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Thermo-Hydro-Mechanical Characteristics and Processes in the Clay Barrier of a High Level Radioactive Waste Repository. State of the Art Report

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CARACTERÍSTICAS Y PROCESOS TERMO-HIDRO-MECÁNICOS DE LA BARRERA DE ARCILLA DE UN ALMACENAMIENTO DE RESIDUOS RADIACTIVOS DE ALTA ACTIVIDAD

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75 pp. 72 fig. 102 refs.

Resumen:

Este documento es un resumen de la información existente en la actualidad sobre propiedades termo-hidro-mecánicas de la barrera de bentonita de un almacenamiento de residuos radiactivos de alta actividad y de los procesos que tienen lugar en ella durante las diferentes fases de operación del almacenamiento. Se analizan especialmente las propiedades térmicas, los procesos de cambio de volumen (hinchamiento y consolidación), la permeabilidad y la capacidad de retención. Se revisa el conocimiento experimental existente sobre la modificación de estas propiedades por efecto de la temperatura, salinidad del agua, humedad y densidad de la bentonita, y su evolución previsible como consecuencia de los procesos que se esperan en el almacenamiento. La información recopilada se refiere fundamentalmente a las bentonitas FEBEX (España), MX-80 (Estados Unidos) y FoCa (Francia), consideradas materiales de barrera de referencia en varios conceptos europeos de almacenamiento.

THERMO-HYDRO-MECHANICAL CHARACTERISTICS AND PROCESSES IN THE CLAY BARRIER OF A HIGH LEVEL RADIOACTIVE WASTE REPOSITORY

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Abstract:

This document is a summary of the available information on the thermo-hydro-mechanical properties of the bentonite barrier of a high-level radioactive waste repository and of the processes taking place in it during the successive repository operation phases. Mainly the thermal properties, the volume change processes (swelling and consolidation), the permeability and the water retention capacity are analysed. A review is made of the existing experimental knowledge on the modification of these properties by the effect of temperature, water salinity, humidity and density of the bentonite, and their foreseen evolution as a consequence of the processes expected in the repository. The compiled evolution refers mostly to the FEBEX (Spain), the MX-80 (USA) and the FoCa (France) bentonites, considered as reference barrier materials in several European disposal concepts.

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ACRONYMS USED

AECL	Atomic Energy of Canada Limited
AITEMIN	Asociación para la Investigación y Desarrollo Industrial de los Recursos Naturales (*Association for the Research and Industrial Development of Natural Resources*) (Spain)
ANDRA	Agence Nationale pour la Géstion des Déchets Radiactives (*National Radioactive Waste Management Agency*) (France)
BCE	Buffer Container Experiment (AECL, Canada)
BMT	Buffer Mass Test (Stripa, Sweden)
Boom Clay	Clay formation used as reference host rock in the Belgian disposal concept, constituted by kaolinite/illite, quartz and inter-stratified
CEA	Commissariat à l'Énergie Atomique (*Commisariat for Atomic Energy*) (France)
CEC	Cationic exchange capacity
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (*Centre for Energy, Environment and Technology Research*) (Spain)
DDL	Diffuse double layer
DOE	Department of Energy (United States of America)
ENRESA	Empresa Nacional de Residuos Radiactivos (*National Radioactive Waste Management Agency*) (Spain)
FEBEX	Full Scale Engineered Barrier Experiment in Crystalline Host Rock
GTS	Grimsel Test Site (NAGRA, Switzerland)
HLW	High level radioactive wastes
ITT	Isothermal Test (AECL, Canada)
JNC	Japan Nuclear Cycle Development Institute
LOT	Long term test of buffer material (Äspö, Sweden)
NF–PRO	
ONDRAF/NIRAS	Organisme National pour les Déchets Radioactifs et les matières fissiles enrichies (*National radioactive waste and enriched fissile materials organisation*) (Belgium)
RESEAL	A large scale *in situ* demonstration test for repository sealing in an argillaceous host rock (Mol, Belgium)
SKB	Svensk Kärnbränslehantering AB (*Swedish Nuclear Fuel and Waste Management Co*)
THM	Thermo-hydro-mechanical
UPC	Universidad Politécnica de Cataluña (Spain)

THERMO-HYDRO-MECHANICAL CHARACTERISTICS AND PROCESSES IN THE CLAY BARRIER OF A HIGH LEVEL RADIOACTIVE WASTE REPOSITORY.

