



## Influence of temperature on the hydro-mechanical behaviour of a compacted bentonite

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### Abstract

This paper reports the results on the influence of temperature (between 20 and 80 °C) on some hydro-mechanical properties of a compacted bentonite. The water retention capacity at higher temperature, both in confined and unconfined conditions, is lower, especially when suction is low. At high temperatures, the swelling capacity of clay decreases, although the influence of temperature is less evident when the applied stress is high. Furthermore, a decrease in swelling pressure as a function of temperature was observed. On the other hand, the measured increase in permeability with temperature is lower than expected on the basis of thermal change in water kinematic viscosity. The transfer induced by temperature between intra-aggregate adsorbed water and inter-aggregate free water may explain most of the features observed, taking into account that the physical characteristics (density, viscosity) of water in each one of these two states are different.

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### 1. Introduction

The design of high level radioactive waste (HLW) repositories in deep geological media—in which bentonite clay is proposed as a sealing material—leads to the need of further studying the behaviour of clayey soils when subjected to hydraulic and thermal changes. The behaviour of a repository is determined, to a large extent, by the changes that may occur in the mechanical, hydraulic and geochemical properties of

the engineered barrier, as a result of the combined effects of heat generated by the radioactive decay and of the water and solutes supplied by the surrounding rock. Temperature changes affect important hydraulic characteristics of compacted clays such as water retention and water permeability, whose knowledge is crucial to predict the hydration rate of the barrier: any small variation can lead to very significant changes in saturation time. In addition, the mechanical response of the material, which has important implications on the design of the repository, is also affected by temperature (swelling pressure, swelling and collapse, thermal dilatation and contraction, compressibility, yielding, effects on time-dependent behaviour). Laboratory tests may help to understand the processes

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that take place in the clay barrier under simple and controlled conditions, and to develop the governing equations. The laboratory tests enable to isolate the different processes, making their interpretation easier, and provide with fundamental data concerning the parameters to be used in the models.

In the last decade, equipment and experimental techniques were developed to test soils under different temperatures (Belanteur et al., 1997; Khemissa, 1998; Romero, 1999; Romero et al., 2001a) and a number of laboratory results, referring to thermal effects on saturated clayey soils, were presented (Towhata et al., 1993; De Bruyn and Thimus, 1996; Tanaka et al., 1997; Delage et al., 2000; Burghignoli et al., 2000; Graham et al., 2001), even though the results that focus on expansive soils are still limited (Lingnau et al., 1996; Romero et al., 2001c).

The temperature effect on hydraulic conductivity of compacted bentonites or additive/bentonite mixtures has been studied by several authors, who generally observed an increase in permeability with temperature, mostly attributed to the thermal decrease of kinematic viscosity (Mingarro et al., 1991; Cho et al., 1999).

The effect of temperature on water retention curves has been scarcely analysed, especially for temperatures much higher than those found in nature. It was observed that the retention capacity decreases with temperature in a manner that cannot be explained solely on the basis of thermal changes in surface tension (Romero et al., 2001b; Bachmann et al., 2002).

The volume change behaviour of non-swelling clays and compacted swelling clays under different temperatures was recently studied by Romero (1999), Romero et al. (2001c) and Sultan et al. (2002), among others. The influence of temperature on the mechanical behaviour of a geological material depends on its overconsolidation ratio, plasticity, expansibility and compaction water content. According to this, temperature may have different opposite effects: swelling/retraction, softening/stiffening, decrease of elastic domain/overconsolidation. These effects may in turn be reversible or irreversible. For example, the volume of normally consolidated clays contracts upon heating, while highly overconsolidated clays may even exhibit volume expansion when heated

(Baldi et al., 1988; Romero et al., 2001c). Besides, the volume change induced by temperature increases with the plasticity of clays (Demars and Charles, 1981).

The work presented here was performed in the framework of the FEBEX Project (Full-scale Engineered Barriers Experiment in Crystalline Host Rock), which studies 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 made of highly compacted bentonite blocks (ENRESA, 1995, 2000). In this paper, experimental results, obtained on compacted specimens subjected to temperature, suction and stress, are presented, as well as an attempt to interpret the cause of the temperature effects observed.

## 2. Material

The material tested is FEBEX bentonite, which was selected by ENRESA (the Spanish Agency for Radioactive Waste Management) as a suitable material to backfill and seal HLW repositories. It is being used in the form of compacted blocks in the large scale experiments of the FEBEX Project performed in the Grimsel Test Site (Switzerland) and CIEMAT (Madrid, Spain) (ENRESA, 2000).

The FEBEX bentonite was extracted from the Cortijo de Archidona deposit (Almería, Spain) and processed as follows: disaggregation and gently grinding, drying at 60 °C and sieving by 5 mm. For the big scale tests of the FEBEX Project, the bentonite blocks were manufactured by uniaxial compaction of the granulated clay with its hygroscopic water content, at dry densities close to 1.6 g/cm<sup>3</sup>.

The most relevant thermo-hydro-mechanical and geochemical characteristics of FEBEX bentonite are shown in detail in ENRESA (1998) and Villar (2000, 2002) and summarised in ENRESA (2000). FEBEX bentonite was formed by the hydrothermal alteration of acid volcanic rocks and has a montmorillonite content higher than 90%. Likewise, it contains variable quantities of quartz, plagioclase, K-feldspar, calcite and opal-CT (cristobalite-trydimite).

The cation exchange capacity (CEC) of FEBEX clay, determined as the sum of exchangeable cations

displaced by 1 M ammonium acetate at pH 8, is of  $102 \pm 4$  meq/100 g, being the major exchangeable cations predominantly divalent: Ca (42%), Mg (33%), Na (23%) and K (2%).

