DIAE/54121/2/99

RESEAL PROJECT

EC Contract FI4W-CT96-0025

TOPICAL REPORT ON LABORATORY TESTS

Version 1

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November 1999

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1. INTRODUCTION

This report summarises the activities carried out by CIEMAT (Programa de Caracterización Hidrogeoquímica de Emplazamientos) and the results obtained during the RESEAL Project concerning the Work Package 1: Laboratory experiments. The tests were focused mainly to the investigation of the hydraulic and gas transport properties of the sealing material and of its geomechanical properties. Most of the tests have been performed with Serrata clay, as the decision of using FoCa clay was not taken until July 97, and most tests were already started. Results of the tests being performed with FoCa clay are also presented. Both compacted samples and pellets/powder mixtures have been used. Some work carried out in the framework of the FEBEX Project that can be useful for the objectives of RESEAL is also presented.

2. SERRATA CLAY CHARACTERISTICS

Most of the tests have been performed with a bentonite coming from the Cortijo de Archidona deposit (Almería, Spain), selected by ENRESA as suitable material for the backfilling and sealing of HLW repositories. It is the same than the clay used for the FEBEX Project in the in-situ (Grimsel, Switzerland) and the mock-up (Madrid, Spain) tests (ENRESA 1998, ENRESA 1999). The processing at the factory has consisted in disaggregation and gently grinding, drying at 60 °C and sieving by 5 mm. It has a content of montmorillonite higher than 90 %. The predominant phylosilicate is in fact a smectite/illite mixed layer, with 10-15 percent of illite layers. Besides, it contains variable quantities of quartz, plagioclase, K-feldspar, calcite and opal-CT (cristobalite-trydimite).

The cation exchange capacity (CEC) is of 113 ± 6 meq/100g, and the major exchangeable cations are: Ca (42 %), Mg (33 %), Na (23 %) and K (2 %).

The chemical composition of an aqueous extract of bentonite/water ratio (b/w) of 1/4 is (mg/l): Cl^- (150±12); SO_4^{2-} (201±20); HCO_3^- (154±6); Mg^{2+} (3±0); Ca^{2+} (4±1); Na^+ (246±11); K^+ (6±2). The pH of the extract is 8.7±0.2.

The liquid limit of the bentonite is 103 ± 4 % and the specific weight is 2.70. The equilibrium gravimetric water content of the clay at CIEMAT laboratory conditions (R.H. 50±10 %, which corresponds to a suction of around 130 MPa) is about 13.7±1.3 %.

The N_2-BET external surface (a_s) is 32±2 m^2/g, while the total specific surface determined by the Keeling hygroscopicity method is 725±47 m^2/g. More than 80% of the pore space, measured by N_2 adsorption in powder samples, lies in a continuous range of the mesopore region (diameter 500-20 Å) with an average value of 91Å.

The structure of the clay has been also analysed by means of mercury porosimetry, which allows the quantification of pores of diameter greater than 60 Å. In the compacted clay, previously freeze-dried, three families of pores can be observed, that will be termed big, medium and small pores. The percentage of pores with diameter greater than 6 µm is 23±6, with a mode of 18±8 µm; the percentage of pores with diameter between 6 and 0.1 µm, is 26±6, with a mode of 0.8±0.6 µm; and the percentage of pores with diameter less than 0.1 µm is 50±10, with a mode about 0.016±0.006 µm. Big and medium pores would correspond approximately to the domain of the macrostructure, and the small pores to the domain of the mesostructure, according to the classification given in Sing et al. (1984). The micropores are not represented in the determinations made by mercury intrusion.

For dry densities higher than 1.45 g/cm^3 the relationship between hydraulic conductivity (K, m/s) and dry density (ρ_d, g/cm^3) can be expressed by:

$$\log K = -3.0\ \rho_d - 8.6$$

This equation has been obtained from tests in which distilled water was used as permeant and the clay was directly compacted in the permeability cell. The variation of the experimental values with respect to this fitting is around 40 % (take into account that the permeability values are in the order of 10^{-13} m/s).

The swelling pressure (P_s, MPa) of compacted samples can be related to the dry density (ρ_d, g/cm^3) through the following equation:

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

3. LABORATORY EXPERIMENTS

3.1 Gas migration through clay based backfill and sealing materials

Gas permeability has been measured in compacted samples of Serrata clay of different initial dry density and water content. The sample is placed in a triaxial cell were a confining pressure of 1.6 MPa is applied. It is covered by two latex membranes with silicone paste between both, and porous stones are placed on top and bottom. The bottom of the sample is connected to a hermetic deposit of known volume in which nitrogen gas has been previously injected to a pressure slightly higher than the atmospheric one. The gas pressure in the deposit is measured by a pressure transducer connected to a DAS system. The upper outlet of the cell is open to the atmosphere. During the test, the air in the deposit is allowed to flow through the sample, while the pressure decrease in the deposit is monitored. The tests have been performed at laboratory temperature. A schematic representation of the experimental set-up is shown in Figure 1.

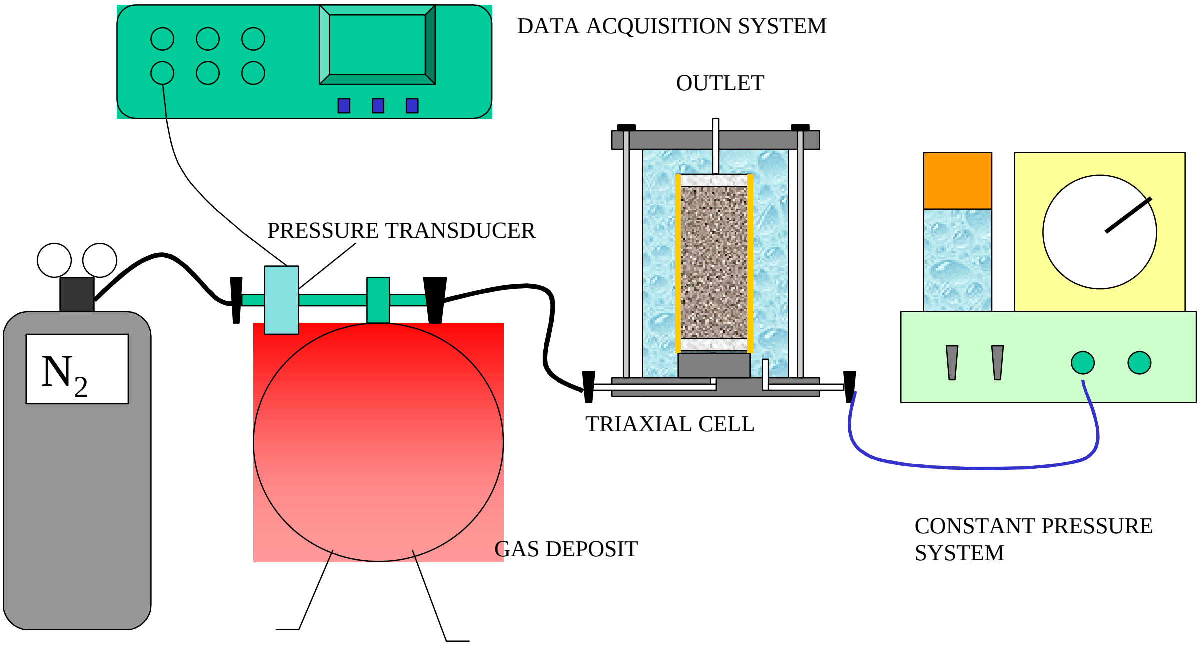

Figure 1. Schematic representation of the set-up to measure gas permeability

The gas permeability is calculated according to the following expression (Yoshimi & Osterberg 1963):

$$k = 2.3 \times \frac{V \times h \times m_a}{A \times \left(P_{atm} + {P_0}/{4}\right)} \times \frac{-\mathrm{Log}_{10}\left({P(t)}/{P_0}\right)}{t - t_0}$$

where k is the intrinsic permeability (m^2), V is the volume of the deposit (m^3), h is the height of the sample (m), A is the section of the sample (m^2), μ_a is the air dynamic viscosity ($N\cdot s/m^2$), P_{atm} is the atmospheric pressure (N/m^2), P_0 is the excess pressure in the deposit over the atmospheric one for time t_0 (s) and P(t) is the excess over the atmospheric pressure for time t.

The volume of the deposit is $2.21\cdot10^{-2}$ m^3, the nominal height of the sample is 8.0 cm and the section 11.4 cm^2. All the tests have been performed with nitrogen gas, whose dynamic viscosity has been taken as $1.79\cdot10^{-5}$ Pa·s. The pressure in the deposit at the beginning of the tests is around 1.03 bar.

Taking into account the dynamic viscosity of nitrogen, and assuming a gas density of 0.04 mol/l (CRC Handbook), the following relationship between the intrinsic permeability measured with nitrogen gas (k, m^2) and the permeability to gas (K_g, m/s) can be established:

$$K_g = \frac{r \times g}{m} \times k = 6.2 \cdot 10^5 \times k$$

Gas permeability determinations have been performed on samples of Serrata clay compacted to nominal dry density of 1.70 g/cm^3 with different water contents. The values obtained are shown in Table I. Values have also been obtained for dry densities of 1.50 and 1.60 g/cm^3.

Table I: Intrinsic and relative permeabilty values obtained for Serrata clay compacted to nominal dry density of 1.70 g/cm^3

ρ_d (g/cm^3)	w (%)	S_r (%)	k (m^2)	log k	K_r (m/s)
1.68	6.1	27	$8.5\cdot10^{-14}$	-13.07	$5.3\cdot10^{-8}$
1.66	6.4	28	$1.4\cdot10^{-13}$	-12.87	$8.3\cdot10^{-8}$
1.67	7.2	32	$6.3\cdot10^{-14}$	-13.20	$3.9\cdot10^{-8}$
1.70	8.4	39	$3.8\cdot10^{-14}$	-13.42	$2.4\cdot10^{-8}$
1.70	10.2	47	$2.4\cdot10^{-14}$	-13.61	$1.5\cdot10^{-8}$
1.70	10.7	49	$2.8\cdot10^{-15}$	-14.55	$1.7\cdot10^{-9}$
1.68	11.0	49	$1.6\cdot10^{-14}$	-13.79	$1.0\cdot10^{-8}$
1.67	12.0	53	$2.9\cdot10^{-14}$	-13.54	$1.8\cdot10^{-8}$
1.69	12.0	54	$2.5\cdot10^{-14}$	-13.61	$1.5\cdot10^{-8}$
1.68	12.4	55	$1.1\cdot10^{-14}$	-13.94	$7.1\cdot10^{-9}$
1.69	12.8	58	$2.6\cdot10^{-14}$	-13.58	$1.6\cdot10^{-8}$
1.70	13.8	63	$3.5\cdot10^{-15}$	-14.45	$2.2\cdot10^{-9}$
1.70	14.0	61	$8.7\cdot10^{-15}$	-14.06	$5.3\cdot10^{-9}$

The gas permeability values obtained with respect to the pore volume accessible to gas, represented as e (1-S_r), are plotted in Figure 2. From this figure the following correlation can be drawn between permeability (K_g, m/s), degree of saturation (S_r) and void ratio (e):

$$K_g = 2.97\cdot10^{-6}\,(e\,(1-S_r))^{4.32} \quad (r^2=0.79,\ 41\ \text{values})$$

This relationship is valid for degrees of saturation between 0.25 and 0.80, for higher degrees of saturation the diminution of intrinsic permeability is very sharp.

It has been observed that, for a given void ratio, the value of intrinsic permeability obtained when the measurement is done with water in the saturated sample, is much lower than that obtained when the measurement is performed with gas in the unsaturated sample. For example, for a dry density of 1.70 g/cm^3, the saturated hydraulic conductivity of the clay, measured with water, is $2.0\cdot10^{-14}$ m/s, which corresponds to an intrinsic permeability in the order of 10^{-21} m^2. For the same dry density, the intrinsic permeability obtained with gas measurements is 10^{-15} m^2 for a degree of saturation of 63 %.

This observation is a consequence of the variation of the structure of the bentonite as a function of the degree of saturation. As the degree of saturation increases, the clay particles swell and fill the big pores among aggregates, what provokes the diminution of the average pore size, and consequently, although the total porosity remains the same, the intrinsic permeability decreases.

