HYDRAULIC AND MECHANICAL PROPERTIES OF COMPACTED BENTONITE AFTER 18 YEARS IN BARRIER CONDITIONS

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Abstract

The FEBEX "in situ" test was performed at an underground laboratory in Grimsel (Switzerland) with the aim of studying the behaviour of components in the near-field of a nuclear waste repository. A gallery of 2.3 m in diameter was excavated through the granite and two heaters, simulating the thermal effect of the wastes, were placed inside, surrounded by a barrier of highly-compacted bentonite blocks. In 2015, after 18 years of operation, the experiment was dismantled. Some of the bentonite samples taken were tested in the laboratory to characterize, among others, their physical state and determine their permeability and swelling capacity.

There were significant changes in water content and dry density across the bentonite barrier: their distribution was radial around the axis of the gallery, with the water content decreasing from the granite towards the axis of the gallery and the dry density following the inverse pattern.

The swelling capacity of the samples was related to their position in the barrier. In the internal, drier part of the barrier an increase of the swelling capacity with respect to the reference bentonite was detected, whereas the samples from the external part swelled less than expected. This was attributed to the different salinity of the samples. The hydraulic conductivity was mainly related to the dry density of the samples and decreased with respect to the reference bentonite. This decrease was not related to the position of the samples and

could be related to the microstructural reorganisation of the bentonite during the 18-year operation –which brought about an average decrease in the pore size– and to the low hydraulic gradients applied to determine the permeability of the samples retrieved.

1. Introduction

The aim of the FEBEX project (Full-scale Engineered Barriers Experiment) was to study the behaviour of components in the near-field of a repository in crystalline rock according to the Spanish reference concept for geological disposal of nuclear waste. As part of this project an "in situ" test, under natural conditions and at full scale, was performed at the Grimsel Test Site (Switzerland), an underground laboratory managed by NAGRA (the Swiss agency for nuclear waste management). A gallery of 2.3 m in diameter was excavated through the granite and two heaters simulating the thermal effect of the wastes –with dimensions and weighs analogous to those of the real canisters– were placed inside a steel liner installed concentrically with the gallery and surrounded by a barrier of highly-compacted bentonite blocks. The external surface temperature of the heaters was 100°C and the bentonite barrier was naturally hydrated by the granitic groundwater (ENRESA, 2006).

The heating stage of the in situ test began on February 1997. After five years of uninterrupted heating at constant temperature, the heater closer to the gallery entrance (heater #1) was switched off. In the following months this heater and all the bentonite and instruments preceding and surrounding it were extracted. A large number of bentonite samples were also taken for analysis in different laboratories (Villar et al. 2006). The remaining part of the experiment was sealed with a new shotcrete plug and a second operational phase started with the test configuration shown in Figure 1.

The clay barrier was built with compacted bentonite blocks arranged in vertical slices with three concentric rings around the heaters (Figure 2). The thickness of the bentonite barrier in the heater areas was 65 cm (distance from liner to granite). The blocks were obtained by uniaxial compaction of the FEBEX clay with its hygroscopic water content (14%) at pressures of between 40 and 45 MPa, what gave place to dry densities of 1.69-1.70 g/cm^3. The initial dry density of the blocks was selected by taking into account the probable volume of the construction gaps and the need to have a barrier with an average dry density of 1.60 g/cm^3 (ENRESA, 2006).

After 18 years of operation, the FEBEX Dismantling Project (FEBEX-DP) undertook the dismantling of the experiment (García-Siñeriz et al. 2016). Heater #2 was switched off in April 2015, the shotcrete plug was demolished and a month later the buffer removal and sampling started. In particular, samples were taken to determine on site their water content and dry density, with the aim of assessing the final state of the barrier. This was very much affected by the processes to which the barrier had been subjected, namely hydration from the granite and thermally-induced moisture redistribution. Some of the more remarkable observations and common patterns were (Villar et al. 2016a):

– All the construction gaps between blocks had been sealed, both those among blocks of the same section and the gaps between bentonite slices. The granite/bentonite contact was also tight at all locations because of the swelling of the bentonite. This had already been observed in the 2002 partial dismantling.
– The water content and dry density in every vertical section followed a radial distribution around the axis of the gallery, with the water content decreasing from the granite towards the axis of the gallery and the dry density following the inverse pattern. The water content at all points in the barrier, even those close to the heater, was higher than the initial one, i.e. greater than 14%. The water content and density gradients were steeper in those sections affected by the heater.

Numerous bentonite samples were also taken and sent to CIEMAT's facilities. The sampling took place in vertical sections normal to the axis of the tunnel –corresponding to original block slices (Figure 1)–, and in each section several samples, mostly whole blocks, were taken (Figure 2). These samples were tested to characterize their physical state (dry density, water content) and determine, among others, their hydraulic and mechanical properties, such as permeability and swelling capacity (Villar et al. 2018).

The aim of the research reported here was to assess if the operation under repository conditions implied irreversible changes on the main properties of the bentonite. For that, the extensive database on the variation of these properties with different parameters (dry density, water content, temperature, salinity) obtained over the last 20 years for the FEBEX bentonite (e.g. Villar 2002, ENRESA 2006, Castellanos et al. 2008, Villar & Lloret 2008) has been used. .

The same kind of analyses was done in the samples retrieved during the first dismantling in 2002 (Villar et al. 2006, Villar & Lloret 2007). Some of the conclusions reached in those works related to the hydro-mechanical properties of the materials were:

- The hydraulic conductivity of the samples retrieved after 5 years of operation was clearly related to dry density and the latter in turn was related to the position of the sample in the barrier. The values of hydraulic conductivity measured for the samples of lower density (more hydrated) were in the order of those expected for untreated bentonite, but for samples of higher densities there was a large dispersion in the values obtained without any clear tendency.
- The final swelling strains upon saturation under a low vertical load of the samples retrieved after 5 years of operation were in the order of those expected for untreated FEBEX bentonite compacted at the same dry density with the same water content.