STATE OF THE ART REPORT.

1 INTRODUCTION

This report has been drawn in the context of the NF-PRO Project, carried out under the European Commission Sixth Framework Programme (2002-2006) within the Euratom Research and Training Programme on Nuclear Energy. It is a summary of the current knowledge on the thermo-hydro-mechanical properties of the bentonite barrier of a high level waste (HLW) repository and of the processes that take place in it. Its main purpose is to highlight which are the critical aspects to be taken into account and to provide a basis for the planning of the subsequent research work.

1.1 The geological disposal of radioactive wastes

Radioactive wastes are considered to be any substance that contains or is contaminated with radionuclides in concentrations higher than those established by the competent authorities and for which no subsequent use is foreseen. Such wastes are produced during the generation of electricity using nuclear means, in the decommissioning of nuclear and radioactive installations and in the use of radioisotopes in industry, medicine, agriculture, research, etc. From a definitive disposal point of view, radioactive wastes are generally classified into low and intermediate level wastes and high level wastes. The latter have high specific short-lived emitter activities, contain appreciable concentrations of long-lived alpha-emitting radionuclides and are major heat producers (Echagüe *et al.* 1989).

The solution as regards protecting people and the environment against the radiations emitted by the radionuclides contained in high level wastes consists in isolating them in such a way that throughout the period in which they remain active they cannot be released to the biosphere along any of the possible paths. The generally accepted option for the final disposal of HLW consists in their disposal in stable deep geological formations (500-1000 m) (Goguel *et al.* 1987), as was suggested by the United States National Academy of Science in the 1950's. In order to guarantee isolation, a series of natural and artificial barriers is put into place (Figure 1):

1. The chemical barrier, which is constituted by immobilising the waste in a solid, stable, long-lasting and chemically inert matrix.

2. The physical barrier, which is the high corrosion resistance canister in which the immobilised wastes are confined.

3. The engineered barrier, which is the installation in which the wastes are placed, made up of a series of chambers, shafts and drifts in which the canisters are introduced. The diameter of these excavations is always larger than that of the canister, the remaining

space being surrounded by appropriate materials known as sealing materials, the chambers and access shafts being plugged with the so-called backfill material.

4. The geological barrier, constituted by the geological medium housing the installation (salts, granites, clays, basalts or volcanic tuffs) and the surrounding geological formations.

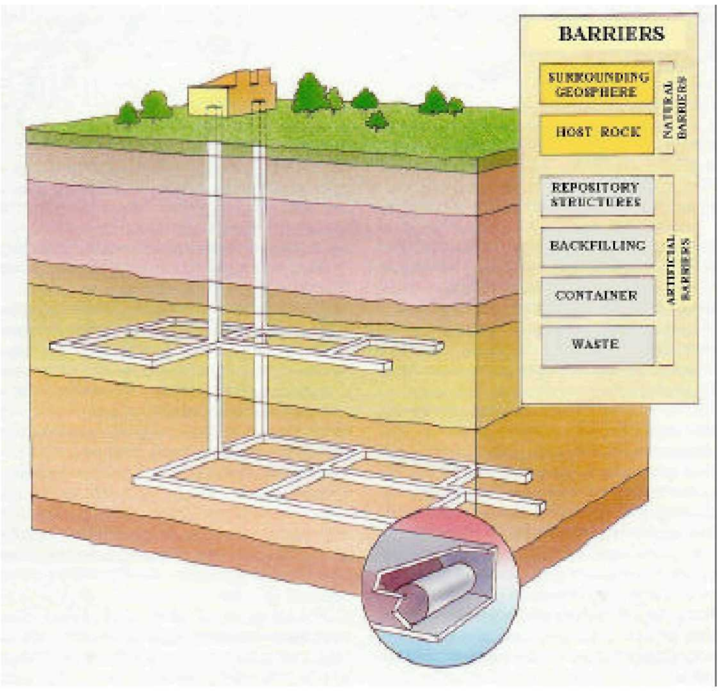

Figure 1: Schematic representation of the concept of multiple barriers in a geological disposal facility

The system of barriers aims to prevent the possible escape paths for the radionuclides to the environment, the most important of which is the circulation of groundwaters. In this context, the basic functions of the sealing material –which is the subject of this report– are to prevent or limit the entry of water to the wastes and to contribute to radionuclide retention. Other additional functions are to contribute to heat dissipation and to provide mechanical protection for the waste canisters.