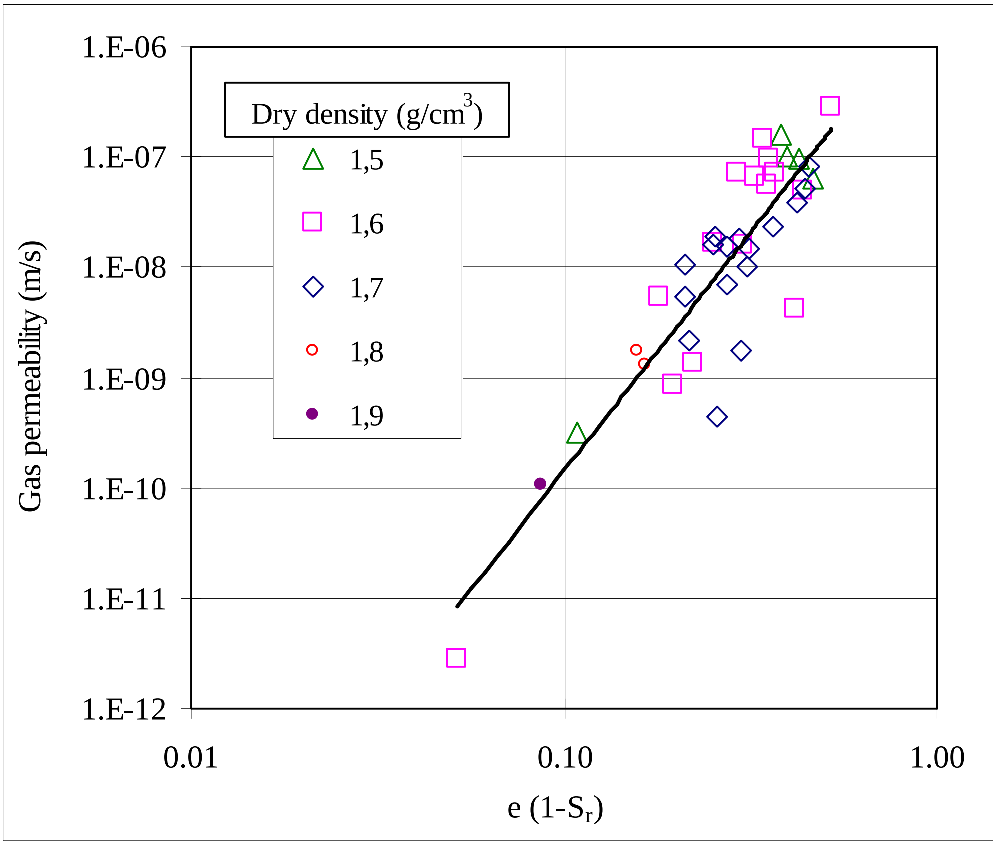

Figure 2: Gas permeability (K_g) values for samples of Serrata clay compacted to different dry density

3.2 Geomechanical properties of the sealing materials

3.2.1 <u>100x100 mm oedometer cell</u>

Several tests have been performed in a cell specially designed to work with pellets/powder mixtures. It has an inner diameter of 10.0 cm and an inner height of 10.0 cm. The sample can be hydrated by both faces or only on top or bottom. At the same time, the swelling pressure exerted by the clay and its deformation can be measured and automatically registered. The water intake is also measured by an automatic volume change apparatus. An schematic representation of the cell is shown in Figure 3.

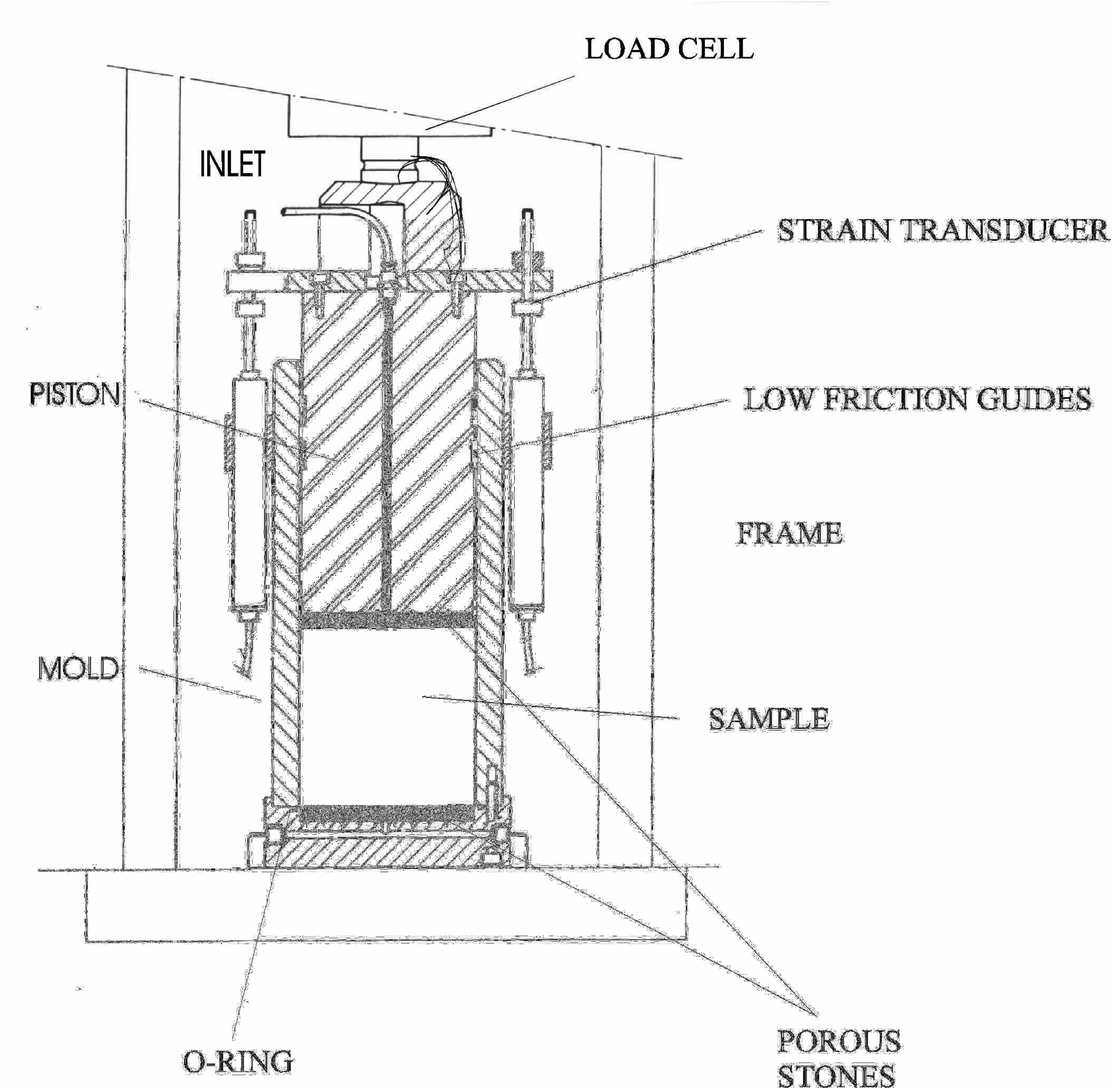

Figure 3: Schematic representation of the oedometer for pellets/powder mixtures

Several tests have been performed. At the beginning, a checking tests with Serrata powder was performed, the results are not presented here. Tests MGR2 and MGR3 were performed with Serrata clay pellets, and test MGR4 and MGR5 with FoCa clay pellets. The last two tests have been performed with the pellets/powder proportion finally chosen for the shaft sealing test. The main characteristics and results of these tests, whose detailed description is given bellow, are summarised in Table II.

Table II: Summary of the tests performed in the big oedometer

Reference	Clay	pellets/powder	ρ_d g/cm^3	w %	Hydration/P (MPa)	Duration hours	P_s MPa	K 10^{-13} m/s
MGR2	Serrata	62/38	1.40	11.0	Both/0.6	800	2.2	1.3
MGR3	Serrata	70/30	1.45	13.6	Bottom/0.6	3930	1.7	2.2

Reference	Clay	pellets/powder	ρ_d g/cm^3	w %	Hydration/P (MPa)	Duration hours	P_s MPa	K 10^{-13} m/s
MGR4	FoCa	50/50	1.54	8.2	Bottom/0.005	6240	1.8	
MGR5	FoCa	50/50	1.60	10.2	Bottom/0.005	2327	3.7	5.3

Test MGR2

This test took place with a 61/38 % pellets/powder mixture of Serrata clay. The pellets belonged to the first manufacturing set and had an initial water content of 9.1% and the powder was Serrata clay, grounded to a size less than 5 mm, with a water content of 14.2%. The resulting water content was 11.0 % and the dry density was 1.40 g/cm^3. It was saturated with tab water by both faces under a water pressure of 0.6 MPa. As a high saturation pressure was used from the beginning, most of the water entered in a few hours. The evolution of water intake and swelling pressure is shown in Figure 4. Saturation has been reached after approximately 800 hours (final water content: 35.9%), although the total duration of the saturation phase was 1262.5 hours. The resulting swelling pressure was 2.2 MPa (after subtracting 0.6 MPa of injection pressure), that is higher than the saturated swelling pressure expected for a compacted specimen of granular clay of the same dry density tested in a standard oedometer (1.5 MPa).

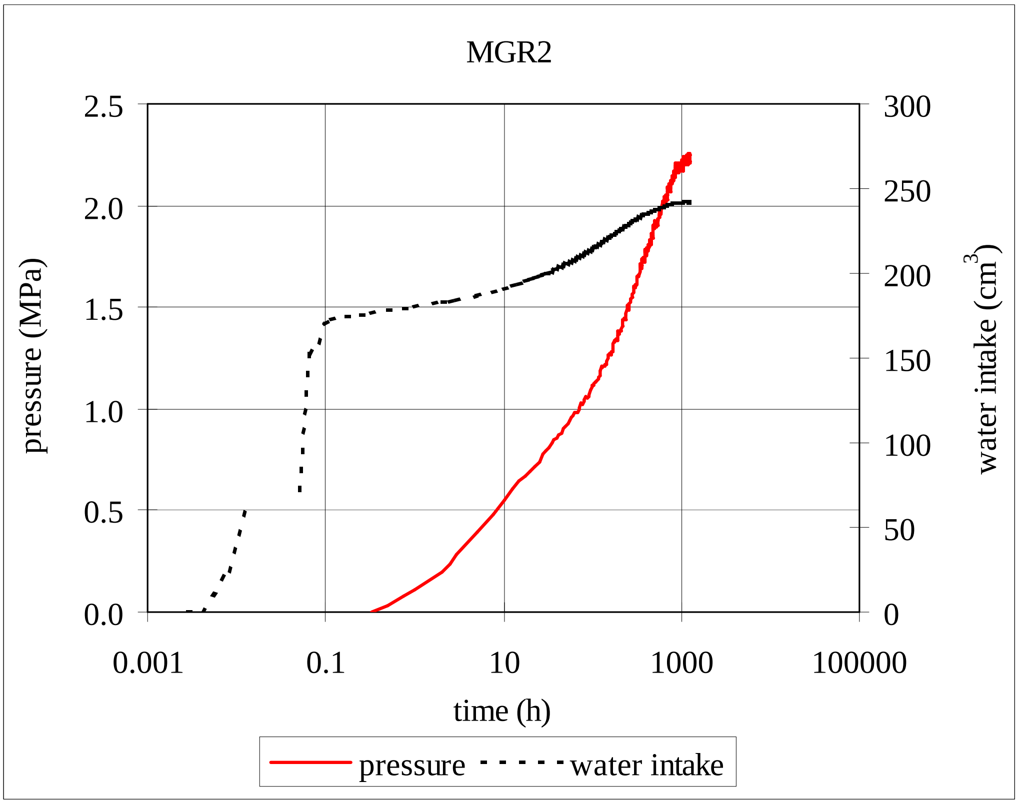

Figure 4: Water intake and swelling pressure evolution in test MGR2

Afterwards, the injection pressure at the bottom of the cell was increased consecutively to three different values, while the water outflow was measured. The hydraulic conductivity was calculated by applying Darcy´s law, giving an average value of $1.3 \cdot 10^{-13}$ m/s, which is in the order of the value obtained for compacted specimens of granular clay of the same dry density ($1.6 \cdot 10^{-13}$ m/s).

The aspect of the clay block after extraction was homogeneous and no pellets could be identified. Figure 5 shows the final distribution of water content and dry density, determined by oven drying and mercury displacement respectively.

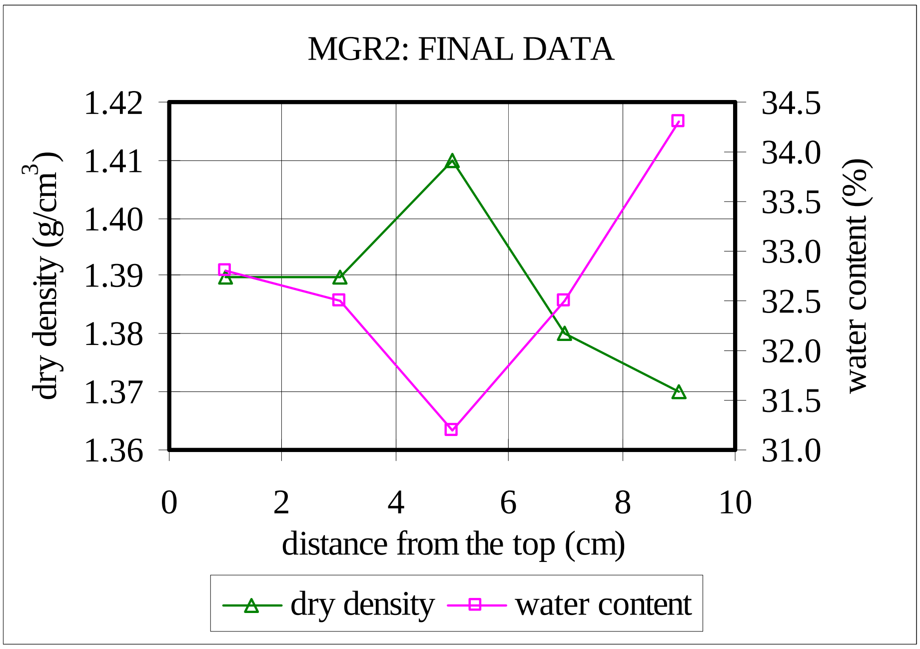

Figure 5: Final water content and dry density at different positions in test MGR2

Test MGR3

250 kg of Serrata clay pellets manufactured by Sahut-Conreur (Raismes, France) and belonging to the second fabrication set were received in CIEMAT. They were used for the third tests in the big oedometer cell. A mixture of 70 % pellets of Serrata clay with initial water content of 13.7 % and 30 % of Serrata clay powder with initial water content of 13.5 % was used. The dry density of the pellets was 1.84 g/cm^3. The resulting water content was 13.6 % and the dry density 1.45 g/cm^3. It was saturated by the bottom face with tab water at a water pressure of 0.6 MPa during 3930 hours. Full saturation was reached in about 3280 hours (Figure 6). It has been observed that when the water injection is stopped, the pressure value registered by the load cell decreases instantly 0.3 MPa, and then, pressure dissipates slowly. So, the swelling pressure after saturation is considered to be between 1.5 and 1.8 MPa (after subtracting the water injection pressure), although it has not reached an equilibrium value and could have been higher if the test had continued. This could be an explanation of having a swelling pressure lower than in test MGR2, in which the dry density was inferior; as in MGR2 swelling pressure had reached an equilibrium value in a shorter time, due to the fact that saturation took place by both faces.