Up to the whole dismantling of the FEBEX in situ test, no bentonite subjected to repository conditions for such a long period of time had ever been studied. Nevertheless, other shorter full-scale experiments reproducing different disposal concepts have been dismantled and analysed in a similar way. Some of the older ones were discussed in Villar & Lloret (2007). In the last decade, several experiments in which compacted bentonite was used as engineered barrier were dismantled at the Äspö Hard Rock Laboratory in Sweden – also excavated in crystalline rock–, and the conclusions regarding the HM analyses performed on the samples retrieved can be summarised as follows:

- The "Long Term Tests of Buffer Material" (LOT) included seven test parcels in which MX-80 bentonite was used as barrier material. Two pilot parcels (A1 and S1) were dismantled and analysed after 1 year operation (Karnland et al. 2000), an additional parcel (A0) was exposed to adverse conditions for 1.5 years (Karnland et al. 2011), and a medium-term parcel (A2) was dismantled after 6 years operation (Karnland et al. 2009). No significant changes in the swelling pressure and hydraulic conductivity were detected in any of the parcels, although some trimmed samples showed a decrease in hydraulic conductivity with respect to the reference material not related to the thermal gradient.
- The "Alternative Buffer Material" (ABM) consisted of three packages where eleven different clays were tested as buffers. Package 1 and 2 were dismantled after 2 and 6.5 years, respectively. After 2 years (one of them with the heater set to 130°C) no difference was seen in hydraulic conductivity between samples from the field experiment and the corresponding reference materials. However, a significant decrease in swelling pressure was seen for two of the bentonites used. The largest deviation was noticed on samples from the warmest part during the experiment. The proposed explanation for this decrease in swelling pressure was the significant redistribution of cations in the test package, which could have influenced the physical properties of the clays (Svensson et al. 2011).

- The “Prototype Repository” consisted of six deposition holes divided into two sections (sections I and II, containing four and two holes, respectively). Each deposition hole had a full-scale buffer of compacted bentonite (MX-80) surrounding a copper canister equipped with heaters. Section II was dismantled after 8 years of hydrothermal exposure, and it was found that the degree of saturation was not homogeneous in any of the two buffers (Olsson et al. 2013). No large variations were observed in the swelling pressure of the field-exposed bentonite with respect to the reference bentonite, however the field-exposed bentonite had lower hydraulic conductivity than the reference specimens, especially at high densities. These changes were not related to the position of the samples in the buffer,
- The “Canister Retrieval Test” (CRT) experiment consisted of a cylindrical deposition hole hosting a canister encapsulated in clay buffer (MX-80). After 5 years operation the experiment was shut down and dismantled in 2006. When compared to reference material properties the hydro-mechanical analyses of the buffer samples showed no significant change in swelling pressure, although there was a slight decrease in hydraulic conductivity for the higher density samples. No coupling was found between these changes in the hydro-mechanical properties and the montmorillonite characteristics (Dueck et al. 2011a, b).

2. Material

The bentonite used to construct the engineered barrier was the FEBEX, extracted from the Cortijo de Archidona deposit in SE Spain. The physico-chemical properties of the FEBEX bentonite, as well as its most relevant thermo-hydro-mechanical and geochemical characteristics were summarised in ENRESA (2006).

The montmorillonite content of the FEBEX bentonite is above 90 wt.% (92±3 %). The smectitic phases are actually made up of a smectite-illite mixed layer, with 10-15 wt.% of illite layers. Besides, the bentonite contains variable quantities of quartz (2±1 wt.%), plagioclase (3±1 wt.%), K-felspar (traces), calcite (1±0.5 wt.%), and cristobalite-trydimite (2±1 wt.%). The cation exchange capacity of the smectite is 102±4 meq/100g, the main exchangeable cations being calcium (35±2 meq/100g), magnesium (31±3 meq/100g) and sodium (27±1 meq/100g). The predominant soluble ions are chloride, sulphate, bicarbonate and sodium.

The liquid limit of the bentonite is 102±4 %, the plastic limit 53±3 %, the density of the solid particles 2.70±0.04 g/cm^3, and 67±3 % of particles are smaller than 2 μm. The hygroscopic water content in equilibrium with the laboratory atmosphere is 13.7±1.3 %. The external specific surface area is 32±3 m^2/g and the total specific surface area is about 725 m^2/g.

Based on numerous swelling under load tests performed with the FEBEX bentonite compacted to different initial dry densities (ρ_{d0}, g/cm^3) with different water contents (w_0, %), an empirical relation predicting the final swelling strain (ε, %) after saturation with deionised water under vertical load of 0.5 MPa was determined (Villar & Lloret 2008):

$$\varepsilon = (37.48\ \rho_{d0} - 50.43) \ln w_0 - 154\ \rho_{d0} + 204.24 \qquad [1]$$

The saturated hydraulic conductivity of compacted bentonite samples is exponentially related to their dry density. An empirical relationship for the reference FEBEX bentonite relating hydraulic conductivity (k_w, m/s) to dry density (ρ_d, g/cm^3) was obtained for samples compacted to dry densities above 1.47 g/cm^3 and permeated with deionised water (Villar 2002):

$$\log k_w = -2.96\ \rho_d - 8.58 \qquad [2]$$

The variation in the experimental values with respect to this fitting is on average of 30%, which should be evaluated taking into account that the values of permeability are of the order

10^{-14} m/s. The hydraulic gradients applied in these determinations were on average of 15200 m/m.

3. Methodology

CIEMAT received samples conveniently preserved on site and the packing was only removed just before subsampling in the laboratory. Subsampling from the blocks was mostly performed by drilling with crowns of different diameters.

Samples were taken specifically to determine the water content and dry density of the blocks at two or three positions along the radius of the block, The gravimetric water content (*w*) is defined as the ratio between the mass of water and the mass of dry solid expressed as a percentage. The mass of dry soil was determined by oven drying at 110°C for 48 h. Dry density (ρ_d) is defined as the ratio between the mass of the dry sample and the volume occupied by it prior to drying. The volume of the specimens was determined by immersing them in a recipient containing mercury and by weighing the mercury displaced (applying Archimedes' principle and considering a specific weight for the mercury of 13.6 g/cm^3). The same samples whose volumes had been determined were used for the water content determination.

For the hydro-mechanical tests the samples were drilled at two locations (subsamples 1 and 2, being 1 the closer to the granite) in each block using a crown of internal diameter 5.7 cm. They were later adapted to the diameter of the cell rings with a cylindrical cutter and finally pushed inside the stainless steel cell rings. Care was taken during the process to preserve the original moisture and density of the samples. The identification of each sample was given by the name of the block from which it was drilled (see Figure 2) and the number of the subsample (1 or 2).

Swelling under load tests

The saturation (or swelling) under load test makes it possible to determine the swelling capacity of the soil when it saturates under a previously established vertical pressure while it is kept laterally confined. The tests were performed in standard oedometers following approximately ASTM D 4546-03 Method A. The samples obtained by drilling and trimming had diameters between 3.6 and 5.0 cm and heights between 1.3 and 1.7 cm. Once in the oedometer, a vertical pressure of 0.5 MPa was applied to the samples. After stabilisation of the deformation, the samples were saturated 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 by linear strain transducers (LSCT) until stabilisation. During most of the tests, the electrical conductivity of the water in the oedometer was measured periodically with an electrical conductivity meter, which was submerged in the oedometer water every time the cell was refilled with deionised water to keep the water level in the cell approximately constant.