The liquid limit of bentonite is  $102 \pm 4\%$ ; the specific gravity is  $2.70 \pm 0.04$ ;  $67 \pm 3\%$  of the particles are smaller than  $2 \mu\text{m}$ . The hygroscopic water content in equilibrium with laboratory atmosphere (relative humidity  $50 \pm 10\%$ , temperature  $21 \pm 3 \text{ }^\circ\text{C}$ , total suction about 100 MPa) is  $13.7 \pm 1.3\%$ . The value obtained for the external specific surface using BET technique is  $32 \pm 3 \text{ m}^2/\text{g}$  and the total specific surface obtained using the Keeling hygroscopicity method is about  $725 \text{ m}^2/\text{g}$ . The low values of the Atterberg limits and the small clay-sized fraction can be explained taking into account the big quantity of silt-sized aggregates that are ‘pseudomorphs’ of the volcanic minerals transformed into smectite (Villar, 2002). The analysis of mercury intrusion data reveals that intra-aggregate pores (smaller than  $0.006 \mu\text{m}$ ) represent 73–78% of total pore volume when bentonite is compacted at a dry density of  $1.7 \text{ g/cm}^3$ .

From X-ray and nitrogen and water vapour adsorption isotherms, it can be derived that, at laboratory atmosphere, smectite lamellae have two monolayers of water and that, in wetting experiments, the montmorillonite takes a maximum of three monolayers of water. The water content associated to two water monolayers is about 14%, which represents a significant part of the total amount of water in the compacted bentonite (Fernández et al., 2003).

The hydraulic conductivity of FEBEX clay at room temperature ( $22 \pm 3 \text{ }^\circ\text{C}$ ) for deionised water can be related to dry density through an exponential law, which is different depending on the dry density interval (Villar, 2000, 2002):

For dry densities of less than  $1.47 \text{ g/cm}^3$ :

$$\log k_w = -6.00\rho_d - 4.09$$

$$(r^2 = 0.97, 8 \text{ measurements}) \quad (1)$$

For dry densities in excess of  $1.47 \text{ g/cm}^3$ :

$$\log k_w = -2.96\rho_d - 8.57$$

$$(r^2 = 0.70, 26 \text{ measurements}) \quad (2)$$

where  $k_w$  is the hydraulic conductivity (m/s) and  $\rho_d$  is the dry density ( $\text{g/cm}^3$ ). The difference between the

experimental values and these fittings is smaller for low densities than for higher values, with an average—in absolute values—of 30%. This should be evaluated taking into account that the values of permeability are of the order of  $10^{-13} \text{ m/s}$ .

The swelling pressure ( $P_s$ , MPa) of samples compacted with their hygroscopic water content and flooded with deionised water up to saturation at room temperature ( $22 \pm 3 \text{ }^\circ\text{C}$ ) can be related to dry density ( $\rho_d$ ,  $\text{g/cm}^3$ ) through the following equation (Villar, 2000, 2002):

$$\ln P_s = 6.77\rho_d - 9.07$$

$$(r^2 = 0.88, 52 \text{ measurements}) \quad (3)$$

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

Various authors have pointed out that in the vicinity of clay laminae, the structure of water molecules is disturbed, their properties differing from those of free water (Low, 1976; Sposito and Prost, 1982; Hueckel, 1992; Ichikawa et al., 1999; Hueckel et al., 2002). Specifically, for the density of water adsorbed in montmorillonite, Martin (1962) showed values of up to  $1.41 \text{ g/cm}^3$ . More recently, molecular dynamics simulations have revealed values of inter-layer water density between  $1.14 \text{ g/cm}^3$  in a Na smectite and  $1.38 \text{ g/cm}^3$  in a Mg smectite (Skipper et al., 1991). Through macroscopic experimentation, average water density values of up to  $1.50 \text{ g/cm}^3$ —depending on clay dry density and exchangeable cations—have been determined in different bentonites (Marcial, 2003). This higher water density provokes that the degrees of saturation of specimens fully saturated turn higher than 100% if they are calculated considering the density of water to be  $1.00 \text{ g/cm}^3$ . The average density of the pore water of FEBEX bentonite was obtained from the final water content of samples of different dry density, which were completely saturated with deionised water at constant volume, by adjusting the final degree of saturation to 100% in each of the tests performed (Villar, 2000, 2002). The values obtained are repre-

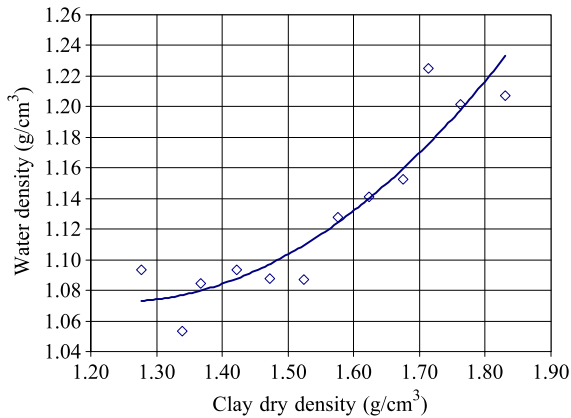


Fig. 1. Values of average density of adsorbed water calculated for specimens of bentonite of different dry density saturated with distilled water at constant volume (from Villar, 2002).

sented in Fig. 1, which shows that average water density increases when the volume of pores, and consequently the amount of free water, decreases.

Deionised water was used in all the tests in order to establish a clear reference condition. Tests performed at room temperature with granitic water (the one more likely in a Spanish HLW repository) showed small

differences in hydraulic conductivity and swelling capacity with respect to those performed with deionised water (Villar, 2000, 2002).

### 3. Test equipment and experimental procedures

#### 3.1. Retention curves

The retention curve (or soil water characteristic curve, SWCC) was determined both in confined and unconfined samples under different temperatures. Although the sealing material will be unconfined only in the outer part of the barrier, where a void exists between the compacted blocks and the host rock, the determination of the SWCC in unconfined samples is justified because, being much easier to determine, it will still be useful to identify the influence of temperature. In drying paths, it was not possible to maintain the volume of the sample constant.

