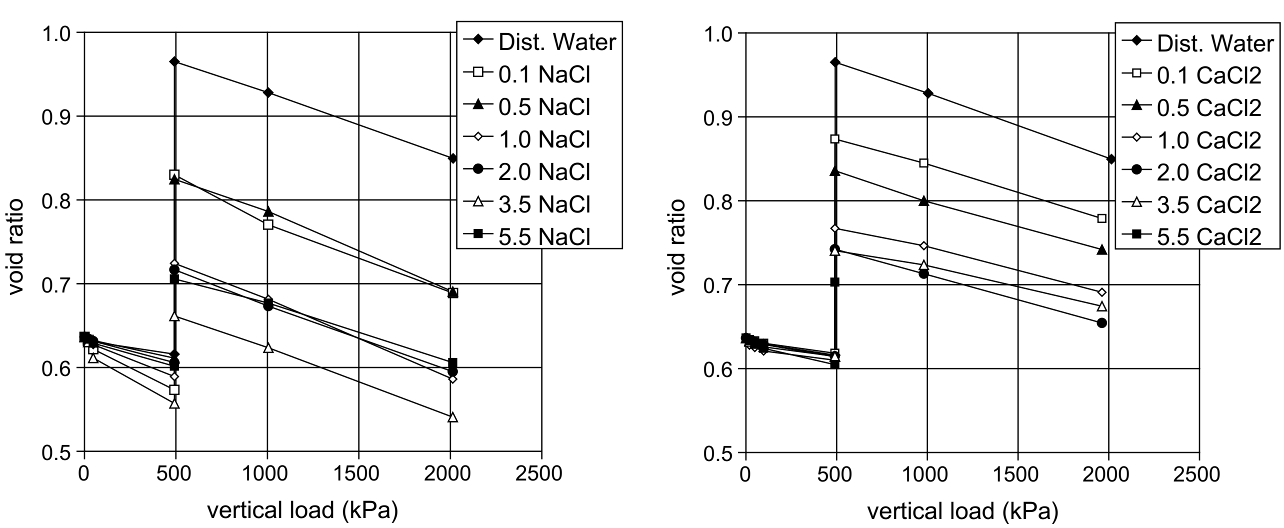

Fig. 11. Changes in void ratio during soaking under load 0.5 MPa and compression up to 2 MPa using different solutions (initial ρ_d 1.65 g/cm^3).

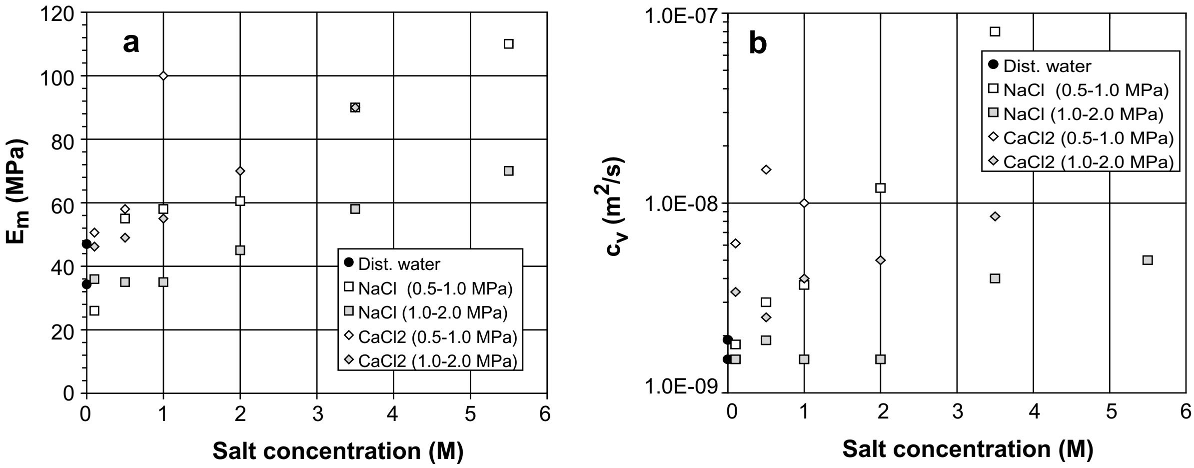

Fig. 12. Variation with salinity of parameters obtained from oedometer tests on samples saturated with different solutions (Lloret et al., 2004).

than those expected for a sample of the same density tested with deionised water, and in addition, they show greater dispersion. This higher permeability to saline water with respect to that expected for deionised water is more pronounced for low densities.