The ratio between the final length increase undergone by the sample in equilibrium with the load applied and its initial length gives the strain value of the material on saturation, the negative values indicating swelling strains. The final result is, therefore, the percentage of strain of a sample of given initial dry density and water content on saturation under a fixed load. The final strain is used to compute the final dry density of the specimens, which is assessed by measurement of the actual specimen dimensions upon dismantling. On completion of the tests, the water content of the specimen was determined by oven drying at 110 °C for 48 hours. The tests were performed at laboratory temperature.

Hydraulic conductivity

The hydraulic conductivity was measured according to a constant-head method developed at CIEMAT for expansive soils, in which the specimens are tested in stainless steel cells that guarantee the constant volume of the samples during the whole measuring process (Villar 2002). The samples obtained by drilling and trimming were confined in rigid cells of internal diameter 5.0 cm. The height of the samples –which was kept constant during the tests– was between 2.2 and 2.8 cm and filter papers and porous stones were placed on their top and bottom. The samples were saturated from both faces with deionised water injected at a pressure of 0.6 MPa over the necessary time period. This was checked by measuring the water intake until stabilisation. The saturation under pressure allowed the dissolution of the air contained in the pores, which volume was not expected to be large because of the high initial degree of saturation of most samples (>83% in all of them). Once the sample was saturated a hydraulic gradient was applied across it by increasing the pressure at the bottom of the cell, while the downstream pressure on top was maintained lower. The complete saturation of the sample and associated swelling guaranteed perfect contact with the walls of the cell, preventing the flow of water between these and the sample. The hydraulic gradients applied in these tests were of between 1600 and 11700 m/m. The water volume passing through the sample was measured online with pressure/volume controllers resolving 1 kPa and 1 mm^3. The tests run over a time period sufficient to determine that the volume of water passing through the specimen was linear and stable with time for a given hydraulic gradient. Hydraulic conductivity was then calculated by applying Darcy's law for flow in porous media.

Two or three different hydraulic gradients were applied to each sample and the tests were performed at room temperature. At the end of the tests, the samples were weighed, measured and oven-dried at 110°C for 48 h to check their final water content and dry density.

4. Results

The water content of the samples tested in the laboratory decreased from the granite towards the inner part of the barrier in all the sampling sections, whereas the dry density increased. In particular, in the sections around the heater analysed in this work the water content and dry density gradients were steep and linear, with values between 18 and 28% for the water content and 1.54 and 1.65 g/cm^3 for the dry density. In the three radii sampled in every section the changes were similar, which confirms the radial distribution pattern. The water content and dry density values obtained in the laboratory agreed very well with those obtained on site (Figure 3), what suggests that the packing and transport conditions were the appropriate to keep the *in situ* state of the blocks even several months after their retrieval.

The broad range of values found in the laboratory for both variables means that the initial conditions of the samples tested to determine their hydro-mechanical properties were very different and this had an impact on the results obtained.

Swelling under load tests

The swelling capacity was tested in 20 samples taken at different distances from the heater. The process of sample preparation, drilling and trimming to make the specimen fit into the cell ring, meant in most cases a decrease of the density of the specimen in the oedometer ring with respect to the dry density of the block from which they were obtained (the one shown in Figure 3).

The evolution of the vertical strain during the swelling tests in samples from section S47 is shown in Figure 4. The samples closer to the gallery wall tended to have higher initial water content and lower initial dry density, in agreement with the values of the blocks from which they were obtained (Figure 3). The swelling capacity is related to both, increasing with initial dry density and decreasing with initial water content (Eq. 1). For this reason the final strain of the samples closer to the heater tended to be higher. It also took longer for the deformation of these samples to stabilise. An exception to this general observation is sample BB47-4-2, which despite belonging to the external ring of the barrier swelled considerably. The most likely reason is its high initial dry density (1.59 g/cm^3), which does not correspond to its position in the barrier (Figure 3, right) and was probably caused by unintended compaction during specimen preparation for the swelling test. The final vertical strain recorded in all the tests is shown in Figure 5, where the trend for finding higher vertical strains in the samples closest to the heater is confirmed, since the samples closer to the gallery wall tended to have higher initial water content and lower initial dry density. Indeed the original dry density of the blocks from which the samples were trimmed was modified during trimming and sample preparation, and that is the reason why the dry densities indicated in Figure 5 do not show a straightforward increase towards the axis of the gallery. Nevertheless, the water content did decrease for each sampling section towards the axis of the gallery (because it was barely affected by sample preparation) and the combination of lower water content and (mostly) higher dry density, made the swelling capacity increase on average towards the internal part of the barrier.

Hydraulic conductivity

The hydraulic conductivity was measured in specimens drilled from blocks after they were saturated with deionised water for periods of time of between 13 and 86 days. Although many samples had a high initial degree of saturation, they took water because, once in the cell, their density decreased with respect to the original value, due to the filling of some irregularities that could have been created during trimming. In fact, there was a decrease in dry density of the samples tested in the permeability cells with respect to that of the blocks from which they were trimmed (the one shown in Figure 3), because of the sample preparation process described above.

After saturation, hydraulic gradients of between 1600 and 11700 m/m were applied to the samples and kept until the outflow rate was constant. Afterwards, the hydraulic gradient was changed and kept again until constant outflow rate. In some cases a third hydraulic gradient was applied. The whole measuring process took between 28 and 106 days. For the first samples tested, lower hydraulic gradients were applied (below 4000). Some of the values obtained have been plotted as a function of the hydraulic gradient applied in Figure 6. Although there is not a clear relationship between hydraulic conductivity and gradient, the lowest permeabilities were measured when hydraulic gradients below 5000 were used.

The results are plotted in Figure 7 as a function of the position in the barrier. The dry density of each sample, computed from the bentonite dry mass and the internal volume of the cell, is also indicated in the Figure. As a general rule the hydraulic conductivity of bentonite is mainly related to dry density (Eq. 2) and the latter in turn should be related to the position of the block in the barrier (Figure 3). Consistently with this, the overall trend in section S47 is for the hydraulic conductivity to decrease towards the heater, where the densities should be higher, whereas for section S53 there is not a clear dependence of hydraulic conductivity on the position of the sample inside the barrier. This is probably because the two samples drilled from the middle block of the barrier in section S53 had –as a consequence of trimming– higher dry density than those in the internal and external blocks, and consequently lower hydraulic conductivity.

The hydraulic conductivity values have been plotted in Figure 8 as a function of the dry density inside the permeability cell. The decrease of hydraulic conductivity with dry density is highlighted in the Figure. The fact that the decrease in dry density of the samples prepared for the permeability tests with respect to the original density of the blocks was not of the same magnitude for all the samples (as observed in section S53) might be the reason why the relation between hydraulic conductivity and distance to the axis was not straightforward.