The initial water content of the samples was the hygroscopic one (about 14%). For the determination of the retention curves under free volume conditions, clay was uniaxially compacted to obtain cylindrical speci-

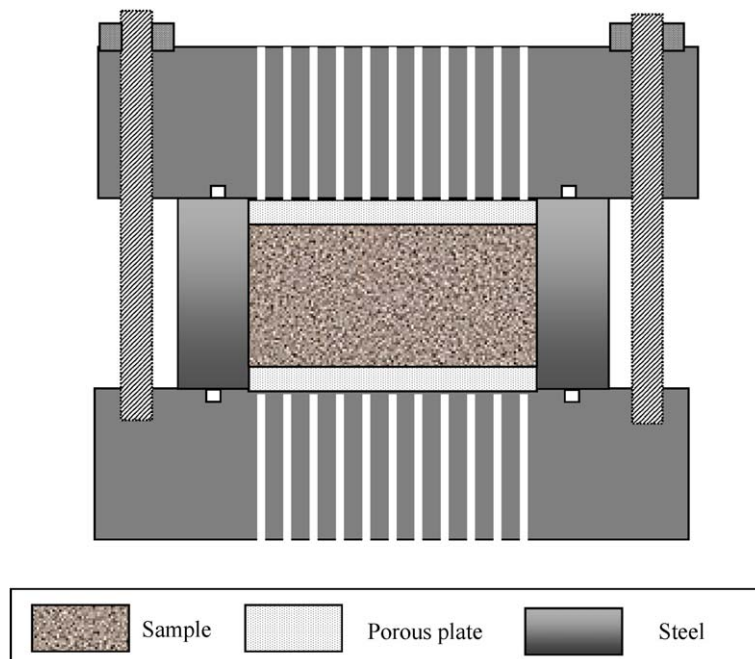


Fig. 2. Schematic layout of the non-deformable cell for the determination of the retention curve.

mens of 1.20 cm in length and a cross section of 7.23 cm<sup>2</sup>. In the case of constant volume determinations, and to avoid the swelling of clay, the samples were compacted in cells, which consist of a stainless steel cylindrical body measuring 0.5 cm in thickness, with two perforated covers joined by bolts (Fig. 2). Granulated clay is compacted directly inside the cell ring at room temperature using static uniaxial compaction. The length of the specimen is 1.20 cm and its cross section, 11.34 cm<sup>2</sup>.

The unconfined compacted samples or the cells are placed in desiccators with a sulphuric acid solution and vacuum is applied. The suction is, therefore, controlled through the control of relative humidity. In the case of constant volume tests, the porous stones and the perforated covers allow the exchange of water in the vapour phase between the clay and the atmosphere of the desiccator. This process is very slow—slower than when the retention curve under free volume conditions is determined in the desiccator—not only because just the upper and lower parts of the surface of the sample act as an exchange surface, but also because the process is performed at constant volume. Once the water content of the clay is stable (it was necessary to wait from 2 to 3 months to reach equilibrium), the solution in the desiccator is changed in order to apply a different suction. Prior to the change of the solution, the unconfined samples are measured to evaluate their volume change. In order to determine the curves at high temperatures, vacuum desiccators with sulphuric acid solutions are, in turn, placed in an oven at controlled temperature (between 40 and 80 °C).

### 3.2. Soaking under vertical load

The influence of temperature on the swelling capacity of clay was checked by soaking under vertical load tests. They were performed in an oedometer, whose cell is placed in a thermostatic bath with controlled temperature. Granulated bentonite with its hygroscopic water content (about 14%) was compacted inside the cell ring, at room temperature, using static uniaxial compaction until it reached nominal dry densities of 1.60 g/cm<sup>3</sup>. Vertical stresses between 22 and 30 MPa were applied to obtain specimens of 5.0 cm in diameter and 1.2 cm in height. After having reached the stabilisation of the

target temperature in the oedometer, vertical pressures of 0.5, 1.5 and 3.0 MPa were applied to the samples. Immediately afterwards, they were soaked with deionised water at atmospheric pressure from the bottom porous plate. The swelling strain experienced by the specimens upon saturation was recorded as a function of time until stabilisation. On completion of the tests, the water content of the specimens was determined and full saturation was verified. The swelling strain experienced by the specimens upon saturation was determined for temperatures ranging from 30 to 80 °C.

### 3.3. Swelling pressure and hydraulic conductivity

The determination of swelling pressure and hydraulic conductivity as a function of temperature was performed in high-pressure oedometer equipments (Fig. 3). Granulated clay was compacted uniaxially and statically at room temperature in the oedometer ring, which had an inner diameter of 5.0 cm, the length of the resulting specimen being 1.2 cm. Nom-

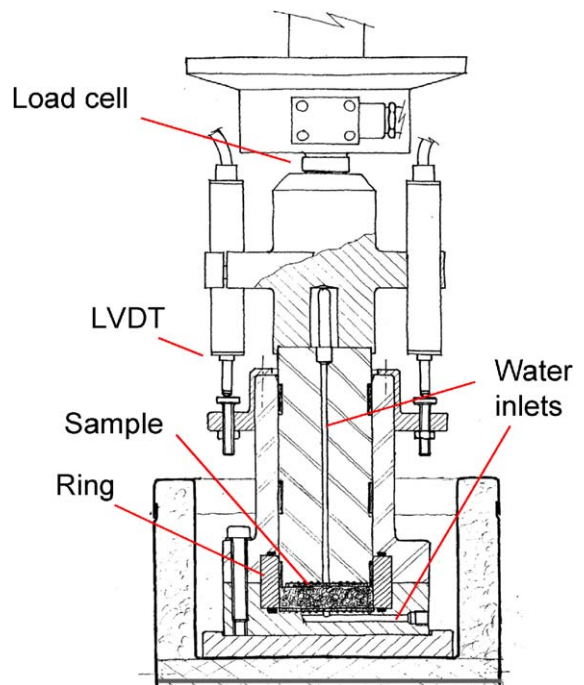


Fig. 3. Schematic layout of the swelling pressure cell inside the thermostatic bath.