1.2 Bentonites as backfill and sealing material

In view of the functions of the sealing material described in the previous section, Pusch (1979) proposed the use of high density compacted sodium bentonite in the form of blocks as a sealing material, since it provides the following characteristics:

- Very low permeability, reducing the percolation of groundwaters, since hydrogeological transport is the main radionuclide transfer mechanism.
- High exchange capacity, and therefore a high capacity for ion adsorption in the event of radionuclide release.

- Sufficient thermal conductivity, to prevent the generation of excessive thermal gradients.
- Mechanical resistance to withstand the weight of the canister.
- Mechanical properties guaranteeing the homogeneous nature of the barrier, this including plastic behaviour to prevent the formation of fissures as a result of differential displacements or the location of stresses, and swelling potential favouring the self-sealing of existing voids.

Other requirements of the material used as barrier that have been underlined in subsequent research (Yong *et al.* 1986) are as follows:

- Good compressibility, ensuring ease in processing, handling and transport to the disposal facility.
- Low shrinkage in response to the drying that will probably occur in the area surrounding the canister, in order to prevent the formation of a network of fissures.
- Non-excessive swelling pressure, to avoid damage to the system.
- Suitable deformability, ensuring that the pressures generated by the rock massif and by the hydration of the expansive component of the barrier are absorbed and reduced by deformation of the barrier itself.
- Physical and chemical stability, ensuring the longevity of the system in relation to the conditions of the disposal facility, *i.e.* high temperatures, chemical gradients and the presence of vapour (Pusch & Gray 1989, Güven 1990).

Since the end of the 1980's, the agencies in charge of the waste management in different countries have proposed other sealing materials, some based on the use of cement but most considering the use of bentonites, either non-sodic smectites (ANDRA, France; ENRESA, Spain) or mixtures of expansive clay and aggregates in different proportions. As regards the aggregates, tests have been performed using crushed granite (AECL, Canada), crushed basalt (DOE, United States), zeolytes and quartz (JNC, Japan), quartz and graphite (ANDRA, France; ONDRAF/NIRAS, Belgium). The main objective of adding inert aggregates is to increase the thermal conductivity of the barrier, improve the mechanical resistance of the compacted blocks and reduce the cost of the material.

Although most of the studies have been carried out considering that the emplacement of the barrier will be made in the form of pre-compacted blocks, other systems have been proposed. Among them, the use of high density bentonite pellets combined with powdered bentonite that, once saturated and with the density of the overall assembly homogenised, give rise to a barrier density equivalent to that achieved with compacted blocks (Salo & Kukkola 1989, Volckaert *et al.* 1996). The advantage of this system is that it is easy to manufacture and install, especially in emplacements in the form of vertical shafts.

1.3 Description of smectites

Bentonites are fine grained rocks made up of crystalline clay minerals formed by the devitrification and accompanying chemical alteration of vitreous igneous materials, normally volcanic tuffs or ashes (Ross & Hendricks 1945). The term bentonite has extended nowadays to any clay rock in which the predominant minerals are smectites. They are constituted by different minerals in variable proportions (quartz, feldspar, micas and amphiboles, among others), but most of the minerals present in bentonites belong to the smectite group, in which montmorillonite is the most common species.

Bentonite, like all clays, is a material that becomes plastic when mixed with water and, since it is made up fundamentally of smectites, has extraordinary expansive properties.

Smectites are clay minerals belonging to the philosilicates group and made up of structural units comprising two layers of polyhedra of tetrahedral coordination (T) between which there is a central octahedral layer (O), as a result of which they are known as 2:1 silicates. Inside each tetrahedron there is an atom of silicon coordinated with 4 oxygens or hydroxiles located at the tips. The octahedral layers contain atoms of aluminium, iron (II or III) or magnesium in their interior, in coordination with 6 oxygens or hydroxiles distributed on two surfaces parallel one to the other (Figure 2). All the vertices of the tetrahedra point in the same direction, towards the centre of the unit, while the bases of each of the layers of tetrahedra constitute the external planes of the unit. The tetrahedral and octahedral layers are combined in such a way that the vertices of the tetrahedra of each layer of silica and one of the surfaces of the octahedral layer are located on the same plane. The atoms common to the tetrahedral and octahedral layer are oxygens instead of hydroxiles.