The empirical relationship for the reference FEBEX bentonite relating hydraulic conductivity to dry density obtained for samples compacted to dry densities above 1.47 g/cm^3 and permeated with deionised water (Eq. 2) is also shown in Figure 8, along with its range of variation. When comparing these values to those expected for untreated FEBEX bentonite of the same dry density, it is found that the values for the FEBEX-DP samples were below the theoretical ones, in many cases even below the expected range of variation of this property for FEBEX bentonite.

5. Discussion

When analysing the hydro-mechanical properties of the bentonite retrieved from the FEBEX in situ test, the changes experienced by the bentonite during sampling on site and during preparation of specimens in the laboratory have to be taken into account. Although the samples were preserved carefully and their water content did not seem to have changed with respect to that the samples had during operation, the stresses in the barrier (which at some points were as high as 6 MPa during operation (Martínez et al. 2016) were released upon dismantling, and this probably implied a decrease in the bentonite dry density. Additionally, as it has been explained above, the preparation of specimens to fit the testing rings required drilling and trimming, which caused a decrease in the final dry density of the samples tested with respect to that of the bentonite blocks from which they were taken. The same observation was made earlier in the samples from the partial dismantling of the FEBEX in situ test (Villar & Lloret, 2007) and by other authors analysing samples retrieved from large-scale tests (Karnland et al. 2009, 2011). This decrease was not in all the cases of the same magnitude, since it depended on the sample conditions and on the operator.

Nevertheless, the aim of the determination of the hydraulic properties reported was to check if they had irreversibly changed during operation with respect to those of the reference bentonite. For this reason, the values obtained are compared in the following with those expected under the same testing conditions for the reference FEBEX bentonite.

In order to evaluate the modification of the swelling capacity of the bentonite as a result of the 18-year operation, the final vertical strain measured has been compared with the theoretical vertical strain of samples of the reference FEBEX bentonite of the same initial dry density and water content saturated under the same conditions, i.e. same vertical load and kind of water, since all these parameters affect the swelling capacity. Thus, the measured values and the theoretical results obtained with Eq. 1 for each sample are plotted in Figure 9. On average the vertical strains actually measured were lower than the theoretical ones (-10% *vs*. -12%). The Figure includes also the distance to the gallery axis of each sample. Most samples from the external, more saturated bentonite ring swelled less than expected, whereas the samples from the inner, drier ring tended to swell as expected or more. This could be related to their higher initial salinity, which would have triggered some additional osmotically-induced swelling. The high salinity of the pore water of the samples close to the heater was confirmed in the geochemical characterisation of samples retrieved from Grimsel performed by Fernández et al. (2017). In this work the very low salinity (lower than the one for the reference bentonite) of the samples from the external ring was also highlighted. The explanation given to the salinity changes across the barrier is that the water coming from the host rock dissolved soluble species and transported them towards the internal part of the barrier, where they accumulated. Hence, there were differences in the initial salinity of the samples tested in the oedometers.The electrical conductivity of the water in the oedometer cells was measured during the tests. In the tests performed with samples from the inner ring (the one closest to the heater), the electrical conductivity increased considerably during the tests, which would indicate that the soluble salts in the bentonite were being dissolved by the deionised water and transported outside the bentonite and into the water in the oedometer cell.

On the contrary, the electrical conductivity of the water in the oedometer cells in which samples from the external ring, the one closest to the granite, were being tested barely increased during the tests. As an example, Figure 10 shows the simultaneous evolution of swelling strain and electrical conductivity of the water in the oedometer cell for two samples of section S53, one of them obtained from the external bentonite ring (taken at 110 cm from the gallery axis) and the other one from the internal ring (taken at 53 cm from the gallery axis). Because of trimming, the initial dry density of these two specimens was similar, despite the fact that the blocks from which they were obtained had considerably different densities. The sample from the internal ring swelled more and for longer, with the electrical conductivity of the water in the oedometer cell increasing considerably, particularly after 10 days, when most swelling had already been developed. This could imply that the osmotically-induced part of swelling (indicated by the increase in electrical conductivity) was less important and was not remarkable until the crystalline swelling had been completed.

The hydraulic conductivity values measured in the samples retrieved were clearly lower than those expected for samples of untreated bentonite of the same dry density (Figure 8). In particular, the samples tested using hydraulic gradients below 4000 m/m had hydraulic conductivities clearly below those expected for the reference bentonite. On the contrary, the samples tested with higher hydraulic gradients had higher hydraulic conductivities. The determinations for the reference bentonite were performed applying hydraulic gradients on average of 15200 m/m, i.e. higher than those used for the testing of the FEBEX-DP samples. Back in 1976, Kharaka & Smalley pointed out the increase in hydraulic conductivity of compacted clays with the increase in hydraulic pressure gradients. Dixon et al. (1999) investigated the hydraulic conductivity of clays, among which bentonites, and found a transitional gradient below which low flow rates were recorded whereas above them the flows, and consequently permeabilities, were higher. A previous research analysing the effect of hydraulic gradient on permeability of the FEBEX bentonite found that –in some cases– the permeability tended to be slightly lower as the hydraulic gradient decreased (Villar & Gómez-Espina 2009). Hence, it is considered that the lowest permeability values measured in the present work can be a consequence of having used hydraulic gradients close to the critical ones in some samples. The critical gradient, as defined by Olsen (1962), would be the hydraulic gradient below which flow occurs but it is non-Darcian (i.e. the relationship between flow and hydraulic gradient is not linear), because of the strong clay-water interactions. The existence of threshold and critical hydraulic gradients for water flow in bentonite has been taken into account to model the hydration of the clay barrier by Sánchez et al. (2007).

Unlike what was observed in the swelling capacity tests, this decrease did not seem to be related to the position of the samples in the barrier. The osmotic effect is ruled out on the permeability samples, because during saturation the samples were confined and not able to swell differently during the permeability measurement as it happened in the swelling capacity tests.