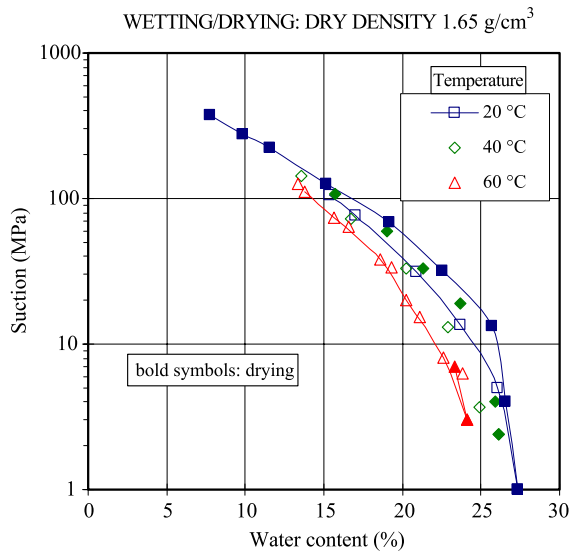


Fig. 4. Soil water characteristic curves at constant volume and at different temperatures in a wetting/drying path for bentonite compacted at dry density  $1.65 \text{ g/cm}^3$ .

inal dry densities of  $1.60 \text{ g/cm}^3$  were reached by applying vertical stresses of  $15 \pm 1 \text{ MPa}$ .

The oedometer assemblage is placed inside a silicone oil thermostatic bath that keeps target temperature. Once the temperature stabilises, the sample, confined between porous stainless steel sinters, is hydrated at constant volume through the bottom face with deionised water injected at a pressure of  $0.6 \text{ MPa}$ , while the upper outlet remains open to atmosphere. At the same time, a load cell installed in the loading frame measures the swelling pressure exerted by the clay. The small vertical deformation of the specimen, due mainly to load cell deformability, is measured by two LVDTs. An automatic volume change apparatus measures the water exchange of the specimen. The values of load, strain and water exchange are automatically recorded.

Once the sample is completely saturated (which is assumed by the stabilisation of water intake and swelling pressure development), the injection of water is stopped, and the pressure registered is considered the swelling pressure value for the dry density attained. The actual density may differ slightly from the nominal one due to the small displacement allowed by the equipment (of the order of  $10 \mu\text{m}$  when a vertical stress of  $2.2 \text{ MPa}$  is applied).

Afterwards, hydraulic conductivity is determined in the same equipment (Fig. 3) and on the same samples kept at constant volume. For that, the water pressure at the bottom of the samples is increased, while a backpressure of  $0.6 \text{ MPa}$  is applied on top, resulting in hydraulic gradients between 11,700 and 20,000. The water outflow is measured and the hydraulic conductivity is calculated applying Darcy's law. Tests performed with different hydraulic gradients have confirmed that there is no effect of the gradient magnitude (in the interval tested) on the hydraulic conductivity value (Villar, 2000, 2002).

## 4. Results

### 4.1. Retention curves

The soil water characteristic curve at constant volume, following a wetting/drying path, was determined for a dry density of  $1.65 \text{ g/cm}^3$  and temperatures of 20, 40 and  $60 \text{ }^\circ\text{C}$ . The curve has followed a wetting path, from suction 130 to  $0.1 \text{ MPa}$  and, afterwards, a drying path to  $130 \text{ MPa}$ . The initial water content of the samples was the hygroscopic one (about 14%). The results obtained are plotted in Fig. 4 in terms of water content reached by clay in equilibrium with a given suction. The retention capacity for

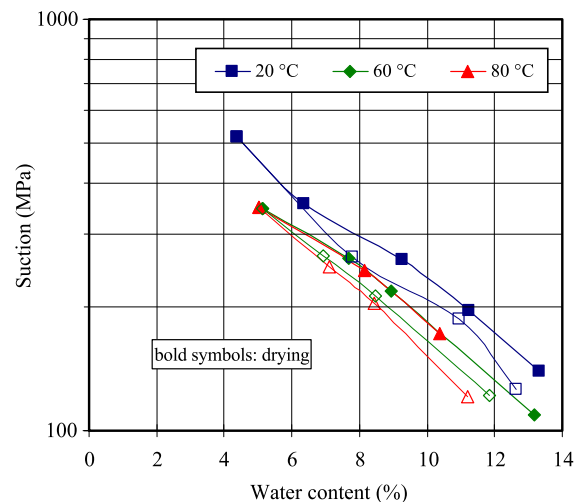


Fig. 5. Soil water characteristic curves in unconfined drying/wetting paths for bentonite compacted at initial dry density  $1.65 \text{ g/cm}^3$ .

the same dry density is lower the higher the temperature, especially for the lowest suctions. In the same figure, the hysteretic behaviour of the curves can be observed, since the water contents reached in drying paths are higher than those obtained during previous wetting.

The bentonite retention curves following a drying/wetting path were determined in unconfined samples for temperatures of 20, 60 and 80 °C and initial dry densities between 1.5 and 1.7 g/cm<sup>3</sup>. In terms of water content, no differences were observed in the retention capacity as a function of initial dry density. Fig. 5 shows the water content/suction relationship for samples compacted at an initial dry density of 1.65 g/cm<sup>3</sup>. As in the tests performed in confined samples, the retention capacity is higher for samples tested at laboratory temperature, although the difference between samples tested at 60 and 80 °C is minor. The lower retention capacity at higher temperature is clearer in the wetting after drying path. The curves obtained show hysteresis, since the water contents in wetting paths are lower than those reached for the same suction in the previous drying.

Fig. 6 shows the changes in dry density experienced by specimens during the determination of retention curves under unconfined conditions. For suctions higher than 100 MPa, the volume changes are small and do not depend on temperature. However, for suctions lower than 100 MPa, the decrease in dry density in the wetting path becomes increasingly important and different for each temperature. Swelling is greater for specimens tested at room temperature, in accordance with the results that will be presented in Section 4.2.