4.4.2. Tests with high-salinity solutions

The water permeability of the samples tested in the oedometers described in the previous section was calculated by analysing the evolution of vertical strains after loading under saturated conditions, using the classical 1-D consolidation theory. Fig. 14 presents the values obtained as a function of bentonite void ratio for high-salinity permeants, where it can be seen that the permeability increases when the concentration of the saline solutions increases, especially for high values of saline concentration. For a given void ratio, the permeability to NaCl solutions is higher than that for $CaCl_2$ solutions of the same concentration.

5. Discussion

Although it is generally accepted that the swelling pressure decreases with the saline content of the solution, the influence of the salinity of the solution on the value of the swelling pressure has been considered negligible or small for montmorillonite compacted at high-density, especially in the case of Ca-montmorillonite (Pusch, 1994). Karnland et al. (2005) measured reductions of the swelling pressure of MX-80 bentonite (a Na-montmorillonite) with the salinity of the saturating fluid, which were relatively lower when the clay density was higher. Sugita et al. (2003) observed the same in Kunigel V1 bentonite. The swelling pressure of less expandable materials (like Friedland Ton clay, with 45% smectite) is insensitive even to high salt content at relatively high clay densities (Pusch, 2001). However, if the salinity of the solution is very high, the swelling decrease may be noticeable even for high bentonite densities. In fact, the tests presented here on FEBEX bentonite show that the swelling pressure of the bentonite compacted to a dry density of 1.65 g/cm^3 decreases to almost half its initial value when 2 M $CaCl_2$ and NaCl solutions are used as saturating fluids. For higher concentrations, the swelling capacity barely changes. This reduction could be related to an increase in osmotic suction. However, it shall be mentioned that the osmotic suction associated with the solution 6.3 M NaCl and with the solution 4.2 M $CaCl_2$ is equal to 40 MPa in both cases (see Section 2), but the reduction of swelling pressure is bigger when NaCl is used as a solute. For low salinites (0.004 M, granitic water), the swelling pressure of the bentonite seems not to be affected in the dry density range from 1.4 to 1.7 g/cm^3.

Bentonite develops slightly lower swelling strains upon saturation with low-concentrated solutions (up to 0.8% salinity) than with deionised water. The reduction of the swelling strains for higher salinities is clearer and can be explained by considering that the NaCl or $CaCl_2$ solutions cause an increase in electrolyte concentration near the clay particle surfaces, diminishing the thickness of the double layer and the swelling potential. Also, samples wetted with $CaCl_2$ solutions swell slightly more than those wetted with NaCl with the same molarity.

The swelling curves shown in Fig. 7 indicate that the samples saturated with low-salinity solutions, develop a quick initial primary swelling as well as a subsequent slower secondary swelling, whereas the samples saturated with high-salinity solutions display a smaller primary swelling and a small to inexistent secondary swelling. Rao et al. (2006) consider that the primary swelling of expandable clays is linked to the rate of matric suction dissipation (which is conditioned by the size of the clay pores), whereas secondary swell is controlled by diffusion of salts and adsorption–desorption reactions. In the case of high-salinity solutions, the diffusion of salts from the saturating solution into the clay particles