There is also the possibility that, during the 18 years operation, the microstructure of the bentonite experienced changes that could affect the water flow. It is known that saturation involves a homogenisation of pore sizes towards the smaller sizes, and this makes the intrinsic permeability of the bentonite decrease with the degree of saturation (Villar & Lloret, 2001). Indeed this process took place during the saturation of the samples in the permeability cell, and it certainly occurred also during saturation of the samples used to determine the permeability of the reference bentonite. But it could have happened that over the 18 years of saturation in the barrier the average pore size had become lower and more homogeneous, and this would explain the lower hydraulic conductivity of the samples that were submitted to barrier conditions. In fact, the pore size distribution of the retrieved samples obtained by mercury intrusion porosimetry showed an increase in the percentage of pores smaller than 50 nm with respect to the reference bentonite. Also, the measurement of the basal spacing of these samples by X-ray diffraction indicated that the interlayer distance between smectite particles had increased from the initial value of 1.48 nm to values higher on average than 1.55 nm, as a consequence of the water content increase in the smectite interlayer (Villar et al. 2018). The modification of the FEBEX bentonite microstructure as a result of prolonged hydration, with overall increase of the microstructural void ratio and of the proportion of adsorbed, interlayer water, was also observed in the 12-year long thermo-hydraulic laboratory tests reported by Villar et al. (2016b). These works concluded that the maturation of the barrier would lead to a decrease in free water availability, since most water would eventually enter the interlayer porosity, a process that has been also described under different testing conditions by other authors (e.g. Karnland et al. 2006). The decrease in the average pore size along with the decrease in free/adsorbed water ratio would make the permeability of the material decrease over time, until equilibrium among pores and kinds of water is reached. Interestingly, the analyses performed in samples retrieved from the large-scale tests performed at the Äspö Hard Rock Laboratory summarised in the Introduction of this paper, showed also in some cases a slight decrease in the hydraulic conductivity of the MX-80 buffer with no spatial trend.

6. Conclusions

The FEBEX in situ test was a full-scale experiment performed under natural conditions to reproduce the conditions of the engineered barrier system in an underground repository of nuclear waste. The barrier around the heater simulating the waste container was composed of FEBEX bentonite blocks and had an average dry density of 1.6 g/cm^3. After 18 years of operation under repository conditions, natural hydration from the granitic host rock and heating from the heater, the heater was switched off and the experiment was dismantled.

This paper has presented the results of part of the hydro-mechanical characterisation of bentonite samples taken during dismantling. The main objective of this work was assessing if these properties had irreversibly changed as a consequence of the conditions during operation. To this aim, the values for these properties obtained in the samples retrieved from Grimsel were compared with those expected for the reference, untreated bentonite tested in the same way.

The bentonite water content decreased from the external part of the barrier (close to the granite) towards the internal part (i.e. the axis of the gallery) and the dry density followed the inverse pattern. Consequently, the samples retrieved had very different water contents and dry densities, both of which affect greatly the hydro-mechanical properties.

The preparation of specimens for the swelling capacity and hydraulic conductivity tests caused some reduction in their dry density with respect to the original dry density of the bentonite blocks. Since the evaluation of the change of these properties during operation was done by comparing with untreated samples of the same dry density and water content, this density decrease does not impair the conclusions obtained, but it has to be taken into account that the permeability and swelling strain values measured do not correspond to those the bentonite had in the barrier, where its dry density was higher and some boundary conditions (water availability and salinity, stress state) were different.

The swelling capacity of the samples taken from the external ring of the barrier, i.e. those more saturated, was lower than expected, whereas the samples from the inner ring of the barrier, those closest to the heater, swelled in the range expected of even more. The reason for these observations could be the different content in soluble salts of the samples depending on their position in the barrier: the pore water of the samples closest to the granite had been leached by the water coming from the host rock and moving inwards. On the contrary, the salinity of the samples closest to the heater was much higher than the initial one because the soluble species concentrated in this area. This salinity would have triggered some additional osmotically-induced swelling, as the salts in the bentonite were dissolved and moved towards the water in the oedometer cell, which was periodically refilled with deionised water. This process did not take place in the reference bentonite or in the more saturated bentonite from the external ring, because in both cases the salinity was lower. It has to be highlighted that this is a process that took place because the samples were let swell under a low vertical stress in a relatively large volume of water, but the same conditions are not to be encountered in the barrier during operation.

The hydraulic conductivity of the samples retrieved was in the order of 10^{-14} m/s and mainly related to the dry density of the samples, but overall it was lower than expected for the untreated FEBEX bentonite. The fact that the hydraulic gradients applied in this work were lower than those used to determine the hydraulic conductivity of the reference bentonite, and in some cases close to the critical hydraulic gradient (i.e. the gradient below which flow is non-Darcian as a result of the strong clay-water interactions), could have contributed to this difference. Also, a decrease of the average pore size of the bentonite during the 18-year operation and a decrease of the free/adsorbed water ratio, more drastic than the one to be expected during the relatively short period of saturation that precedes the permeability determination, could play a part in the decrease of hydraulic conductivity observed.

Despite the changes observed in the properties analysed with respect to those of the reference bentonite, the performance of the barrier was very good, the permeability continued to be low and the swelling capacity was very high and enough to fill all the construction gaps of the barrier. The dry density and water content gradients observed in the barrier have a repercussion on its hydro-mechanical properties, since most of them depend greatly on the density and water content of the bentonite. This could lead to an inhomogeneous distribution of swelling pressure and permeability in the barrier that should be taken into account when modelling the barrier behaviour and evolution.

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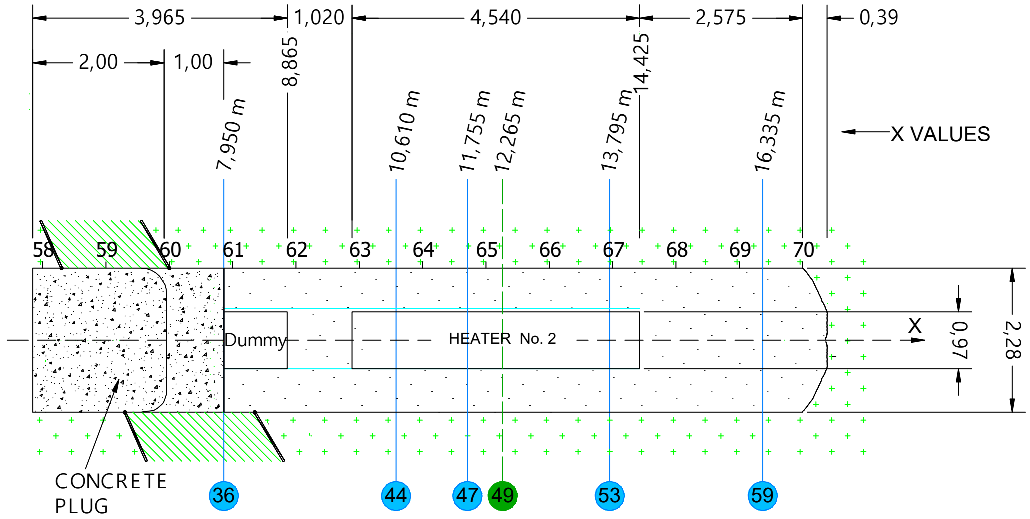

Figure 1: General layout of the in situ test during phase II and location of the sampling sections from which the samples sent to CIEMAT were taken (S49 was analysed on site)