As discussed by several authors, the retention capacity of swelling materials is highly affected by the strain state, and it is higher for samples that can swell freely (unconfined samples) than for those kept at constant volume (Yahia-Aissa, 1999; Romero and Vaunat, 2000; Villar, 2000, 2002). The comparison between the curves determined under both conditions is shown in Fig. 7, where the above-mentioned aspect can be observed.

#### 4.2. Soaking under vertical load

The final strains upon soaking up to saturation with deionised water, which were reached for specimens

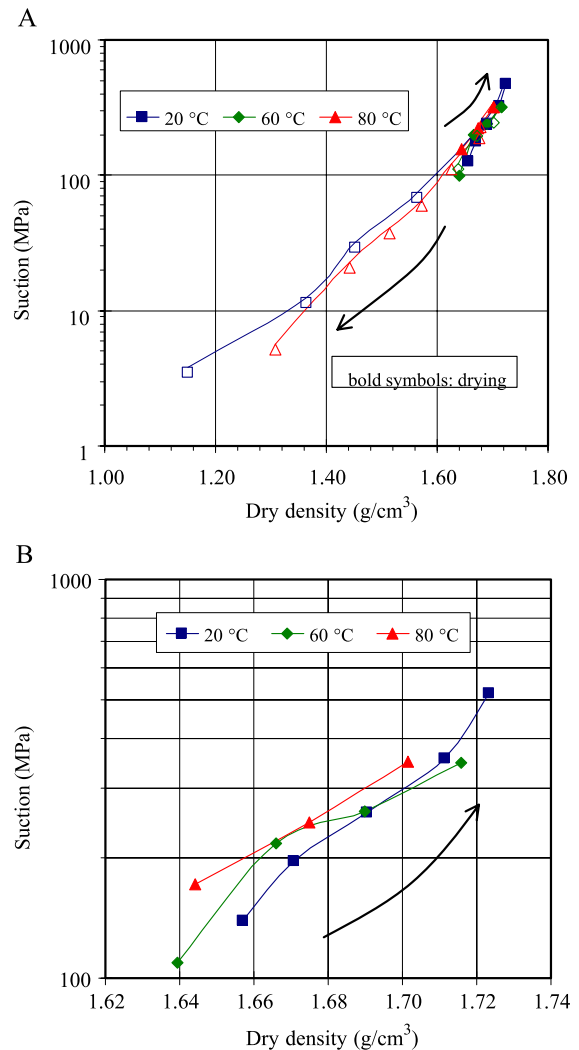


Fig. 6. Evolution of dry density in the retention curves in unconfined specimens of bentonite compacted at initial dry density 1.65 g/cm<sup>3</sup>. (A) Drying/wetting paths and (B) enlargement of drying paths.

compacted at an initial nominal dry density of 1.60 g/cm<sup>3</sup>, are plotted as a function of temperature and vertical stress in Fig. 8 (some of the results were presented in Romero et al., 2001c). The strain percentage is calculated as the increase of height with respect to the initial height of the sample, the negative values indicating swelling strains. The vertical loads applied were 0.5, 1.5 and 3.0 MPa, and the temperature of the tests were set to values between 30 and 80 °C. The swelling capacity decreases with temperature, al-

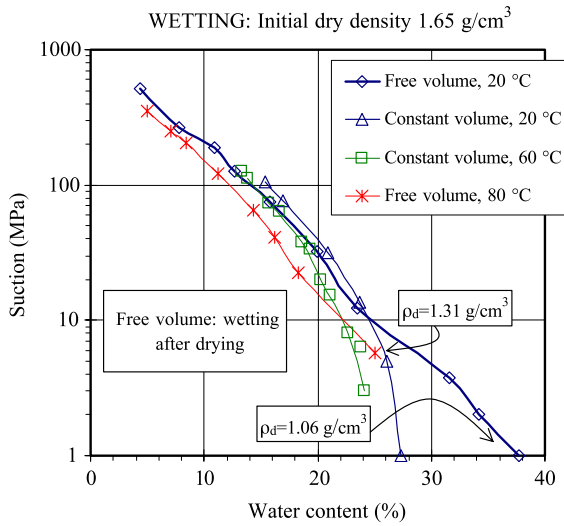


Fig. 7. Comparison of the retention curves obtained in confined and unconfined volume conditions at different temperature.

though the influence of temperature is less evident when the vertical stress is higher. A fitting was found between swelling strain ( $\varepsilon$ , %), temperature ( $T$ , °C) and vertical stress ( $\sigma$ , MPa):

$$\varepsilon = (-0.11\sigma + 0.38)T + (8.80\sigma - 36.72) \quad (4)$$

The lines displayed in Fig. 8 were obtained by using this equation. Although the fittings came from

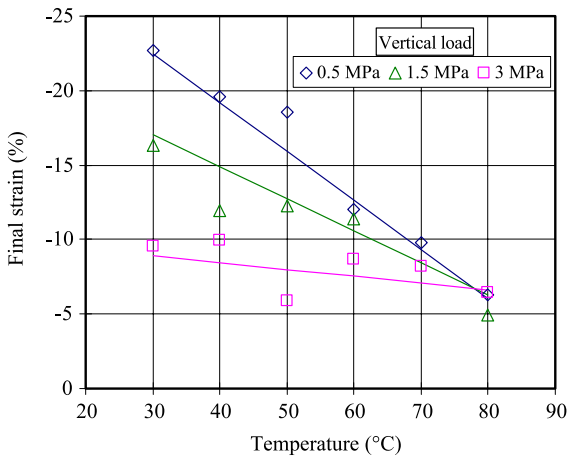


Fig. 8. Final vertical strain of samples compacted at initial dry density 1.60 g/cm<sup>3</sup> saturated with deionised water under different vertical pressures and temperatures.

tests performed at temperatures ranging from 30 to 80 °C, the extrapolation of lines towards higher temperatures seems to indicate that the swelling capacity under the lowest vertical stresses would disappear for temperatures higher than 100 °C.