When the cation of the octahedral layer is trivalent, as for example in the case of aluminium, one of every three cation positions is unoccupied, this causing the octahedral layer to have the structure of gibbsite, $Al(OH)_3$, and the smectite to be known as dioctahedral. This group includes montmorillonite, beidellite and nontronite. In trioctahedral smectites, the cation in the octahedral layer is divalent, as a result of which all the cation positions are occupied, this giving rise to an octahedral layer with the geometry of brucite, $Mg(OH)_2$. This is the case for the saponites and hectorites (Hurlbut & Klein 1982).

The T-O-T units are known as laminae and are continuous in directions *a* and *b*, in other words their extent is theoretically indefinite, while in the *c* direction they are piled one on top of the other. When piling of the T-O-T units occurs, the oxygen planes of each unit are left adjacent to the oxygen planes of the neighbouring units. Depending on the isomorphic replacements that take place in the structure, the laminae will have a negative charge of a greater or lesser magnitude. For this reason, the positions between adjacent planes of oxygen are usually occupied by exchange cations. This means that water may enter between these units for solvation of the cations and other polar molecules, causing the crystalline network to expand in the direction of the *c*-axis. For this reason, the dimension of smectite on *c*-axis is not fixed, but varies depending on the size of the molecule located between the units and on its hydration state. When there are no molecules between the laminae, the dimension of the *c*-axis will be between 9 and 10 Å (0.0009-0.001 μm), this possibly increasing to 16 Å when there are polar molecules or hydrated cations (Grim 1968). The number of cations required to reach electrical equilibrium is the cation exchange capacity (CEC) of the clay, which may be measured experimentally.

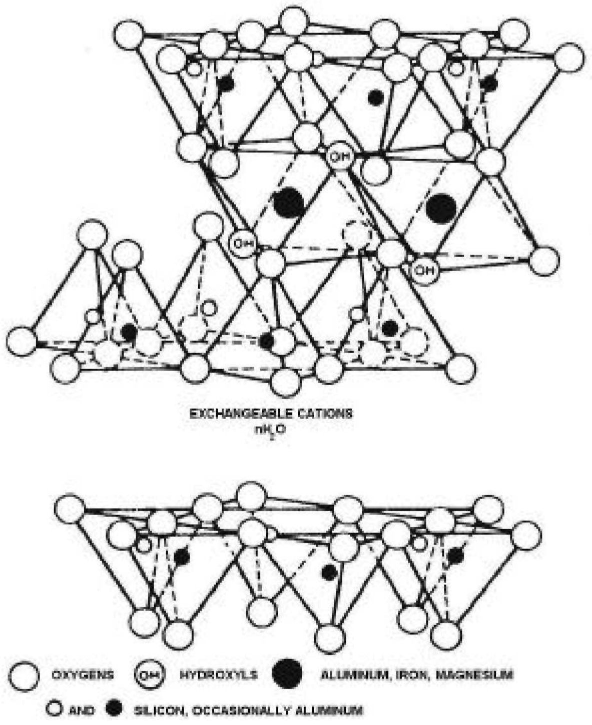

Figure 2: Schematic representation of the structure of montmorillonite (Grim 1968)

The piling of clay laminae forms primary particles (or stacks) with a thickness of between 0.0015 and 0.0150 µm, this possibly varying in the case of smectites depending on the hydration state and reaching up to 0.0600 µm (Ben Rhaiem *et al.* 1986). The piled laminae are separated by layers of water. The number of clay laminae in each particle depends very much on the type of exchangeable cations, being higher in the case of bivalent cations. When these layers are organised, the primary particles are known as quasi-crystals. In turn, a group of quasi-crystals gives rise to aggregates.