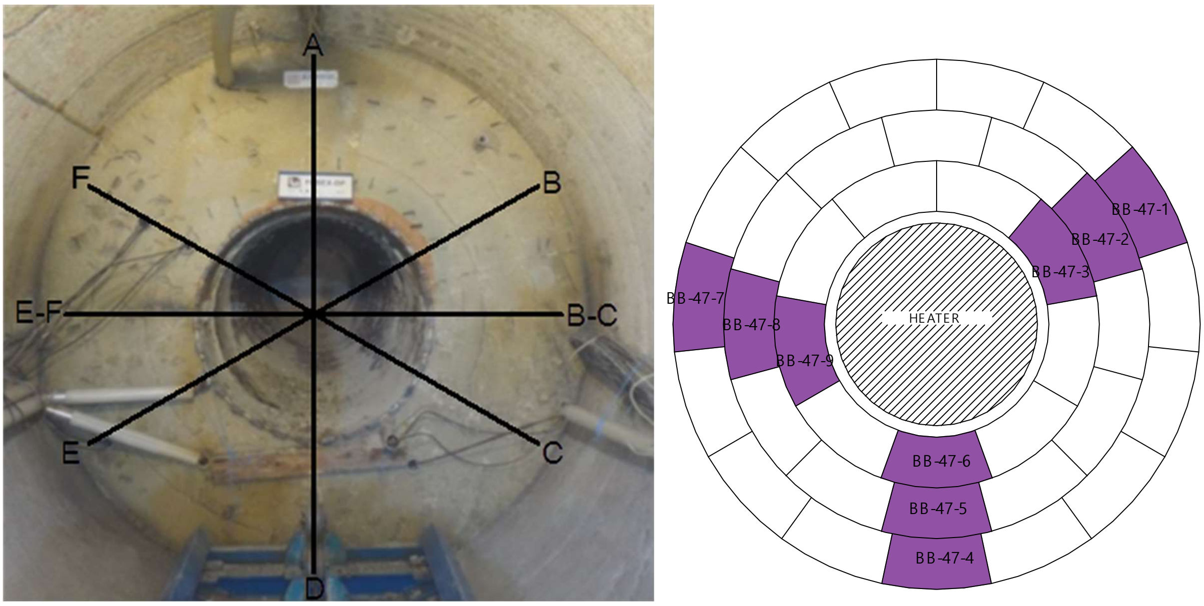

Figure 2: Left: appearance of the bentonite barrier after extraction of heater 2 in 2015, with indication of sampling radii; Right: location and reference of block samples from sampling section S47 sent to CIEMAT (coloured, BB stands for Bentonite Block, 47 is the sampling section as in Figure 1)

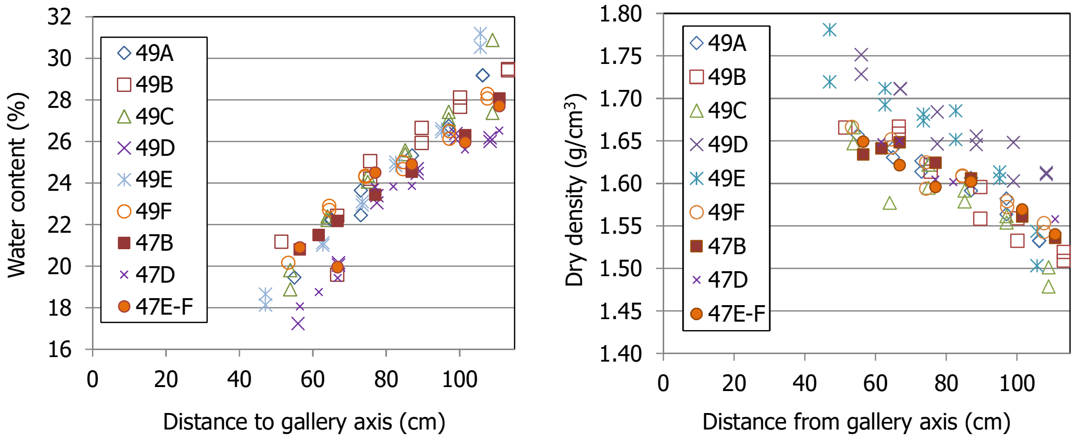

Figure 3: Comparison of water content and dry density measured in close-by sections S47 (CIEMAT's lab) and S49 (onsite, Villar et al. 2016a). The location of the sections is indicated in Figure 1.The letters in the legend indicate the sampling radius according to Figure 2

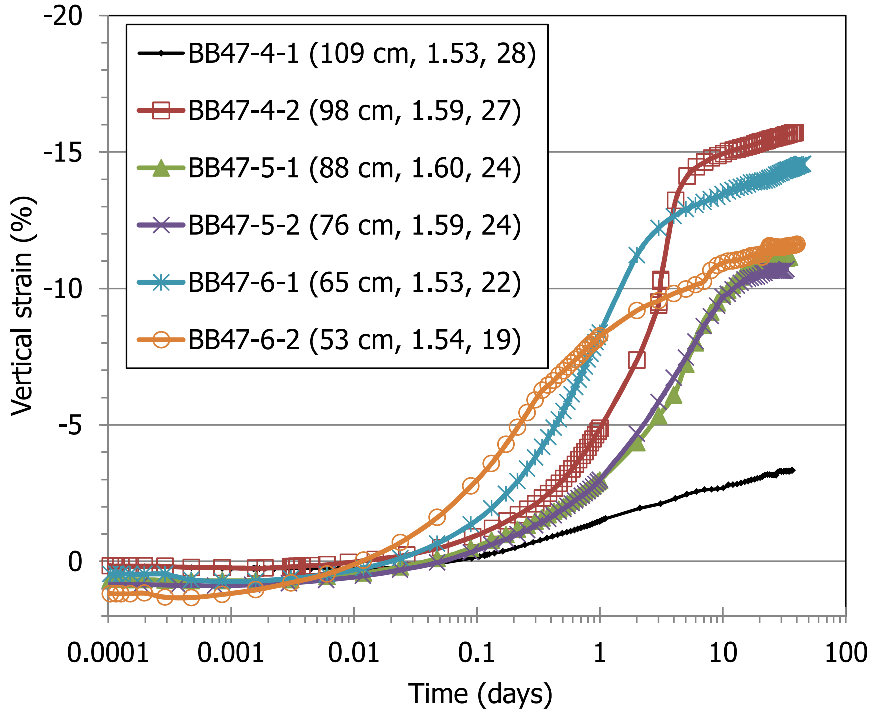

Figure 4: Evolution of vertical strain during the swelling under 0.5 MPa stress for samples of section S47. The reference of the samples corresponds to the block reference indicated in Figure 2. The distance to the axis, the dry density (g/cm^3) and water content (%) of each sample are indicated in the legend