### 4.3. Swelling pressure

Several tests were performed at different temperatures with clay compacted uniaxially with its hygroscopic water content in the oedometer ring at a nominal dry density of 1.60 g/cm<sup>3</sup>. The swelling pressure cell displayed in Fig. 3 was used to perform these tests. The results are shown in Fig. 9, in which the dispersion of data can be mostly attributed to the variations in dry density (whose average value was in fact 1.58 g/cm<sup>3</sup>). This is caused by the small displacement allowed by the equipment, as the swelling pressure value is very sensitive to little density changes (see Eq. (3)). The error bar shown in Fig. 9 was obtained from values measured in tests performed at laboratory temperature to obtain Eq. (3). A decrease of swelling pressure as a function of temperature is observed. However, the extrapolation of the logarithmic correlation towards higher temperatures would indicate that swelling pressures around 2 MPa would be developed even for temperatures of 100 °C. This would be in accordance with the results obtained in soaking under load tests, which

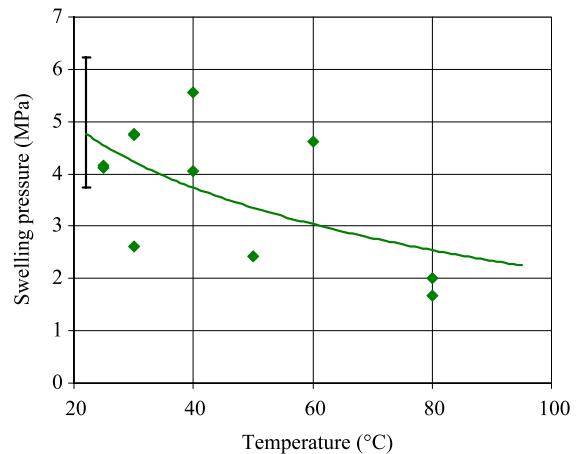


Fig. 9. Swelling pressure values as a function of temperature for the saturated clay compacted at an average dry density of 1.58 g/cm<sup>3</sup>.



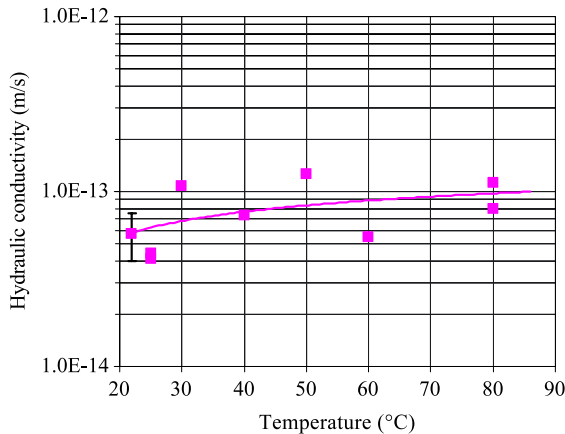


Fig. 10. Hydraulic conductivity values as a function of temperature for the saturated clay compacted at an average dry density of 1.58 g/cm<sup>3</sup>.

predict a smaller decrease in swelling capacity with temperature for high vertical stresses (Fig. 8). Lingnau et al. (1996) also observed a reduction in swelling pressure with temperature for a sand/bentonite mixture, although it did not show any loss in the self-healing capability of the material, even for temperatures of up to 100 °C.

#### 4.4. Hydraulic conductivity

Following the tests described in Section 4.3, and once the samples were completely saturated with deionised water, the hydraulic conductivity was determined in the same equipment (Fig. 3) and on the same samples with an average dry density of 1.58 g/cm<sup>3</sup>.

The results obtained are shown in Fig. 10. The error bar shown in Fig. 9 was obtained from values measured in tests performed at laboratory temperature to obtain Eq. (2). The effect of temperature on permeability is less evident than on swelling pressure, although it slightly increases in a logarithmic way with temperature, as expected from the decrease in water kinematic viscosity.

## 5. Discussion

The variation of the properties of pore water with temperature undoubtedly affects the hydro-mechanical properties of soils. The property of water that may

have, at most, an influence in the retention capacity of a soil is the surface tension, due to its repercussion in capillary phenomena. However, Bachmann et al. (2002) find out that there is a dependence of the SWCC of natural soils on temperature (between 5 and 38 °C) much greater than what could be expected from the change in surface tension, and they consider that the variation of the contact angle with temperature could also had an influence, as well as the changes in liquid–gas interfacial tension because of solute effects. Romero et al. (2001b) conclude also that the influence of temperature on the retention capacity of a compacted kaolinitic-illitic clay (Boom clay) cannot be only due to the thermal dependence of surface tension and that alteration of clay fabric and intra-aggregate fluid chemistry with temperature could also occur. In FEBEX bentonite, and due to the high suctions involved (usually higher than 10 MPa), the capillary tube model (equation of Laplace) may have little sense and the changes in water surface tension are not decisive. Consequently, the changes in other physico-chemical properties of micropore water and in the microfabric must play a determinant role in the dependence on temperature of the retention capacity. Specifically, the transfer of tightly bound intra-aggregate water to free inter-aggregate water caused by temperature (Derjaguin et al., 1986; Ma and Hueckel, 1992, 1993) could explain the higher retention capacity at lower temperatures, if we accept that the density of adsorbed water is higher than that of free inter-aggregate water (see Section 2). The influence of temperature on the retention capacity of FEBEX clay at constant volume becomes greater for the lowest suctions (Fig. 4), what would suggest that the samples tested at high temperature saturate earlier. We think this could be caused by the thermally driven degeneration of high-density intra-aggregate water into inter-aggregate water of density equal to that of free water. This inter-aggregate water will consequently occupy a greater pore volume and give rise to higher degrees of saturation.

Pusch et al. (1990) pointed out that the influence of heating on the swelling capacity decreases as the confining stress increases. They observed that heating is of negligible importance for swelling pressure development at very high bulk densities, while it is rather significant at low bulk densities. The dependence on temperature of the volume changes upon saturation was recently analysed by Romero et al.