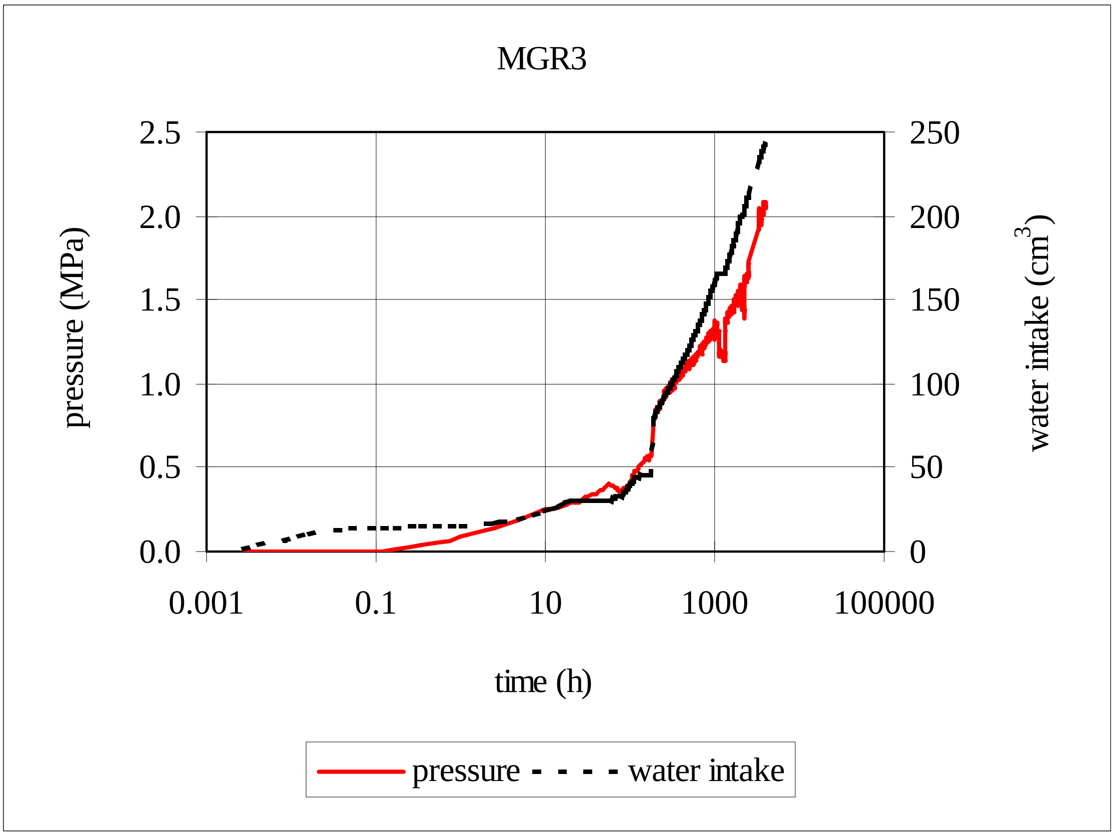

Figure 6: Evolution of the water intake and of the pressure registered by the load cell in test MGR3 performed with Serrata clay

Afterwards, the hydraulic conductivity was measured by applying a hydraulic gradient, giving a value of $2.2 \cdot 10^{-13}$ m/s for a dry density of 1.45 g/cm^3. Subsequently, consolidation at 4 MPa and 5 MPa took place, with measurement of the water outflow and deformation evolution. A final dry density of 1.46 g/cm^3 was attained after 912 hours of consolidation at 5 MPa. Hydraulic conductivity was measured again, giving a value of $8.2 \cdot 10^{-14}$ m/s. Both the swelling pressure and the permeability values are in the order of those values obtained for compacted specimens of granular clay. The sample was extracted at the end of the test, fully saturated, and no pellets could be seen. The final water content was 32.8 %, and the final dry density and water content distribution is shown in Figure 7.

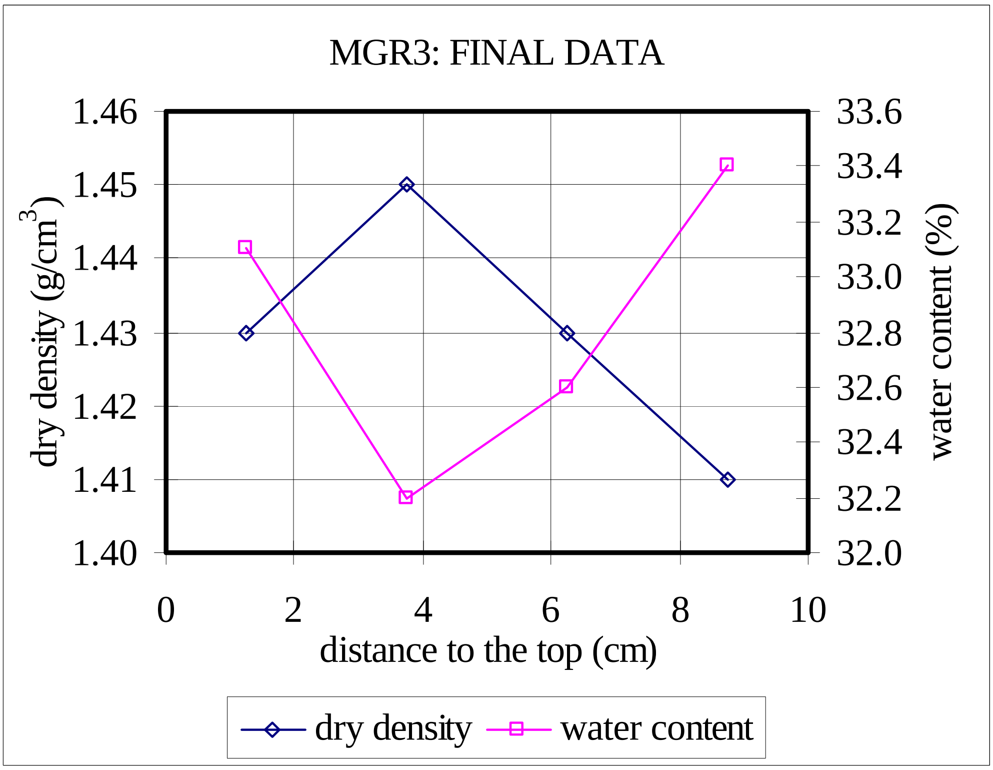

Figure 7: Final water content and dry density at different positions in test MGR3

Test MGR4

Pellets of the French clay FoCa were received in CIEMAT on January 1998. Its average dry density was 2.02 g/cm^3 and its water content 8.2 %. Powder has been obtained by grinding the pellets. For test MGR4 in the big oedometer a mixture 50/50 pellets/powder has been used, with a resulting dry density of 1.54 g/cm^3. It has been saturated by the bottom face with distilled water at a pressure of 0.005 MPa, while pressure exerted by the clay and water intake were measured (Figure 8). The duration of the test was 6240 hours, and full saturation was reached, with a final water content of 30.3 % and a swelling pressure of 1.8 MPa. After 3000 hours, when the water uptake started to stabilise, the swelling pressure began to increase in a sharper way. In fact, although the water uptake seems to stabilise at the end of the test, swelling pressure does not, what suggest certain microstructural organisation that gives place to further development of swelling pressure.

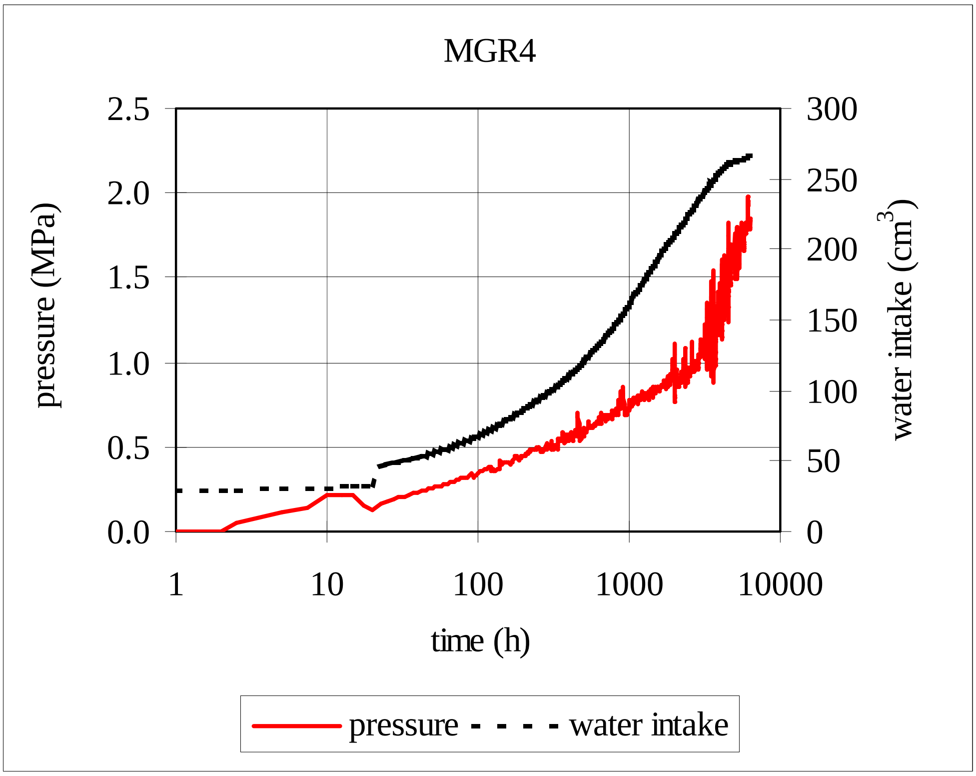

Figure 8: Evolution of the water intake and of the pressure registered by the load cell in test MGR4 performed with FoCa clay

The final distribution of dry density and water content is shown in Figure 9.

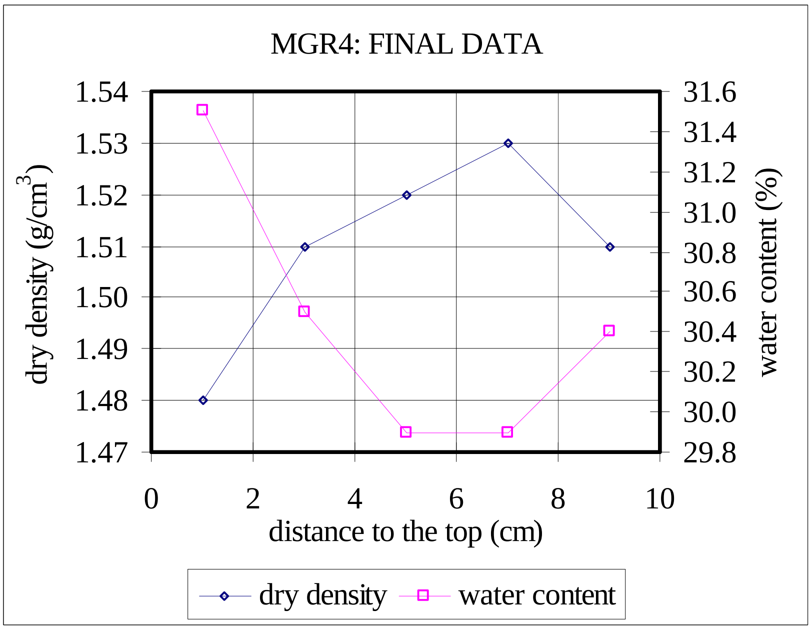

Figure 9: Final water content and dry density at different positions in test MGR4

Test MGR5

For test MGR5 in the big oedometer a mixture 50/50 pellets/powder has been used, with a resulting dry density of 1.60 g/cm^3 and water content of 10.2 %. It has been prepared following the same protocol than that followed by CEA, the height of the sample being 5 cm.

At the beginning, the intrinsic permeability of the dry mixture has been measured with nitrogen gas, giving a value of 2.1·10^{-13} m^2.

Afterwards, it has been saturated by the bottom face with distilled water at a pressure of 0.005 MPa, while pressure exerted by the clay at the top surface and water intake were measured (Figure 10). The duration of hydration was 2327 hours, and full saturation was reached, with a final water content of 28.3 % and a swelling pressure of 3.7 MPa. After 100 hours of hydration, the development of swelling pressure seems to stabilise, but after 300 hours, it starts to increase again. These two episodes in the development of swelling pressure could correspond to the saturation of the powder and subsequently, of the pellets. This behaviour was not appreciated in test MGR4 probably due to the higher height of the sample, that causes the overlapping of both processes.