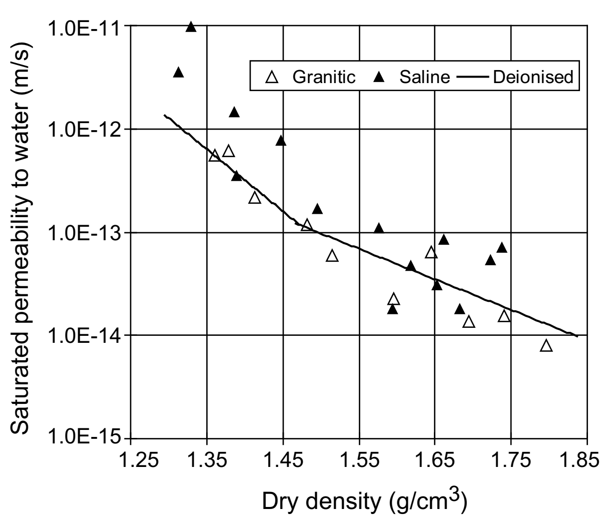

Fig. 13. Hydraulic conductivity for granitic or saline water versus the dry density of the clay and fittings obtained for deionised water (Eqs. (1) and (2)) (Lloret and Villar, 2007).

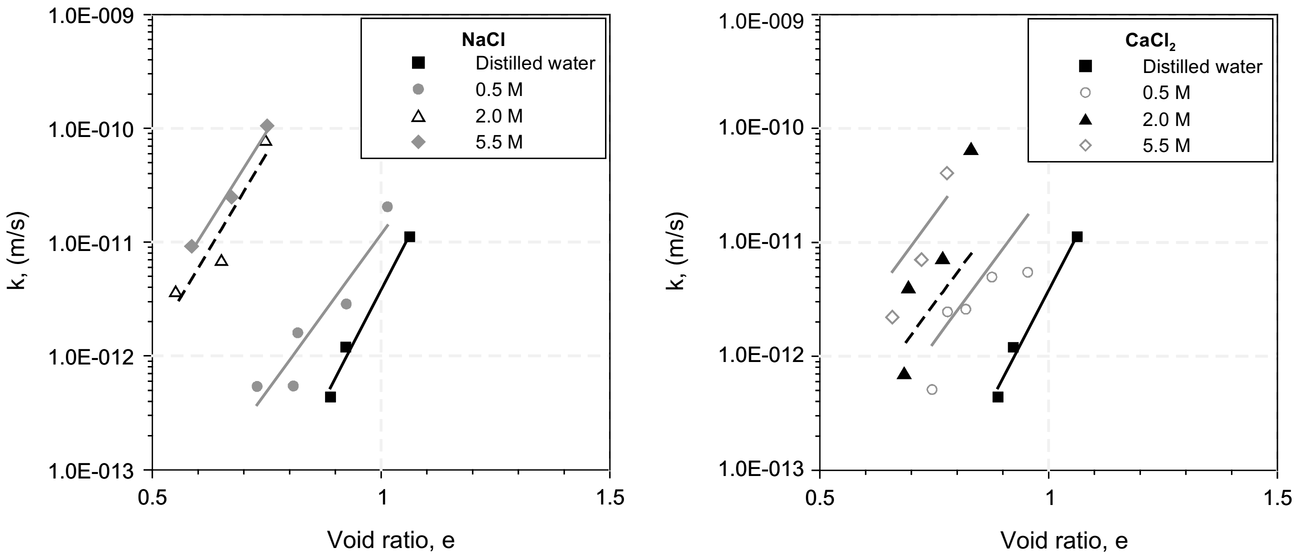

Fig. 14. Water permeability obtained from compression tests as a function of void ratio and salinity of the permeant.

would cause osmotic consolidation that would, not only decrease the primary swelling, but also counteract or hinder the secondary swelling.

The changes in salt concentration of the solutions can be related to changes in osmotic pressure (see Section 2), since the changes in salt concentration generate an osmotic suction difference, osmotic suction referring to the suction arising from the presence of dissolved salts in water. Hence, the impact on the swelling of the clay caused by salt solutions (taking into account their osmotic pressure) can be compared with the impact on clay strain caused by suction changes imposed in suction-controlled oedometers. Fig. 15 shows the volume changes during suction-reduction stages at constant vertical stress, measured in suction-controlled oedometers that use the vapour control technique for suctions higher than 14 MPa and the axis translation technique for lower suctions (Lloret et al., 2004). The same figure includes the swelling strain measured in the soaking tests using different salt solutions (shown previously in Fig. 9), considering the value of the osmotic suction associated to each of them. It seems that the magnitude of the two types of strains is comparable.