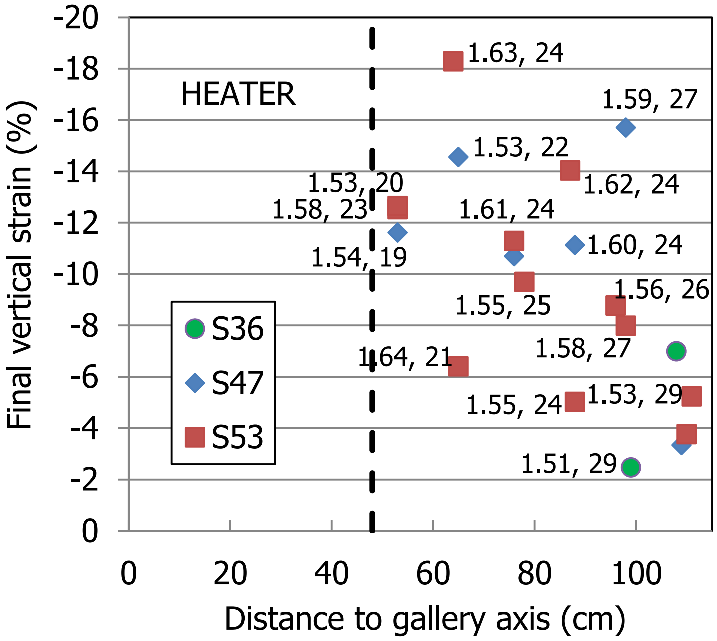

Figure 5: Final vertical strain for samples of different sections saturated under vertical stress 0.5 MPa (the initial dry density and water content of some of the specimens is indicated in g/cm^3 and %)

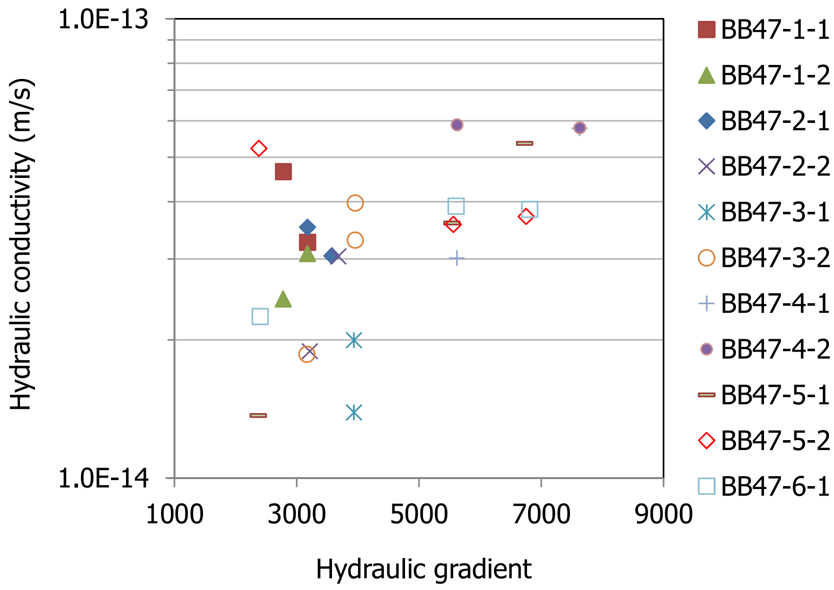

Figure 6: Hydraulic conductivity of samples from section S47 as a function of the hydraulic gradient applied (the references of the samples correspond to the names of the blocks indicated in Figure 2)

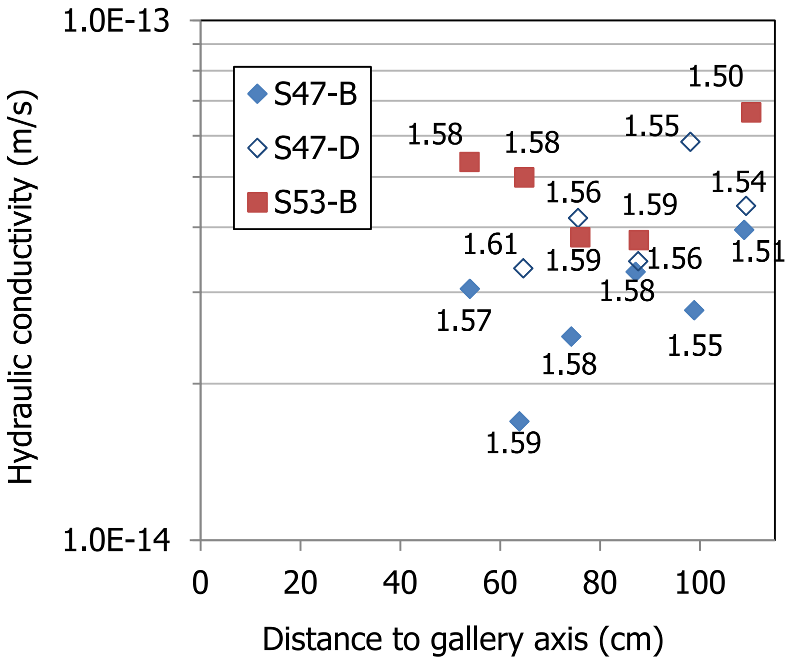

Figure 7: Hydraulic conductivity of samples from section S47 and S53 taken along different radii (the dry density of the specimens is indicated in g/cm^3)

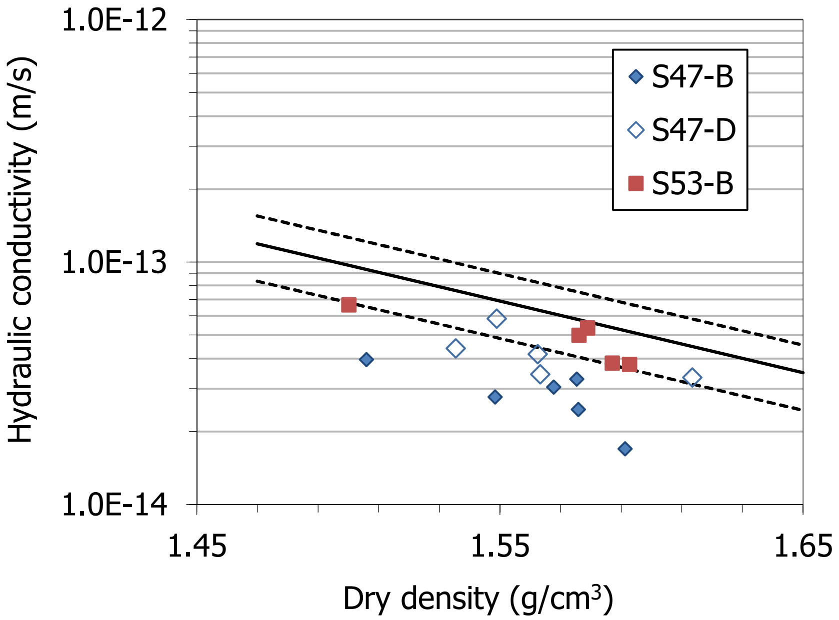

Figure 8: Hydraulic conductivity of samples from different sections and radii (indicated in the legend) and empirical correlation for untreated FEBEX bentonite obtained with Eq. 2 (solid line, the dashed lines indicate the expected range of variation, 30%)