By increasing the hydration pressure at the bottom of the sample, while measuring the water outcome at the top, the hydraulic conductivity has been calculated. An increase of the injection pressure of 1.5 MPa gave rise to an increase in the registered swelling pressure of almost 1 MPa. The hydraulic conductivity value obtained, for a dry density of 1.59 g/cm^3, is 5.3·10^{-13} m/s.

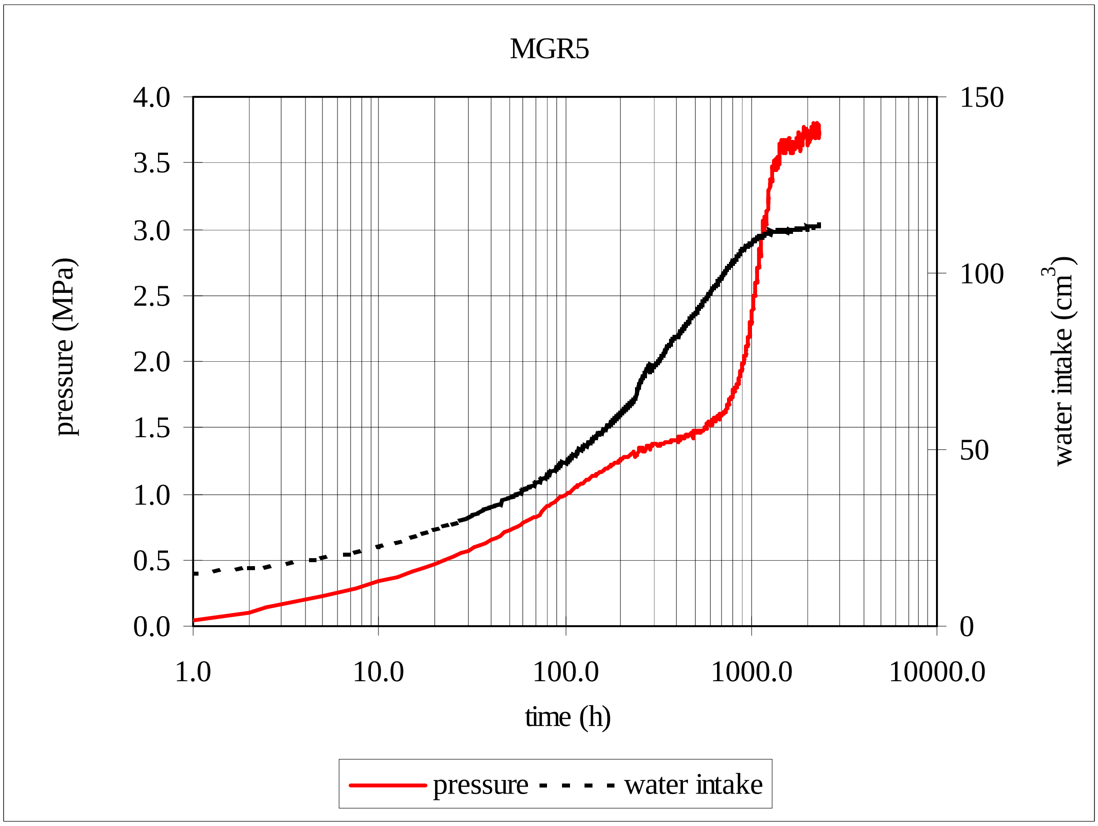

Figure 10: Evolution of the water intake and of the pressure registered by the load cell in test MGR5 performed with FoCa clay

The final distribution of dry density and water content is shown in Figure 11. Pellets were not appreciable.

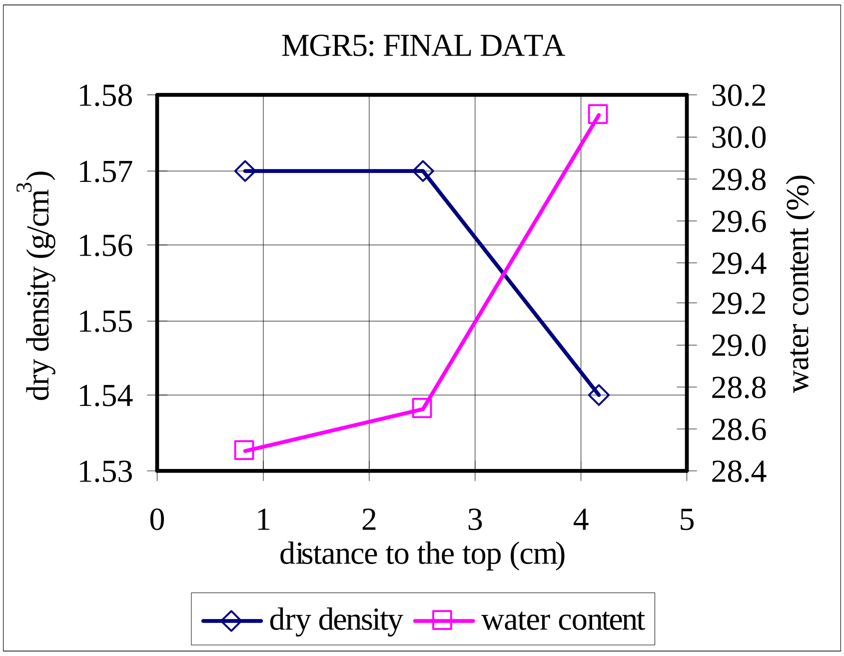

Figure 11: Final water content and dry density at different positions in test MGR5

3.2.2 Retention curves at 20 °C

The retention curves have been determined on compacted samples of Serrata clay and of FoCa clay and on pellets of Serrata clay. The samples have a height of 1.2 cm and a diameter of 1.5 cm, and they have been uniaxially compacted with the water content at equilibrium with the laboratory conditions. The test consists on submitting the samples to a given suction until their water contents reach an equilibrium value. This process takes around 1 month. Once the equilibrium is achieved, the samples are weighted and measured, to determine their water content and dry density. In the case of pellets, the dry density is determined by mercury displacement. Afterwards, the sample is submitted to a different suction and the process is repeated. All the process takes place at a temperature of 20 °C. As the retention curves have been determined for the sealing material, only wetting paths have been followed.

Suction is imposed, but not measured, as there are not conventional techniques to measure high suction in expansive materials. To impose the suction two different methods have been used: control of the relative humidity by means of sulphuric acid solutions (Esteban & Sáez, 1988), and the axis translation technique in membrane cells (Escario & Sáez, 1973). In both cases the suction (s) is imposed to the sample by changing the values of air pressure (u_a) or of water pressure (u_w) in the pores, as suction changes are given by the difference between both ($s=u_a-u_w$). In the first technique, the sample is placed in an atmosphere in which the relative humidity is controlled by means of a sulphuric acid dissolution of known water activity. This relative humidity is correlated to suction through the Kelvin law. Suctions from 4 to 500 MPa can be obtained with sulphuric acid solutions. In the membrane cells, suction is applied by changing the pressure of the gas phase in the pores of the sample by injecting nitrogen in the cell to the desired pressure. At the same time, the sample is in contact with water at atmospheric pressure through a membrane permeable to water but not to air. Due to the mechanical limitations of the cell, only suctions below 14 MPa can be applied (Villar & Martín 1996).

Serrata clay

The retention curve at 20 °C was determined for Serrata clay samples following a wetting path from the "as compacted" condition (uniaxial compaction: ρ_d=1.75 g/cm^3; w=14.2%; s=130 MPa) down to 0.1 MPa, through 10 suction steps (ENRESA 1999). The evolution of water content (w, %) as suction (s, MPa) decreases can be described by the following relationship:

$$w = -5.0 \ln(s) + 38.3 \qquad (r^2=0.98, 30 \text{ points})$$

and, as the determination is performed at free volume, the sample swells as it takes water and the dry density decreases. A potential relationship has also been found between suction (s, MPa) and dry density (ρ_d, g/cm^3). For a wetting path between 148 and 0.1 MPa the fitting obtained is:

$$\rho_d = 1.14\, s^{\,0.08} \qquad (r^2=0.99, 30 \text{ points})$$

These curves have been determined also for Serrata clay pellets, following a wetting path, from suction 130 to 0.1 MPa. The initial dry density of the pellets was 1.78 g/cm^3 and its water content 13.7 %. The average values obtained are shown in Table III and the single values are plotted in Figure 12, with the fitting found for compacted samples.

Table III: Average values obtained in a wetting path for pellets of Serrata clay

suction MPa	ρ_d g/cm^3	w %	S_r %
118.5	1.78	13.7	71
90.9	1.71	14.9	70
28.4	1.52	20.0	70
7.4	1.38	26.2	74
2.0		36.3	
1.0	1.18	38.5	80
0.5	1.16	40.4	82
0.1		53.0	

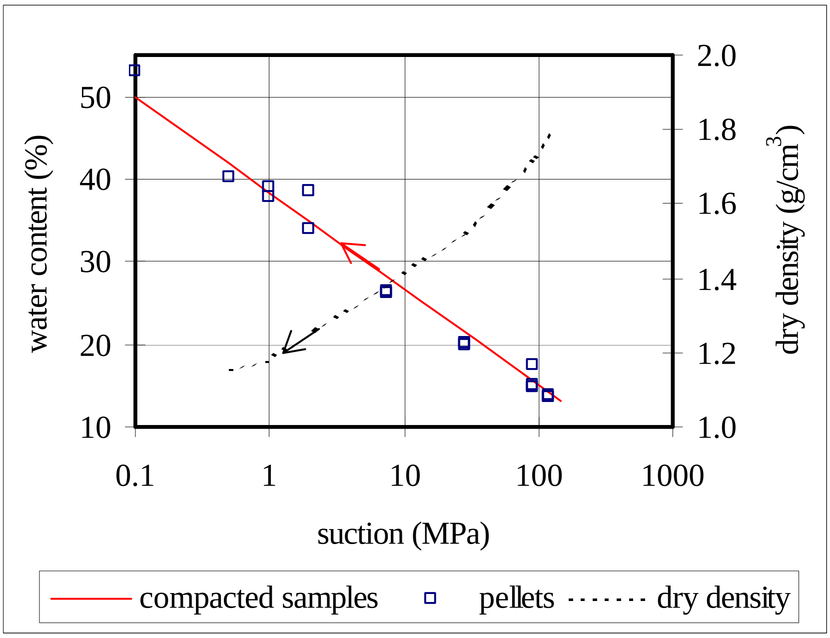

Figure 12: Values of water content and of dry density of pellets of Serrata clay following a wetting path and fitting obtained for compacted samples

No significant differences have been found between the values of water content for a given suction of pellets and of compacted samples. A logarithmic relationship has been found between water content (w, %) and suction (s, MPa) during the wetting process from 120 to 0.1 MPa at free volume, that can be described by the following expression:

$$w = -5.3 \ln(s) + 38.5 \qquad (r^2=0.98, \text{ 21 points})$$

The dry density decreases during the wetting process according to the following expression:

$$\rho_d = 1.19\ s^{0.08} \qquad (r^2=0.90, \text{ 20 points})$$

As a consequence of this dry density variation, there is not a good correlation between suction and degree of saturation, as the density decrease makes the degree of saturation remain almost constant.

FoCa clay

The retention curve at 20 °C has been determined for FoCa clay samples following a wetting path from the “as compacted” condition (uniaxial compaction: ρ_d=1.65 g/cm^3; w=10.3 %; s=140 MPa) down to 0.1 MPa, through suction steps. The average values obtained are shown in Table IV and the single values are plotted in Figure 13. The evolution of water content (w, %) as suction (s, MPa) decreases, can be described by the following relationship:

$$w = -5.0 \ln(s) + 36.1 \qquad (r^2=0.98, \text{ 21 points})$$

and, as the determination is performed at free volume, the sample swells as it takes water and the dry density decreases. A potential relationship has also been found between suction (s, MPa) and dry density (ρ_d, g/cm^3). For a wetting path between 140 and 1 MPa the values obtained are:

$\rho_d = 0.98\ s^{0.10}$ (r^2=0.98, 19 points)

Table IV: Average values obtained in a wetting path for compacted samples of FoCa clay

suction MPa	w %	ρ_d g/cm^3	S_r %
144.3	11.3	1.62	47
74.3	14.6	1.49	49
31.6	18.8	1.37	53
14.0	23.0	1.25	54
4.6	28.0	1.13	55
2.0	34.9	1.01	57

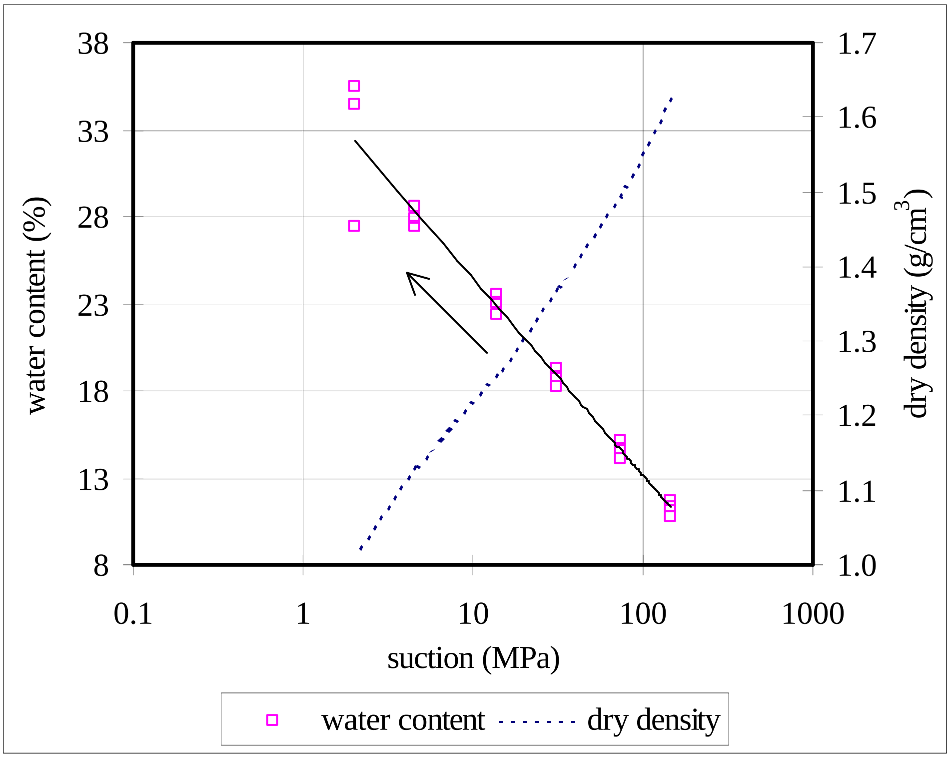

Figure 13: Values of water content and of dry density for compacted samples of FoCa clay following a wetting path (initial dry density 1.65 g/cm^3)

These values can be fitted to the Van Genuchten expression, that relates effective volumetric water content to suction:

$$q_e = \left[\frac{1}{1+(ah)^{1/_{1-m}}}\right]^m$$

where θ_e is the effective volumetric water content, h is the water head, expressed in MPa, α is a parameter related to the soil air entry value and m is a parameter related to the residual water content. To calculate the effective volumetric water content a value of 1.00 has been taken for the saturated volumetric water content and of 0.07 for the residual volumetric water content.