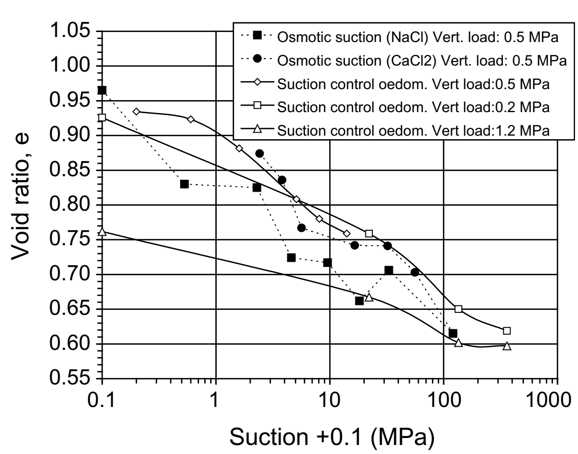

Fig. 15. Comparison between swelling strains due to suction-reduction and strains due to soaking with different NaCl and $CaCl_2$ solutions under different vertical loads (Lloret et al., 2004).

The effect of salinity on the hydraulic conductivity of the highly compacted saturated FEBEX bentonite is not very important when the permeant is a low-salinity solution. For the bentonite saline water (0.22 M, 0.8% salinity) the permeability values obtained were 184% higher on average than those expected for a sample of the same density tested with deionised water, and they showed greater dispersion. This higher permeability to saline water is more significant for low densities. The increase in permeability with salinity for high-salinity permeants (up to 5.5 M) is much obvious (up to two orders of magnitude), especially when the stress level is low and when Na is the predominant cation in the solution (Fig. 14).

Karnland et al. (1992) found an increase in permeability for MX-80 bentonite of half an order of magnitude when the salinity of the permeant increases from 0% to 3.5% NaCl. Villar (2005) found that the hydraulic conductivity of MX-80 bentonite was 135% higher for saline water (0.5% salinity) than for deionised water. These variations are in the order of those shown here for the FEBEX bentonite under similar conditions. However, the hydraulic conductivity of less expandable materials (such as Friedland Ton clay, with 45% of smectite) is more affected by the changes in the salinity of the permeant, increasing up to two orders of magnitude when salinity rises from 0% to 20% (Pusch, 2001), sodium giving place to a higher increase in permeability than calcium.

It is an accepted fact that the type of water used as a permeating agent, and especially its salinity, has an impact on the coefficient of permeability of clay (Klute, 1965; Olsen, 1962). The increase in permeability with the salinity of the permeating fluid was emphasised by several authors. In particular, it has been reported that the permeability of clays and clayey sandstones increases with the concentration of NaCl in the water. Rolfe and Aylmore (1977) attribute the changes in permeability observed in their tests with montmorillonite and illite to changes in ion distribution associated with variations in the cation exchange complex and to the concentration of the electrolyte used as permeating agent. There are various mechanisms that contribute to these changes, among them: (1) alterations in pore dimension distribution as a result of variations in swelling pressure in the clay matrix, (2) variations in the mobility of the molecules of water associated with the exchangeable cations adsorbed on the surfaces or forming diffuse double layers, and (3) alterations in the viscous behaviour of the structure of the water. As a result of these mechanisms, when the concentration of the electrolyte increases there is a reduction in the swelling capacity of the clay particles, the size of the flow channels increasing to the detriment of the number of small channels, this causing flow – and therefore, permeability – to increase. On the contrary, the higher development of diffuse double layers on reduction of the concentration of the electrolyte causes a decrease in permeability, because the size of the flow channels decreases. In expansive materials, it is the intrinsic permeability of the material itself that is altered by interactions between the fluid and the solid, and the largest variations in permeability with the composition of the fluid are found in clays with high content of montmorillonite (McNeal and Coleman, 1966). According to the diffuse double layer theory, its thickness decreases as the concentration of water in the pores increases, as a result of which, for a given porosity, the effective porosity of the clay would increase with increasing concentration of the solution, with the corresponding increase in permeability. In their studies with Na-montmorillonite, Studds et al. (1998) observed a clear increase in hydraulic conductivity as the saline concentration of the permeating agent increases, which they attributed to modifications induced by the latter in the effective porosity of the clay. The reduction of effective porosity with decreasing salinity of the permeating agent would result from occupation of the pore space by the bound water (DDL), the viscosity of which is higher than that of free water.