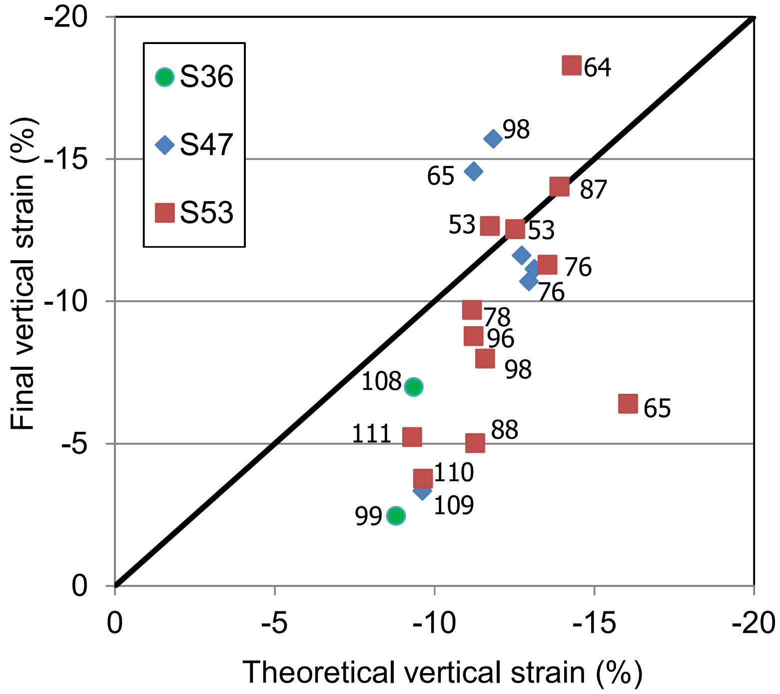

Figure 9: Comparison between the vertical strain measured at the end of the swelling under load tests and the strain corresponding to equivalent samples of the reference bentonite tested under the same conditions obtained with Eq. 1. The distance to the gallery axis of each sample is indicated in cm

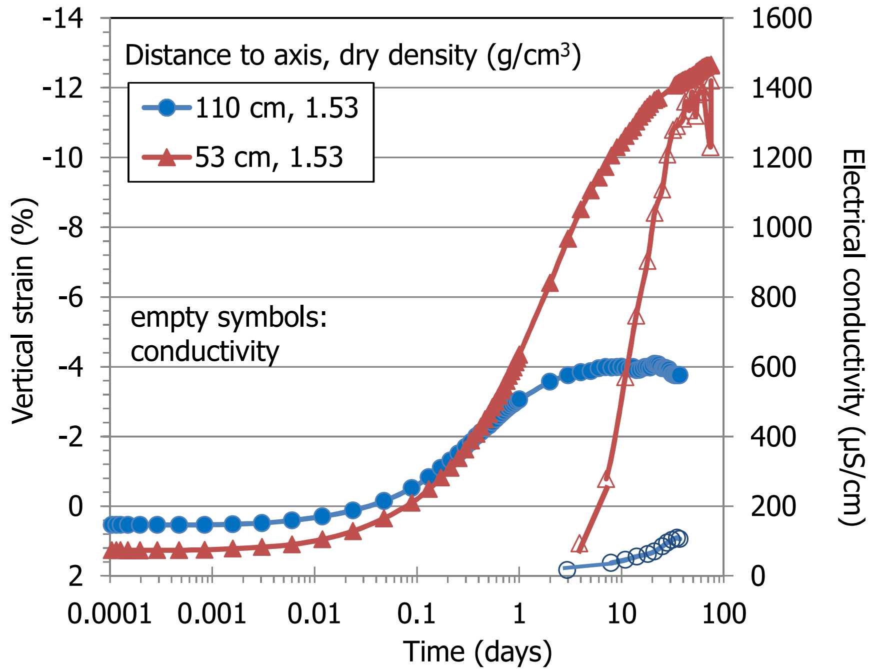

Figure 10: Evolution of vertical strain and electrical conductivity of the water in the oedometer cell during the swelling under 0.5 MPa vertical stress for two samples of section S53

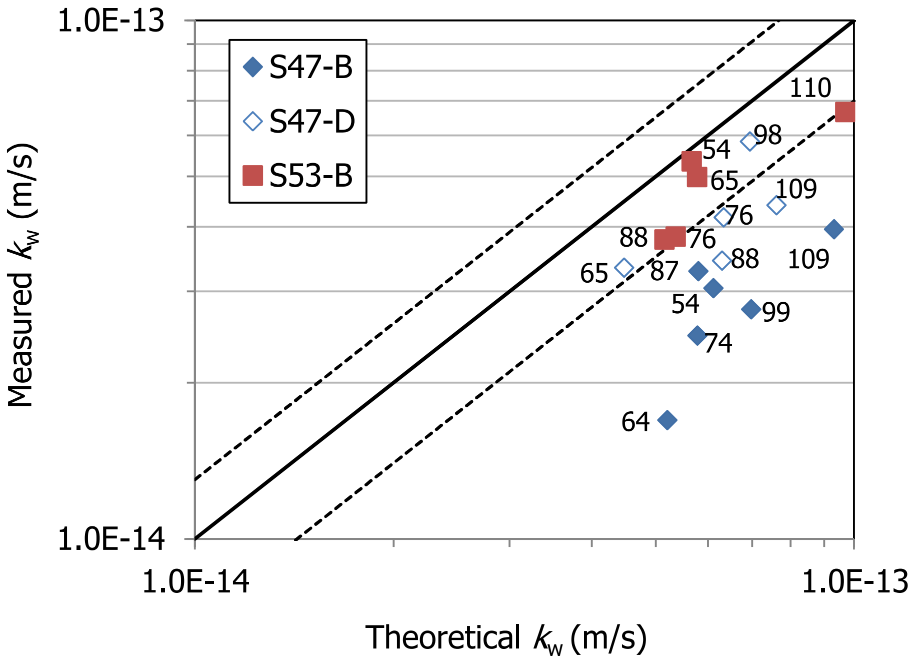

Figure 11: Comparison between the hydraulic conductivity measured and that corresponding to samples of the reference bentonite of the same dry density obtained with Eq. 2. The distance to the gallery axis of each sample is indicated in cm