The fittings obtained for the wetting paths are plotted in Figure 14 for samples of FoCa clay compacted to an initial dry density of 1.65 g/cm^3 and for samples of Serrata clay compacted to an initial dry density of 1.67 g/cm^3 (the experimental values for Serrata clay can be found in ENRESA 1999). For FoCa clay α takes a value of 295 MPa^{-1} and m of 0.13, while for Serrata clay the values obtained are 42 MPa^{-1} and 0.14, respectively. It has to be taken into account that the Van Genuchten expression was derived for samples whose volume does not change during hydration, which is not the case of these experiments.

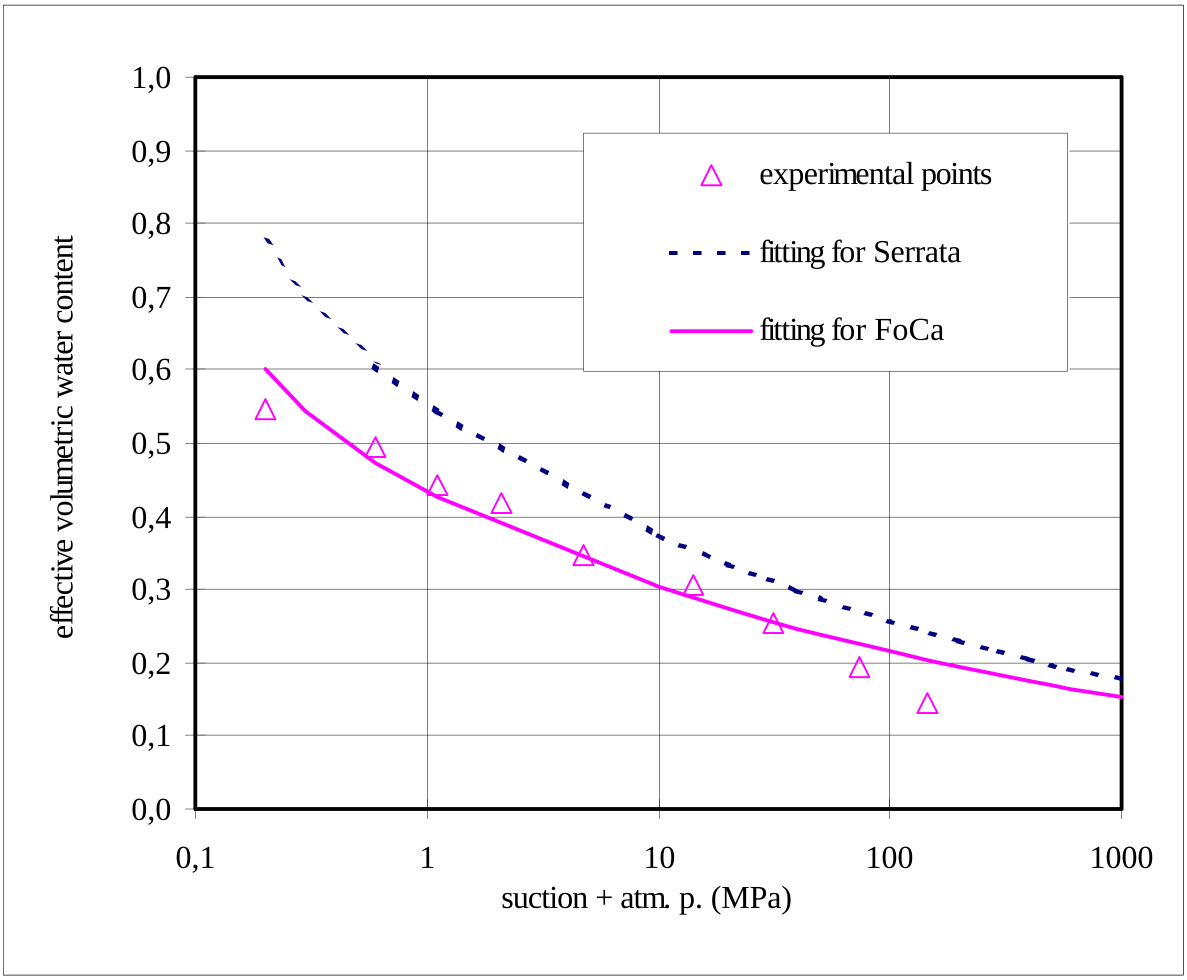

Figure 14: Van Genuchten fitting for the experimental points obtained with compacted samples of FoCa clay (initial dry density 1.65 g/cm^3) and of Serrata clay (initial dry density 1.67 g/cm^3) in wetting paths

The relationship between suction and water content for samples of FoCa clay kept at constant volume, has been determined in suction controlled oedometers, in which it is possible to apply the appropriate load to avoid swelling. A wetting path has been followed, the initial dry density is 1.65 g/cm^3, the suction range is from 130 to 0 MPa for test EDS2_11 and from 14 to 0 MPa for test EDN2_13. Test EDN2_13 continues with a drying path. The method allows also the determination of the swelling pressure for every suction step, as the density of the sample is kept "approximately" constant, between 1.65 and 1.60 g/cm^3 for the lower suctions.

If this dry density diminution is not taken into account, fictitious values of the degree of saturation greater than 1 are obtained. The results are shown in Table V.

Table V: Evolution of water content and swelling pressure during a confined wetting path for a sample of FoCa clay compacted to dry density 1.65 g/cm^3

Test EDS2_11				Test EDN2_13			
Suction MPa	w %	S_r %	P_s (MPa)	Suction MPa	w %	S_r %	P_s (MPa)
130	8.9	38	0.0	130	10.3	45	0
127	9.0	39	0.2	14	22.0	94	0.3
62	13.4	58	1.1	8	24.2	100$^+$	1.1
32	17.0	73	2.2	5	24.2	100$^+$	3.7
14	20.1	87	2.5	3	24.7	100$^+$	5.3
4.2	24.5	100*	2.6	1.5	24.2	100$^+$	7.6
1.5	25.8	100*	2.6	0.5	25.1	100$^+$	7.8
				0.1	24.7	100$^+$	7.8
				0	24.7	100$^+$	8.4

* dry density 1,61-1,58 g/cm^3

$^+$ dry density 1,62-1,60 g/cm^3

Figure 15 represents the values obtained in these two tests plus those obtained in the retention curve determined at constant volume that has been shown above (the drying path of test EDN2_13 is not represented in it). The water content values attained by the compacted clay for a given suction are dependent on the stress situation of the clay. When vertical load is applied and the swelling hindered, the retention capacity of the clay decreases, and the water content reached at the equilibrium is lower. This effect becomes more significant as suction becomes lower, specially bellow 10 MPa.

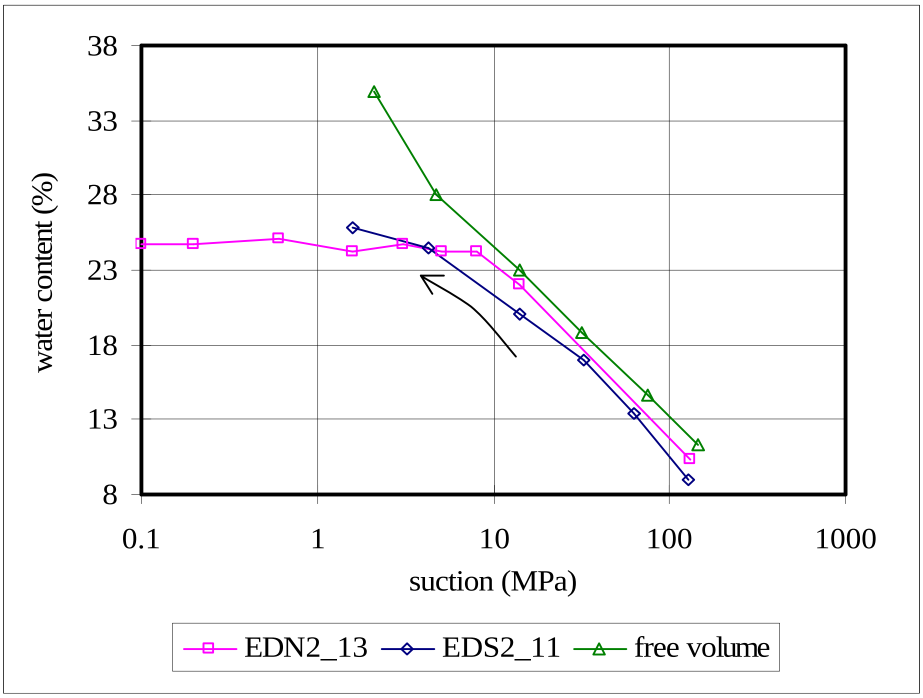

Figure 15: Suction/water content relationship in a wetting path for compacted samples of FoCa clay of constant dry density 1.65 g/cm^3 (EDN2_13 and EDS2_11) and for samples of initial dry density 1.65 g/cm^3 allowed to swell during the hydration process

The results of test EDS2_11 can be fitted fairly well to the Van Genuchten expression shown bellow, with a value of 26 MPa for P_w and of 0.36 for the parameter λ_w. For the low suction range anomalous high values of degree of saturation have been obtained, probably due to some not detected swelling of the clay, that would have brought the dry density to 1.60 g/cm^3. Figure 16 shows the values experimentally obtained and the Van Genuchten fitting.

$$S_r = \frac{1}{\left(1+\left(s/P_w\right)^{\frac{1}{1-\lambda_w}}\right)^{\lambda_w}}$$

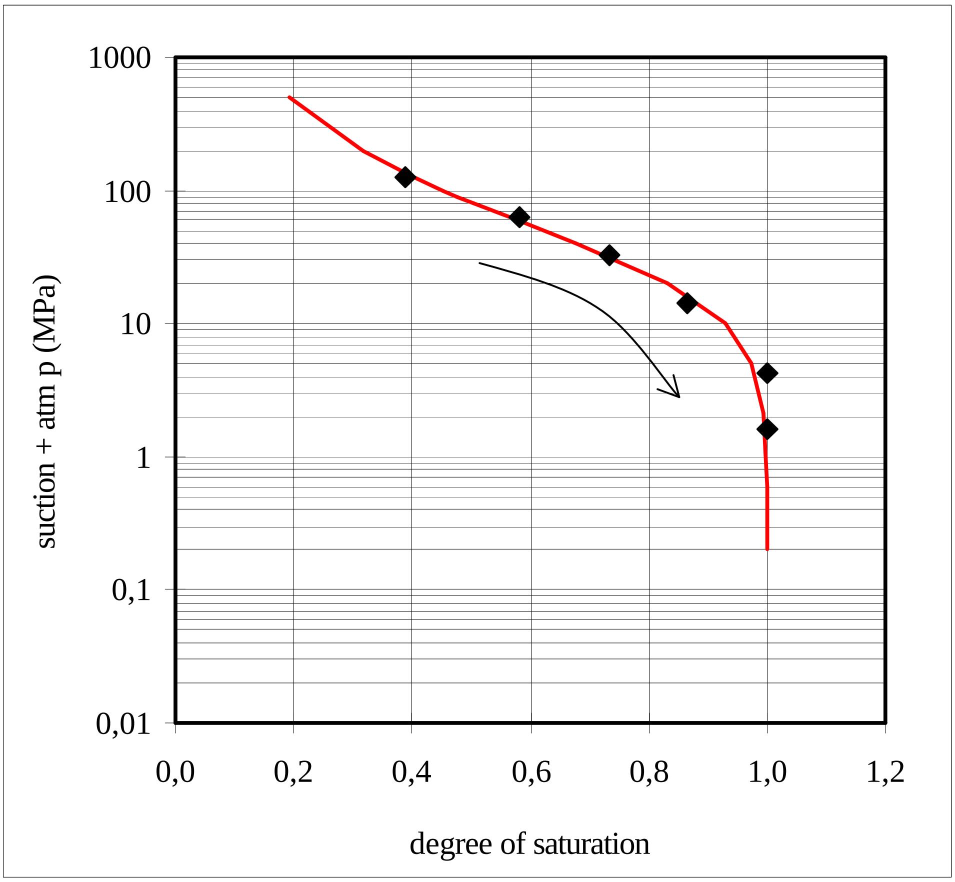

Figure 16: Van Genuchten fitting for the results of test EDS2_11: wetting path for a sample of FoCa clay compacted to dry density 1.65 g/cm^3

3.2.3 Suction controlled oedometer tests

The tests have been performed in oedometer apparatus equipped with cells specially designed to apply the desired suctions, described in Villar & Martín (1996). Suction is controlled using one of the two techniques described in section "Retention curves": control of the relative humidity by means of sulphuric acid solutions, and the axis translation technique in membrane cells. Suctions from 13 to 500 MPa can be obtained with sulphuric acid solutions. Due to the mechanical limitations of the membrane cells, only suctions below 14 MPa can be applied in them.