6. Conclusions

The effect of the salinity of the saturating fluid on the hydro-mechanical properties of compacted bentonite was investigated by means of swelling, compressibility and permeability tests in which deionised water and solutions of different concentrations and compositions were used as saturating fluids. The low and high-salinity ranges were chosen to simulate the expected and the extreme conditions in a high-level radioactive waste repository excavated in crystalline or clay host rocks.

The swelling capacity of the bentonite decreases with the increase in salinity of the pore water, although this change is less patent for high vertical loads and high densities, and when the salinity of the solution is very low. For an equivalent concentration and in the high-salinity range, sodium seems to reduce more the swelling capacity than calcium. In addition, the secondary swelling of samples saturated with high-salinity solutions is reduced or disappears.

The samples saturated with solutions containing high concentration of ions are less deformable and consolidate more rapidly than the samples saturated with low-salinity solutions.

The effect of salinity on the hydraulic conductivity of the highly compacted saturated FEBEX bentonite is not very important when the permeant is a low-salinity solution. However, the saturated permeability of the compacted bentonite clearly increases when high-salinity permeants are used. This higher permeability to saline water is more significant for low densities and when the stress level is low. For the high-salinity range, the permeability increases more when sodium is the predominant cation in the solution. This behaviour is contrary to what it is observed at saline concentrations of 0.5 M, in which a higher permeability is observed at low void ratios when calcium is the predominant cation. This higher permeability has been usually associated with microstructural changes and the tendency of calcium solutions to form denser aggregates, leaving larger macropores in between them. Further studies of these aspects are required to explain the unexpected higher permeability with sodium solutions at high concentrations.

For the salinities reasonably expected in the groundwater of a HLW repository excavated in crystalline or clay host rocks, the swelling and permeability properties of the FEBEX compacted bentonite will remain in the range of acceptable values. Much higher salinities (above 2 M) would drastically reduce the performance of the bentonite barrier.