(2001c) for different types of clay. For FEBEX bentonite, an overconsolidated active clay, they explain the observed reduction in swelling strains at elevated temperatures (Fig. 8), as a consequence of the predominance of water transfer phenomena between micro and macrostructure over aggregates thermal expansion. The fact that the effect of temperature be less important when the applied stress is high, would indicate that the processes that reduce the swelling capacity at high temperature are hindered when the volume change is obstructed. This would support the hypothesis of the temperature-driven transfer of high-density intra-aggregate water to free inter-aggregate water, which occupies a higher volume. On the other hand, the minor contribution of mineral thermal expansion phenomena to overall volume change upon saturation is also manifested in the evolution of dry density in the wetting paths of retention curves determined under unconfined conditions: the volume increase is higher for samples tested at room temperature (Fig. 6A). Even in the drying paths and when clay is very dry (water contents below 14%), the samples tested at different temperatures experience a similar volume reduction (Fig. 6B).

The reduction in swelling pressure with temperature could be explained both by the reduction of the swelling capacity of the above-mentioned microstructure and by the decrease of the yield surface size (LC) of the macrostructure (Hueckel and Borsetto, 1990; Gens, 1995). According to Pusch et al. (1990), the effect of heating on the swelling pressure of montmorillonite differs depending on the predominant cation in the exchange complex: swelling pressure is found to increase in Na bentonite while it decreases in Ca bentonite. The reason invoked is that the net effect of two mechanisms—lattice contraction due to dehydration of the interlamellar space and increase of the osmotic pressure at stack contacts—is different for the two types of clay. Lattice contraction predominates in Ca bentonite while the increase in osmotic pressure predominates in Na bentonite, where the number of stack contacts is higher. A decrease in swelling pressure due to temperature was also observed in the FEBEX Project mock-up test performed at CIEMAT (ENRESA, 2000), where an accidental increase of the temperature of the heaters from 100 to 200 °C provoked a decrease in the stresses measured by sensors placed in the bentonite blocks (Martín and Barcala,

2001). This overheating episode was successfully analysed with a generalised plasticity model that considers two structure levels in bentonite (Sánchez et al., 2001). Decreases of swelling pressure with temperature were also measured in other materials: beidellite (Karland et al., 1994), sand/bentonite mixtures (Lingnau et al., 1996) and Boom clay (Romero et al., 2003). On the other hand, increases in swelling pressure with temperature were found for saponite (Karland et al., 1994).

The increase of hydraulic conductivity with temperature has been frequently attributed to the changes in water kinematic viscosity. This increase, resulting from the decrease in water viscosity with temperature, was calculated for FEBEX bentonite, taking as a starting point the permeability value determined at laboratory temperature (22 °C) from Eq. (3). The results are plotted in Fig. 11, where the experimental values were also plotted. Despite the scarce experimental data, the increase in hydraulic conductivity with temperature observed could be considered as slightly lower than that predicted solely on the basis of water viscosity change.

However, Kharaka and Smalley (1976) reported hydraulic conductivity values of a Na bentonite and Mingarro et al. (1991) of a granite/Ca bentonite mixture, which increased with temperature (ranging from 20 to 100 °C) roughly according to estimates from the decreases in the kinematic viscosity of water.

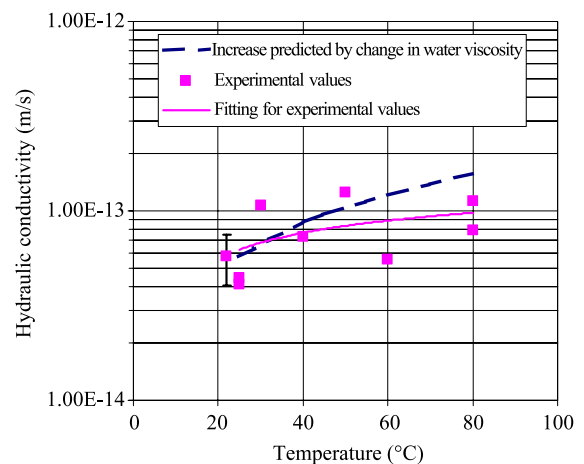


Fig. 11. Comparison of hydraulic conductivity values determined experimentally at different temperatures for clay compacted at dry density  $1.58 \text{ g/cm}^3$  and those predicted for the same temperatures taking into account the change in water kinematic viscosity.

Table 1  
Values of coefficient  $\beta_T$  found for different materials (modified from Romero et al., 2001b)

Soil	$\beta_T$ (K <sup>-1</sup> )	Reference
FEBEX bentonite	0.010	This work
Ca–Mg–Na montorillonite	0.020	Rivas et al. (1991)
Saponite	0.067	Rivas et al. (1991)
Kaolin	0.042	Towhata et al. (1993)
Kaolin	0.010	Khemissa (1998)
Ca bentonite	0.022	Cho et al. (1999)
Boom clay (illite-kaolinite)	0.005	Romero et al. (2001b)

Likewise, Cho et al. (1999) found similar results for a Ca bentonite, and Khemissa (1998), for a kaolinite. On the other hand, Towhata et al. (1993) found a higher hydraulic conductivity increase with temperature than that calculated by using the properties of free and pure water in a kaolinite and a bentonite. They consider that the degeneration of adsorbed water into bulk pure water at elevated temperatures (Derjaguin et al., 1986) may result in an increase in the dimension of flow channels and therefore of intrinsic permeability to liquid phase. Romero et al. (2001b) analysed the influence of temperature on the unsaturated water permeability of Boom clay and concluded that, although it is more relevant at high degrees of saturation, it is smaller than it could be expected from the thermal change in water viscosity. They consider that thermo-chemical effects altering clay fabric and porosity redistribution could be relevant processes that affect the change of permeability with temperature.