Vertical loads of up to 5 or 9 MPa, depending on the equipment, can be applied. The samples are 1.2 cm in height and 4.95 or 3.81 cm in diameter. The tests have been performed at 20 °C.

The bentonite has been compacted in the oedometer rings to initial nominal dry densities of 1.70 g/cm^3 by applying a uniaxial load of about 20 MPa. The initial water content of the clay is the equilibrium one at laboratory conditions.

The stress paths of the tests were proposed by the modeller team. Loading and wetting paths have been followed, and in each of them the modification of vertical load or of suction is performed in a stepwise way. The paths followed have been of changing suction under constant vertical load, or of changing vertical load under constant suction. The time taken to stabilise the deformation under a given step is longer than 20 days. The same stress path has been followed in nitrogen oedometers (tests EDN) and in sulphuric oedometers (tests EDS), as the range of suctions that can be applied in both is different. Besides, the same stress path has been sometimes performed in equipments with different vertical load capacity, giving rise to two different oedometric test.

The stress paths of the tests performed in sulphuric acid cells, in which the maximum suction available is higher, are plotted in Figure 17.

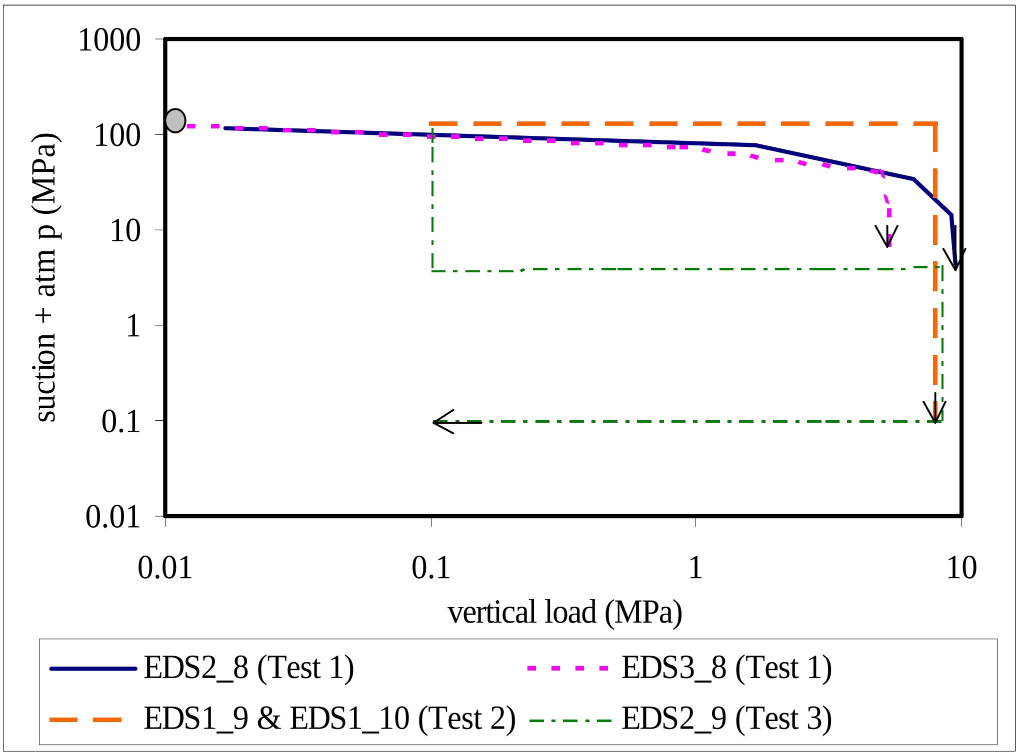

Figure 17: Stress paths followed in the sulphuric acid cells, with indication of the initial conditions (circle)

The detailed description of every test is given bellow:

- Test EDS2_8 (corresponding to Test 1 of UPC): Stabilisation at initial compaction conditions (s=130 MPa) and swelling pressure test at progressively decreasing suctions down to saturation (final suction: 4.4 MPa). The maximum vertical load available was 9 MPa.
- Test EDS3_8 (corresponding to Test 1 of UPC): Stabilisation at initial compaction conditions (s=130 MPa) and swelling pressure test at progressively decreasing suctions up to saturation (final suction: 4.6 MPa). The maximum vertical load available was 5 MPa. Due to the limitations of the equipment, it has not been possible to counteract the swelling pressure of the clay at the end of both tests.
- Tests EDS1_9 and EDS1_10 (corresponding to Test 2 of UPC). The first one had to be interrupted. They started by stabilisation at the initial compaction conditions under a nominal vertical stress (0.1 MPa). Increase of vertical stress up to the swelling pressure value (determined in Test 1: 9 MPa) under the “as compacted” suction pressure (130 MPa). Wetting stage by controlled suction reduction down to the minimum value available, maintaining the previously reached vertical stress.

- Test EDS2_9 (corresponding approximately to Test 3 of UPC): Stabilisation at initial compaction conditions (s=130 MPa). (The initial compression up to a small vertical stress, proposed by the modellers, was not performed). Wetting stage by controlled suction reduction down to 4 MPa under a vertical load of 0.1 MPa. Increase of vertical stress up to 8.4 MPa under constant suction. Unloading at full saturation.

The stress paths of the tests performed in nitrogen cells are plotted in Figure 18. The higher suction that can be applied in these apparatuses is of 14 MPa, what implies that in the first step of any of these tests, the suction is suddenly reduced from the initia1 value of the compacted sample, which is about 130 MPa, to 14 MPa. Besides, as all the tests start under a vertical load of 0.1 MPa, when the sample is placed in the oedometer, it swells largely and the dry density suffers a significant diminution.

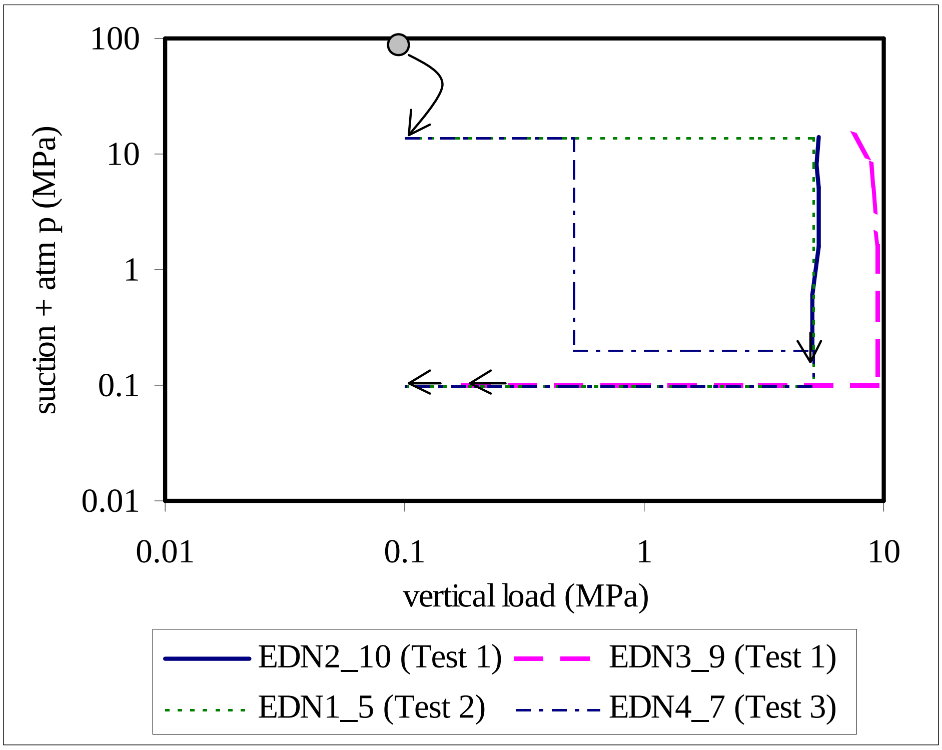

Figure 18: Stress paths followed in the nitrogen pressure cells, with indication of the initial conditions (circle)

- Test EDN3_9 and EDN2_10 (corresponding to Test 1 of UPC): Swelling pressure test at increasing water contents, by decreasing suction from the maximum value available (14 MPa) down to the minimum value available in the apparatus (0 MPa). Due to the limitations of the equipment, it was not possible to counteract the swelling pressure of the clay, and the tests have been in fact wetting tests under a high vertical load.

- Test EDN1_5 (corresponding to Test 2 of UPC): Compression under suction 14 MPa from 0.1 to 5 MPa. Wetting stage by controlled suction reduction down to 0.1 MPa. The subsequent increase of vertical stress proposed by the modellers was not performed, as the maximum vertical load was already reached. Unloading at constant suction 0.1 MPa.
- Test EDN4_7 (corresponding to Test 3 of UPC): Compression test under constant suction (14 MPa) from a vertical stress of 0.1 MPa to 0.5 MPa. Wetting stage by controlled suction reduction down to the minimum value available in the apparatus (0.1 MPa) at

constant vertical stress (0.5 MPa). Increase of vertical stress to the maximum available one in the apparatus (5.0 MPa) under a suction of 0.1 MPa. Unloading at full saturation.

A summary of the results of every oedometer test is presented in the following sections, while more detailed results can be found in Annex I. The final void ratio of each step is plotted against the correspondent suction or vertical load. As both variables have the same units, in some cases, they are represented in the same axis, but paths of changing suction or of changing vertical load are represented with lines of different styles indicated in the legends.

Test 1 in sulphuric acid cells

They are swelling pressure tests for suctions increasingly lower. Due to the limitations of the equipment, it has not been possible to counteract the swelling of the clay for the lower suctions, and the dry density decreased in the last steps. Figure 19 shows the evolution of swelling pressure and of void ratio in both tests.

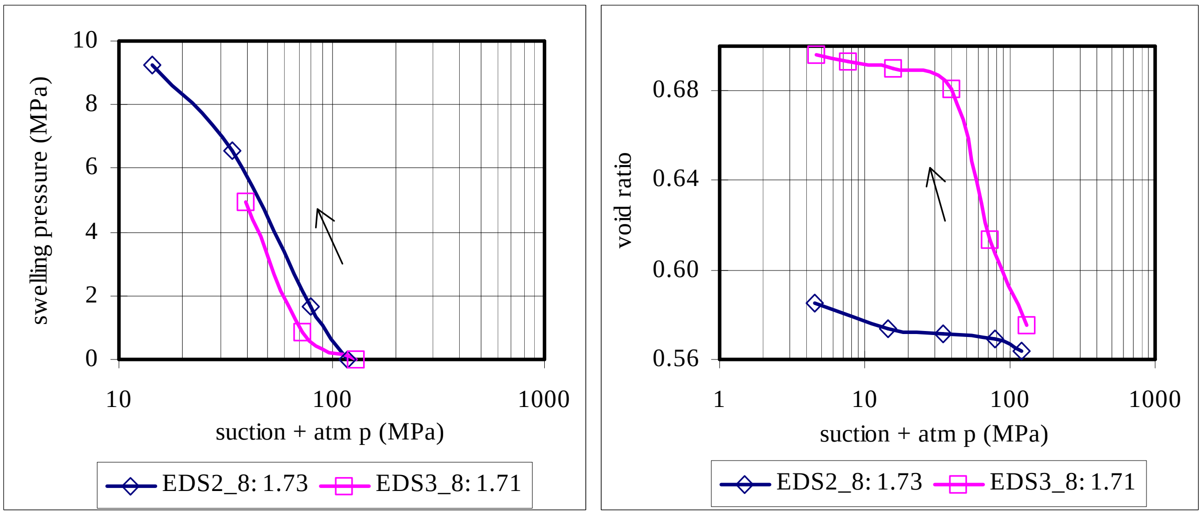

Figure 19: Evolution of the swelling pressure and of the void ratio at decreasing suctions in suction controlled test EDS2_8 and EDS3_8, with indication of the initial dry densities in g/cm^3

Test 2 in sulphuric acid cells

Test 2 type is represented by Test EDS1_9, that was interrupted and replaced by EDS1_10. Keeping constant the suction corresponding to the initial water content (123 MPa), the sample is loaded up to the maximum capacity of the equipment (which is about the swelling pressure value). Figure 20 shows the final void ratio of each load step for both tests, and of the final unloading step of test EDS1_9.