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References

Arcos, D., Grandia, F., Domènech, C., 2006. Geochemical evolution of the near field of a KBS-3 repository. SKB Technical Report TR-06-16, Stockholm, 97 pp.
Castellanos, E., Gens, A., Lloret, A., Romero, E., 2006. Influence of water chemistry on the swelling capacity of a high-density bentonite. In: Miller, G.A., Zapata, C.E., Houston, S.L., Fredlund, D.G. (Eds.), Unsaturated Soils 2006. Geotechnical Special Publication 147 (1). ASCE, pp. 962–971.
Das, B.M., 1983. Advanced Soil Mechanics. Hemisphere Publishing Corporation, Washington.
ENRESA, 2000. FEBEX project. Full-scale engineered barriers experiment for a deep geological repository for high level radioactive waste in crystalline host rock. Final report. Publicación Técnica ENRESA 1/2000, Madrid, 354 pp.
ENRESA, 2006a. Full-scale engineered barriers experiment. Updated Final report 1994–2004. Publicación Técnica ENRESA 05-0/2006, Madrid, 590 pp.
ENRESA, 2006b. FEBEX project final report. Post-mortem bentonite analysis. Publicación Técnica ENRESA 05-1/2006, Madrid, 183 pp.
Fernández, A.M., Cuevas, J., 1998. Estudios del agua intersticial de la arcilla FEBEX. CIEMAT report. FEBEX Report 70-IMA-L-9-44, Madrid, 40 pp.
Hoffmann, C., Alonso, E.E., Romero, E., 2007. Hydro-mechanical behaviour of bentonite pellet mixtures. Physics and Chemistry of the Earth 32 (8-14), 832–849.
Karnland, O., Pusch, R., Sandén, T., 1992. The importance of electrolyte on the physical properties of MX-80 bentonite. SKB Report AR 92-35, Stockholm (in Swedish).
Karnland, O., Muurinen, A., Karlsson, F., 2005. Bentonite swelling pressure in NaCl solutions – experimentally determined data and model calculations. In: Alonso, E.E., Ledesma, A. (Eds.), Advances in Understanding Engineered Clay Barriers. Balkema, Leiden, pp. 241–256.
Klute, A., 1965. Laboratory measurements of hydraulic conductivity of saturated soil. In: Black, C.A. (Ed.), Methods of Soil Analysis. Agronomy Series, vol. 9. American Society of Agronomy, Inc., pp. 210–221.
Lambe, T.W., Whitman, R.V., 1979. Soil Mechanics. John Wiley & Sons, New York.
Lloret, A., Villar, M.V., 2007. Advances on the knowledge of the thermo-hydro-mechanical behaviour of heavily compacted FEBEX bentonite. Physics and Chemistry of the Earth, Parts A/B/C 32 (8–14), 701–715.
Lloret, A., Villar, M.V., Sánchez, M., Gens, A., Pintado, X., Alonso, E., 2003. Mechanical behaviour of heavily compacted bentonite under high suction changes. Géotechnique 53 (1), 27–40.
Lloret, A., Romero, E., Villar, M.V., 2004. FEBEX II project. Final report on thermo-hydro-mechanical laboratory tests. Publicación Técnica ENRESA 10/2004, ENRESA, Madrid, 165 pp.
McNeal, B.L., Coleman, N.T., 1966. Effect of solution composition on soil hydraulic conductivity. Soil Science Society of America Proceedings, 308–317.
Olsen, H.W., 1962. Hydraulic flow through saturated clays. In: Ninth Nat. Conf. on Clays and Clay Minerals. Pergamon, Oxford, pp. 170–182.
Pusch, R., 1994. Waste Disposal in Rock. Developments in Geotechnical Engineering, vol. 76. Elsevier, Amsterdam. 490 pp.
Pusch, R., 2001. Experimental study of the effect of high porewater salinity on the physical properties of a natural smectitic clay. SKB Technical Report TR-01-07, Stockholm, 35 pp.
Rao, S.M., Thyagaraj, T., Thomas, H.R., 2006. Swelling of compacted clay under osmotic gradients. Géotechnique 56 (10), 707–713.
Rolfe, P.F., Aylmore, L.A.G., 1977. Water and salt flow through compacted clays: I. Permeability of compacted illite and montmorillonite. Soil Science Society of America Journal 41, 489–495.
Romero, E., 1999. Characterisation and thermo-hydro-mechanical behaviour of unsaturated Boom-clay: an experimental study. Ph.D. Thesis, Universidad Politécnica de Cataluña, Barcelona, 405 pp.
Studds, P.G., Stewart, D.I., Cousens, T.W., 1998. The effects of salt solutions on the properties of bentonite–sand mixtures. Clay Minerals 33, 651–660.

Sugita, Y., Chijimatsu, M., Suzuki, H., 2003. Fundamental properties of bentonite pellet for prototype repository project. In: Proc. Int. Workshop Large scale field tests in granite. Advances in understanding and research needs, Universitat Politècnica de Catalunya-ENRESA, Sitges, November 12–14th, 2003.
Turrero, M.J., Peña, J., 2003. Arcilla española de referencia. Integración de resultados de la formación geológica: sondeos IC-1 y SS-1. CIEMAT/DIAE/54460/3/03, versión 1. ENRESA 2003, GTI 2- Descripción del Emplazamiento (in Spanish).
Villar, M.V., 2002. Thermo-hydro-mechanical characterisation of a bentonite from Cabo de Gata. A study applied to the use of bentonite as sealing material in high level radioactive waste repositories. Publicación Técnica ENRESA 01/2002, Madrid, 258 pp.
Villar, M.V., 2005. MX-80 bentonite. Thermo-hydro-mechanical characterisation performed at CIEMAT in the context of the prototype project. Informes Técnicos Ciemat, 1053, Madrid, 39 pp.
Villar, M.V., Lloret, A., 2001. Variation of the intrinsic permeability of expansive clay upon saturation. In: Adachi, K., Fukue, M. (Eds.), Clay Science for Engineering. Balkema, Rotterdam, pp. 259–266.