The effect of temperature on hydraulic conductivity in the interval of 20–80 °C can be approximated through a factor  $\beta_T$ , as suggested by Romero et al. (2001b):

$$k_w(T)/k_w(T_0) \approx 1 + \beta_T(T - T_0) \tag{5}$$

where  $k_w$  is the saturated hydraulic conductivity, and  $T$  and  $T_0$  are the temperature of the measurement and the temperature of the reference measurement (usually room temperature).

The value of  $\beta_T$  evaluated taking into account the change in water density and viscosity is of 0.030 K<sup>-1</sup>. For FEBEX bentonite, the value of  $\beta_T$  is 0.0097 K<sup>-1</sup>, i.e. much smaller than the theoretical

one. The values of this parameter, found for different materials, are shown in Table 1 (modified from Romero et al., 2001b), in which it can be observed that the changes of hydraulic conductivity with temperature can be above or below those expected on the basis of the change in water viscosity. To further illustrate this aspect, the variation of hydraulic conductivity with temperature of two bentonites compacted at dry density 1.59 g/cm<sup>3</sup> is shown in Fig. 12. S-2 bentonite comes from the same deposit than FEBEX bentonite and they are both very similar, as they belong to the Ca–Mg–Na montmo-

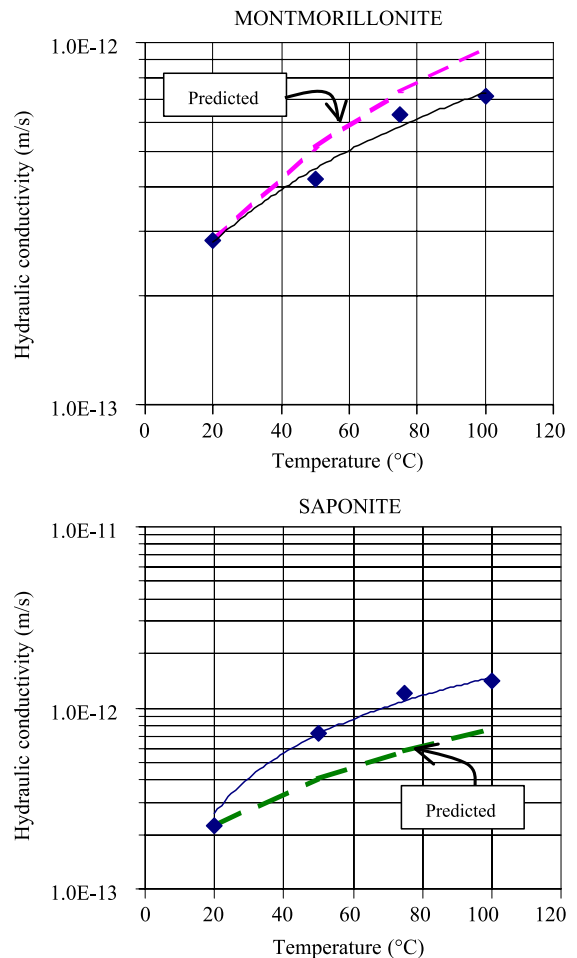


Fig. 12. Hydraulic conductivity of S-2 (montmorillonite) and MCA-C (saponite) bentonites compacted at dry density 1.59 g/cm<sup>3</sup> as a function of temperature: experimental values (Rivas et al., 1991) and predictions according to the water kinematic viscosity change.

rillonite type (Volckaert et al., 1996; ENRESA, 1998). The MCA-C bentonite is a saponite containing a 10% of sepiolite that comes from the Madrid sedimentary basin and was characterised by Cuevas et al. (1993). The tests were described in an unpublished report by Rivas et al. (1991). The foreseen variation of permeability with temperature, on the basis of the thermal change of water viscosity, was calculated and plotted in the same figures. For S-2 bentonite the actual increase is, as in the case of FEBEX bentonite, lower than predicted, while the contrary happens with the saponite.

## 6. Conclusions

Results on the influence of temperature on the hydro-mechanical properties of a compacted bentonite have been presented. Temperature effects on water retention curves in confined and unconfined conditions were determined and swelling pressure, hydraulic conductivity and swelling strains as a function of temperature were successfully measured.

The water retention capacity at higher temperature is lower, both under confined and unconfined conditions, being this fact more significant for lower suctions. It is also evident by the hysteretic behaviour of the clay, the water contents reached during drying higher than those obtained during the previous wetting (in confined samples) and the water contents in the wetting paths lower than those reached for the same suction in the previous drying (in unconfined samples). Below a given value of suction, the retention capacity under unconfined conditions is higher than under confined conditions.

The swelling strains of bentonite upon saturation are a function of temperature and vertical stress: at high temperatures, the swelling capacity of clay decreases, although the influence of temperature is less evident when the vertical stress is high. Likewise, a decrease in swelling pressure as a function of temperature (down to 2.5 MPa at 80 °C) was observed. On the other hand, the measured increase in permeability with temperature is small and slightly lower than what would be expected on the basis of the thermal changes in water kinematic viscosity.

Temperature induced transfers between intra-aggregate adsorbed water—of density higher than that

of free water—and inter-aggregate free water could explain most of the features observed. However, a better knowledge of the properties of adsorbed water (density and viscosity) and of their variation with temperature and with density and water content of clay is crucial to understand the thermo-hydro-mechanical behaviour of compacted expansive soils.

The literature survey and some old data reviewed revealed that the effects of temperature may differ depending on the type of material, and even on the type of cations in the exchange complex. Consequently, the conclusions reached in this work cannot be extrapolated to any other soil. Besides, the influence of drainage conditions and the long-term evolution of the properties studied are issues that must be tackled to fully understand the hydro-mechanical behaviour of clays at high temperature.

FEBEX bentonite remains suitable as a sealing material in HLW repositories (from the hydro-mechanical point of view) for temperatures of up to 80 °C, as it keeps its high water retention capacity, low permeability and self-healing ability. No data are still available for higher temperatures, although the extrapolation of results points out to the preservation of properties for at least up to 100 °C.

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