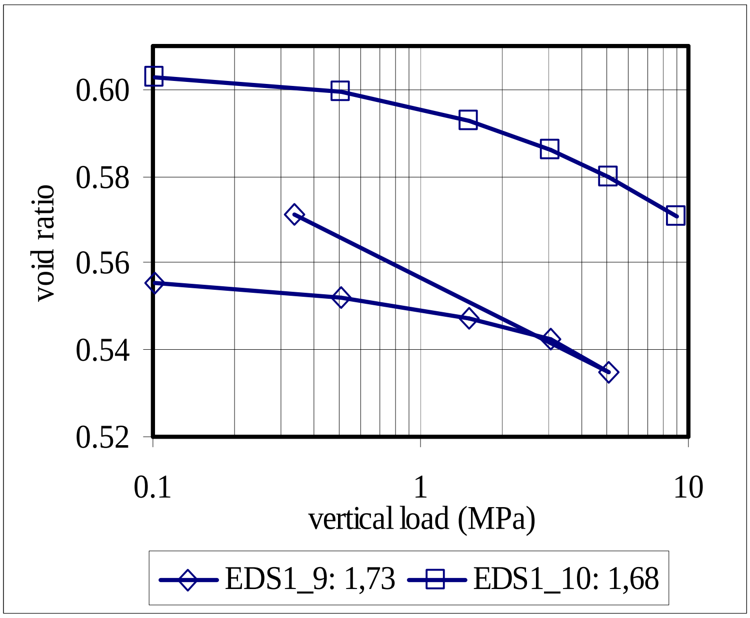

Figure 20: Final void ratios corresponding to the different load steps under constant suction (123 MPa) in tests EDS1_10 and EDS1_9, with indication of the initial dry densities in g/cm^3

Afterwards, under the maximum vertical load, the sample is wetted by decreasing suction (Test EDS1_10, Figure 21). When suction decreases from 15.3 to 5.4 MPa the volume of the sample decreases, but it recovers once full saturation is reached. The collapse observed can be explained as a consequence of the transfers between micro and macrostructure. Under a high vertical load of 9 MPa, a limited quantity of water is able to enter the sample during the wetting process, that, at the beginning, affects only the microstructure. When suction decreases to 15 MPa, the big mesopores start to be affected, and become suddenly saturated, what provokes a loss of its resistance and certain collapse.

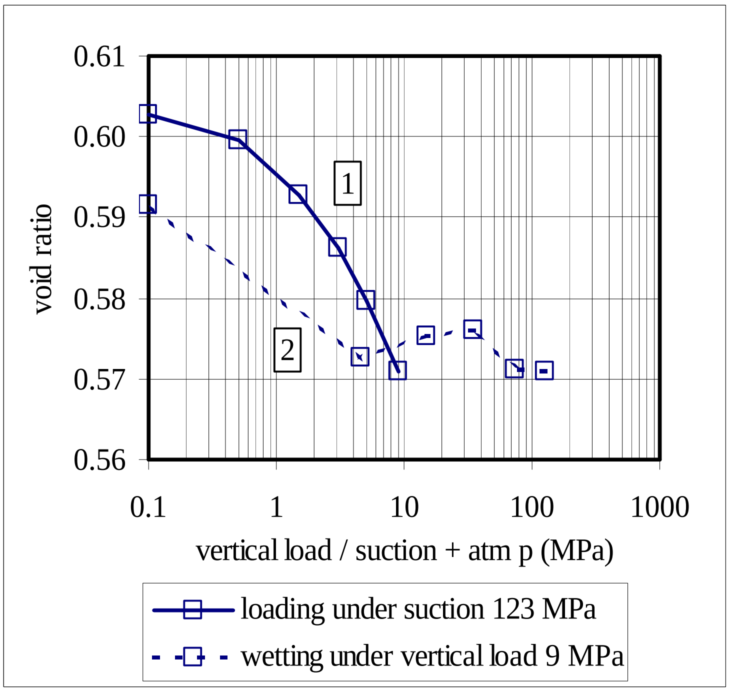

Figure 21: Consecutive void ratios of the loading / wetting test EDS1_10

Test 3 in sulphuric acid cells

This type of test is represented by test EDS2_9. It starts by the wetting of the sample from the initial suction value (130 MPa) to a suction of 5 MPa under a vertical load of 0.1 MPa. The void ratio increases in a logarithmic way during this path, plotted in Figure 22 (1). Afterwards the sample is loaded up to 9 MPa and finally unloaded at full saturation. The evolution of the void ratio in this part of the test is plotted in Figure 22 (2 and 3). It can be observed that the void ratio increase experienced during the initial wetting, was not recovered after loading to a pressure equivalent to the swelling pressure value. It must be recalled that this swelling pressure was determined avoiding the deformation of the sample as it saturates, which is not the case of test EDS2_9.

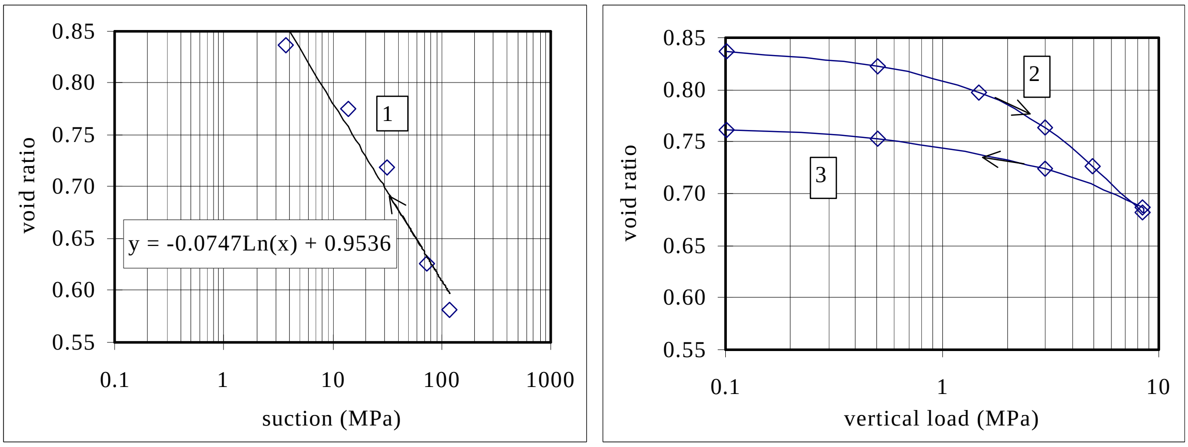

Figure 22: Evolution of the void ratio during wetting under vertical load 0.1 MPa (1); loading under suction 4 MPa (2) and unloading at full saturation (3) in test EDS2_9

Test 1 in nitrogen cells

It is a swelling pressure test under suctions increasingly lower. In test EDN3_9, the loading capacity of the equipment (9 MPa) was surpassed for suctions lower than 1.5 MPa. For this reason the void ratio increased with respect to the initial value. The results obtained are plotted in Figure 23.

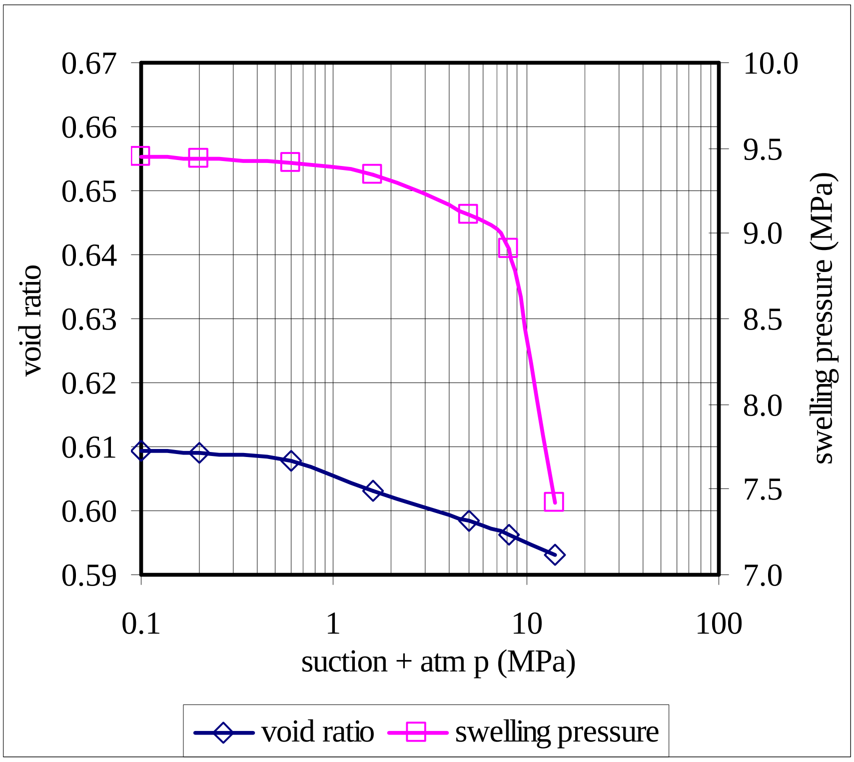

Figure 23: Evolution of swelling pressure and void ratio as suction decreases in suction controlled test EDN3_9

Test EDN2_10 was in fact a test of wetting under constant vertical load, as the maximum vertical load that could be reached in the oedometer was 5.0 MPa, inferior to the swelling pressure of the compacted clay. The evolution of the void ratio, that increases logarithmically as suction decreases, is shown in Figure 24.

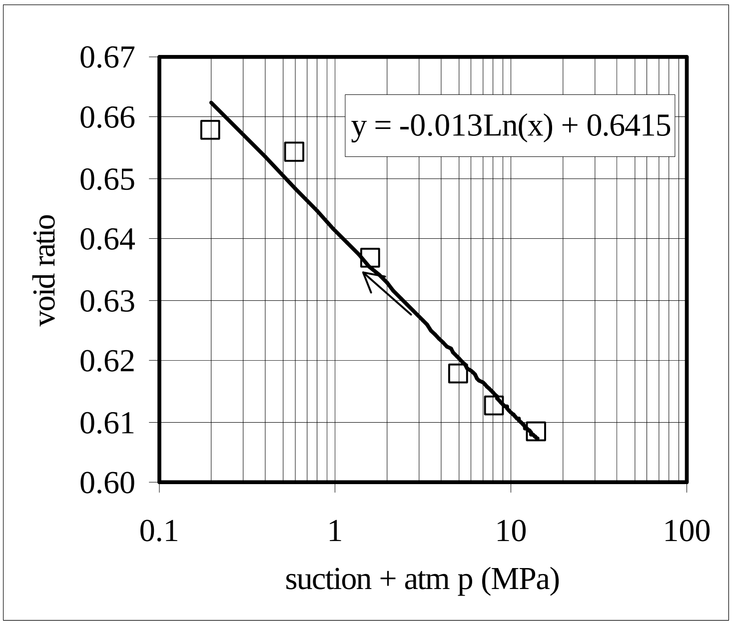

Figure 24: Void ratio at the end of each step of a wetting path performed under a vertical load of 5 MPa (Test EDN2_10)

Test 2 and 3 in nitrogen cells

These types of tests are represented by test EDN1_5 and EDN4_7, respectively. The results of both are presented together for comparison. Figure 25 shows the final void ratio of each step.

Test EDN1_5 and test EDN4_7 have been performed in nitrogen cells and start by the stabilisation of the sample at suction 14 MPa and vertical load 0.1 MPa, what produces an initial density decrease from 1.72 g/cm^3 to 1.50 g/cm^3 in test EDN1_5 and from 1.69 g/cm^3 to 1.51 g/cm^3 in test EDN4_7. In test EDN1_5, the sample is loaded from 0.1 to 5.0 MPa, under a suction of 14 MPa, and finally wetted by suction reduction down to 0 MPa. On the contrary, test EDN4_7 starts by slightly loading the sample up to 0.5 MPa and then the sample is wetted by suction reduction down to 0 MPa. Afterwards, the sample is loaded up to 5 MPa. So, the final condition of both tests is the same, vertical load 5 MPa and full saturation. The void ratio increase provoked by a suction reduction is higher if it takes place under a low vertical load (Test EDN4_7), but the void ratio decrease during consolidation at full saturation is similar to that under a suction of 14 MPa.

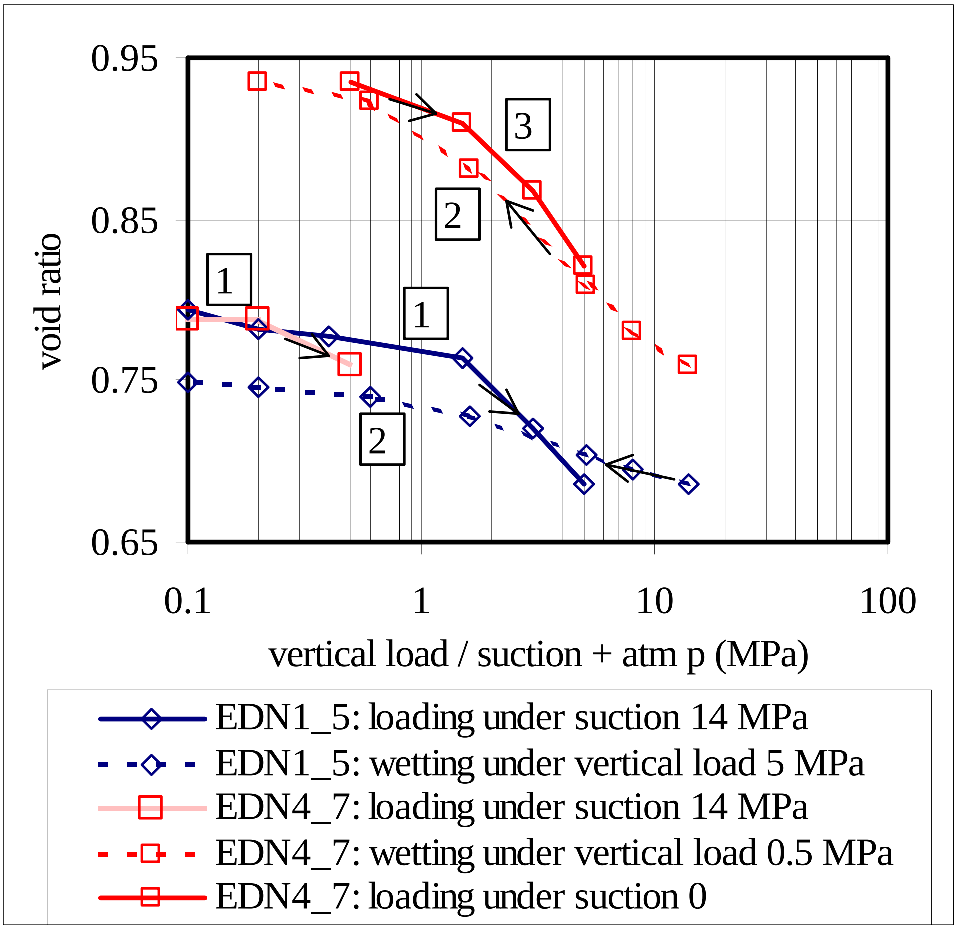

Figure 25: Final void ratio of each step of the loading and wetting paths in tests EDN1_5 and EDN4_7

3.3 Transport properties of the sealing materials

3.3.1 Infiltration tests on Serrata powder/pellets mixture

An infiltration test (CT25) has been carried out in a hermetic cell of 15 cm height and 15 cm diameter, to measure the transport properties of a pellets/powder mixture and, at the same time, verify the behaviour of the humidity sensors. Hydration took place by the bottom surface at atmospheric pressure during the first 600 hours and afterwards at 6 bar. The evolution of the relative humidity registered by the sensors and the actual water content physically measured at the end of the test have been compared and evaluated, as though as the difference between pellets and powder water content and dry density.

The sensors used have been capacitive VAISALA HMP-233, with sinter filters. Sensor 1 was placed between 8.0 and 11.0 cm from the hydration front and sensor 2 between 5.5 and 8.0 cm from the hydration front.

The mixture was 67 % pellets and 33 % powder of Serrata clay. The initial dry density of the pellets was 1.84 g/cm^3 and its water content 13.7 %. The powder was oven dried, so the average water content of the mixture was 8 %, and its dry density 1.54 g/cm^3.

A calibration curve was obtained for these sensors by measuring the effective gravimetric water content of compacted blocks in which the sensors had been previously placed. The relative humidity registered by the sensors (RH, %) can be converted to water content of the clay (w, %) through the equation:

$$w = 6.45 \times \ln\left(\frac{3.06}{1 - \frac{RH}{100}}\right)$$

Figure 26 shows the evolution of the relative humidity registered by both sensors and of the water contents calculated according to the calibration curve, and the evolution of the bulk water intake with time.

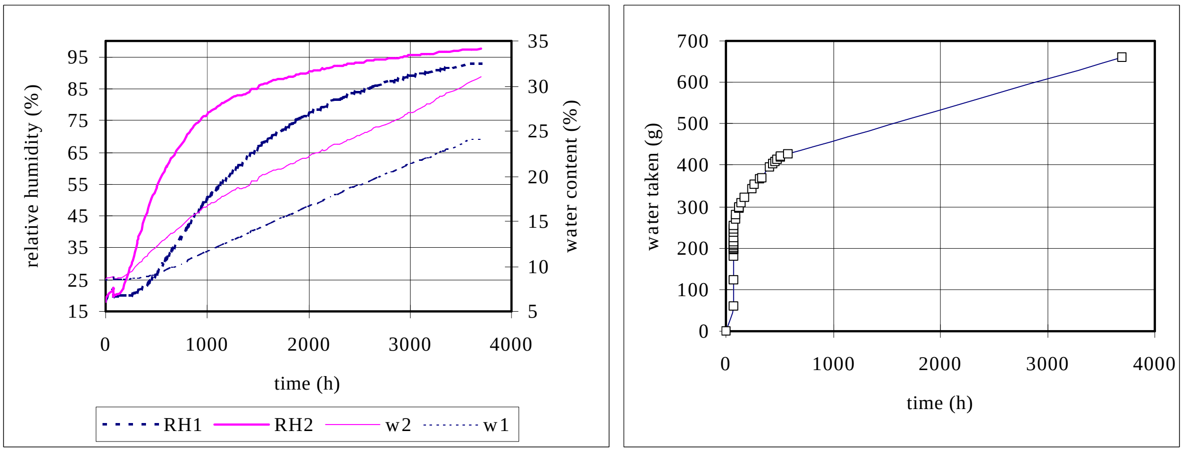

Figure 26: Evolution of the relative humidity registered by sensors 1 and 2 and of the water contents calculated through the calibration curve. Evolution of the bulk water intake

At the end of the test the clay sample was extracted and it could be observed that the aspect of the clay was homogeneous in the bottom part and that pellets could be only recognised in the drier upper part. The water contents of pellets and powder have been measured and compared, being the latter higher, which is explained because powder is a preferential hydration pathway (Figure 27).

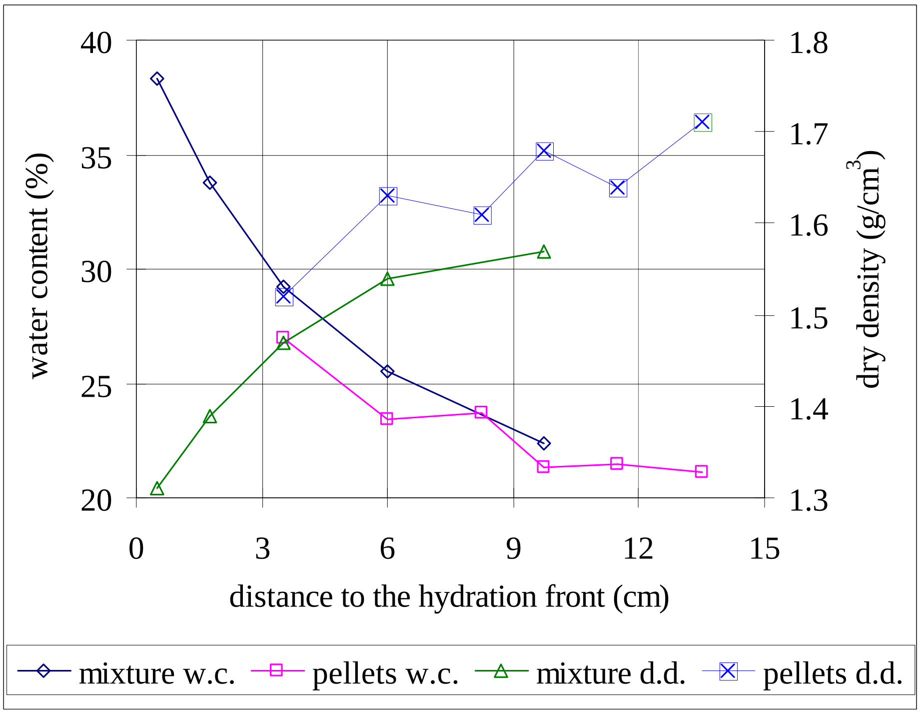

Figure 27: Final water content and dry density of the pellets and of the mixture measured in test CT25

The final measured water contents have been compared to those registered by the sensors. Sensor 1 registered a final relative humidity corresponding according to the calibration curve to a final water content of 24 %, while the measured water content in the zone the sensor was placed was 22 %. Sensor 2 registered a final water content according to the calibration curve of 31 %, while the measured value was 25 %. A possible reason to explain these differences is that the calibration curve used has been obtained in unconfined compacted samples in which the water content is homogeneous. In the case of pellets/powder mixtures, the water content of the powder and of the outer part of the pellets can be much higher than that of the inside of the pellets, and the relative humidity measured by the sensors is not that at the inside of the pellets.

Another possibility to convert the values of relative humidity to water content values is to use the equation (Kelvin's law) that relates relative humidity (RH, %) to suction (s, MPa):

$$s = -10^{-6} \frac{R \times T}{V_w} \ln\left(\frac{RH}{100}\right)$$

where R is the universal constant of gases (8.3143 J/mol·K), T the absolute temperature (295.6 K) and V_w, the molar volume of water ($1.80 \cdot 10^{-5}$ m^3/mol). Suction can be converted to water content (w, %), through the retention curve determined for compacted samples of Serrata clay (cf. section "Retention curves"). This method presents the limitation that the retention curve has been obtained at free volume, and it has been observed that the water content values obtained for the same suction at confined volume (which is the case of test CT25) are much lower than those obtained at free volume. In fact, the values of water content obtained transforming the RH values by this method are higher than those obtained through the calibration curve.

However, a preliminary curve, relating suction to degree of saturation at confined volume, has been obtained for Serrata clay, following the method described in the section “Retention curves” (Villar 1998a). When this curve is used to infer the water content values, instead of the one obtained at free volume, we can obtain a much better fitting between the actual water content values and those deduced from the sensors readings.

4. CONCLUSIONS

Several tests have been performed in order to get a better knowledge of the geomechanical behaviour of the sealing material and of its gas and water transport properties. The Spanish and the French reference materials, Serrata and FoCa clay, have been used. Some of the tests have been performed on pellets/powder mixtures, but not all of them as the available equipment had not the appropriated dimensions.

To study the swelling behaviour of pellets/powder mixtures a special oedometer cell has been designed. It allows the measurement of swelling pressure and water intake as saturation goes on. Hydraulic conductivity can also be determined. The tests have been performed on low density mixtures of Serrata and FoCa clay. It has been observed that the development of swelling pressure does not follow a continuos rate: when the water intake has stabilised, the swelling pressure develops in a faster way, what must be due to a redistribution of water that gives place to the hydration of the pellets, whose dry density, and consequently, swelling pressure, are higher. This makes the equilibrium time very long.

The behaviour of humidity sensors placed on pellets/powder mixtures has been checked by performing an infiltration test on a pellets/powder mixture of Serrata clay. It has been observed that the powder constitutes a preferential way for the water entrance and that, except for the zones completely saturated, in which the water content is homogeneous, the water content of the powder is higher than that of the pellets. When the sample is not yet saturated, the values registered by the sensors are higher than the actual ones. This can be due to the fact that the sensors measure the water content of the powder, as they are not placed inside the pellets.

An experimental set-up has been assembled to measure the gas permeability under low injection pressures of compacted samples with different degree of saturation. The tests have been done on Serrata clay samples and it has been observed a strong difference between the values of intrinsic permeability determined with air or water as permeant for the same dry density. This points to a significant change in the structure of the clay when it is saturated, due to the reduction in mean pore diameter caused by the swelling of the aggregates.

The retention curves have been determined at free volume for compacted samples of Serrata and FoCa clay and for pellets of Serrata clay. No differences have been observed between the values obtained for compacted samples and for pellets. On the contrary, the retention curve determined for compacted FoCa clay at constant volume differs significantly from that determined at free volume. The water content at equilibrium with a given suction is lower when the sample is confined and its swelling hindered.

Suction controlled oedometer tests, following the stress paths proposed by the modellers, have been performed on Serrata clay compacted to an initial dry density of 1.70 g/cm^3. Some trends of behaviour have been identified:

- When wetting takes place under a low vertical load (Tests EDN4_7 and EDS2_9), the void ratio increases largely and the external load applied later is not able to counteract this swelling. However, if swelling during saturation is prevented by previous (EDN1_5) or simultaneous loading (Test EDS2_8 and EDS3_8), the final volume change is smaller. This behaviour may be an example of the irreversible macroscopic deformation induced by the microstructural swelling, whose magnitude (of the plastic deformation) is stress path dependent (Gens & Alonso, 1992).

- The collapse observed in test EDS1_10 can be explained as a consequence of the transfers between micro and macrostructure. Under a high vertical load of 9 MPa, a limited quantity of water is able to enter the sample during the wetting process, that, at the beginning, affects only the microstructure (pores smaller than 20 Å). When suction decreases to 15 MPa, the big mesopores start to be affected, and become suddenly saturated, what provokes a loss of its resistance and certain collapse. But when full saturation is reached, the clay continues swelling, as the microstructure (interlamellar region of expansive minerals) has a higher availability of water coming from the saturated macro and mesopores (Villar 1998b). A similar behaviour was observed in a montmorillonite coming from the same deposit, that slightly collapsed under a vertical load of 5 MPa when suction decreased to 30 MPa (Villar 1995). The initial dry density of this sample was 1.60 g/cm^3, which may explain the lower external load necessary to induce its collapse.

5. ACKNOWLEDGEMENTS

This work has been financed by EC and ENRESA through Contract FI4W-CT96-0025. The laboratory work has been performed by A. Pelayo and R. Campos. P.L. Martín supported the work with sensors. Some results obtained for the FEBEX Project (UE Contract FI4W-CT95-006) have been useful for the development of this work.

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