As has been seen, the surface of the primary particles in 2:1 silicates is an interface with a negative electrical charge. Given that the water in the soil contains dissolved solutes, among which there are different cations, the interaction between the charged surface of the clay particle and the cations in the pore water gives rise to an electric double layer (EDL), that is to say, to ordering of the negative and positive charges around the electrical interface. However, given that the ions in a solution are endowed with mobility, the cations and anions will be mixed following the first layer of positive charges (that constitutes the EDL), with the former predominating close to the surface of the particle and with the charges equalling out as the distance from the particle increases. The area in which the electrical potential varies depending on the distance from the particle is known as the diffuse double layer (DDL), or more accurately the diffuse ion layer (DL). Figure 3 is a schematic representation of the development of the EDL and the DL. When two particles are located close to each other, their DL's interact, giving rise to a repulsion between them whose magnitude is conditioned in part by the chemistry of the pore water. The thickness of the DL depends on a number of factors, among which are the concentration of salts, the valence of the exchange cations and the temperature, which cause the thickness to decrease. On the other hand, the size of the hydrated ion and the dielectric constant and pH of the solution increase the DL thickness. These factors in turn influence the swelling capacity of the clay, since it increases with the thickness of the DL.

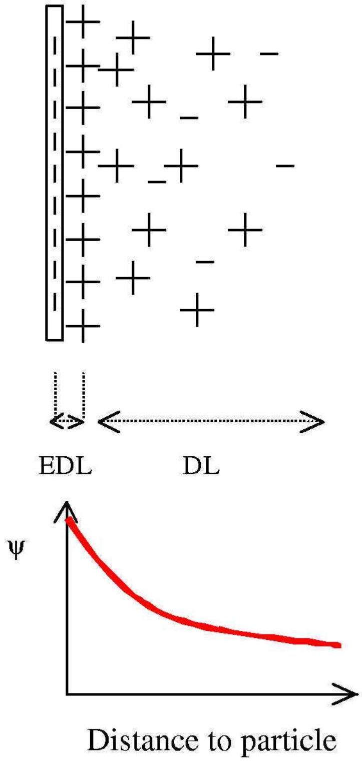

Figure 3: Schematic representation of a negatively charged clay particle interacting with the ions in the pore water. EDL: electric double layer, DL: diffuse ion layer, ψ: electrical potential (Yong *et al.* 1992)

The organisation of the laminae, particles and aggregates in a clay material gives rise to different types of porosity. Insight into these types is necessary to understand the mechanisms and processes that take place at different structural levels inside the material, since each porosity is associated with a type of water having different properties (Stepkowska 1990, Wersin *et al.* 2004). A distinction may be made between the following types of porosity, which are shown schematically in Figure 4 and Figure 5:

- Interlaminar or interfoliar porosity, to which only polar molecules have access and whose spacing ranges from 2 to 10 Å (although this may be larger in the case of hydrated sodium smectites). This constitutes the cation solvation water. The water accessing this porosity is influenced by the electrical field and is strongly bound, as a result of which its properties are very different from those of free water, which has less mobility. Some authors claim that this water may move freely across the surface of the lamina but not perpendicularly to it (Stepkowska 1990).

- Intra-aggregate porosity, inside the primary particles and between adjacent laminae piles. This is also known as lenticular porosity or microporosity, and includes pores having diameters of less than 0.0020 µm, although in high saturation states they may reach up to 0.0035 µm. The diffuse layers develop in these pores. The water that forms part of the diffuse layers is also adsorbed and cannot be separated from the clay particle as a result of hydrodynamic action until the distance to the particle is sufficiently large.

- Inter-aggregate porosity (greater than 0.002 µm), which in turn may be classified as macroporosity or mesoporosity, depending on whether the diameter is above or below 0.05 µm. In these pores, the water is retained by capillary or gravitational forces, and is known as external water, in constrast to the water contained in the rest of the porosity, which is known as internal. The proportion of external water in smectites decreases with increasing material density and decreasing water content, and may be lower than 20 percent in the case of compacted calcium smectites with a dry density of 1.60 g/cm^3 (Pusch 1994). However, in the case of a calcium montmorillonite paste without confinement subjected to very low values of suction (0.032 bar), Ben Rhaiem *et al.* (1986) give a percentage of external water of 85 percent.

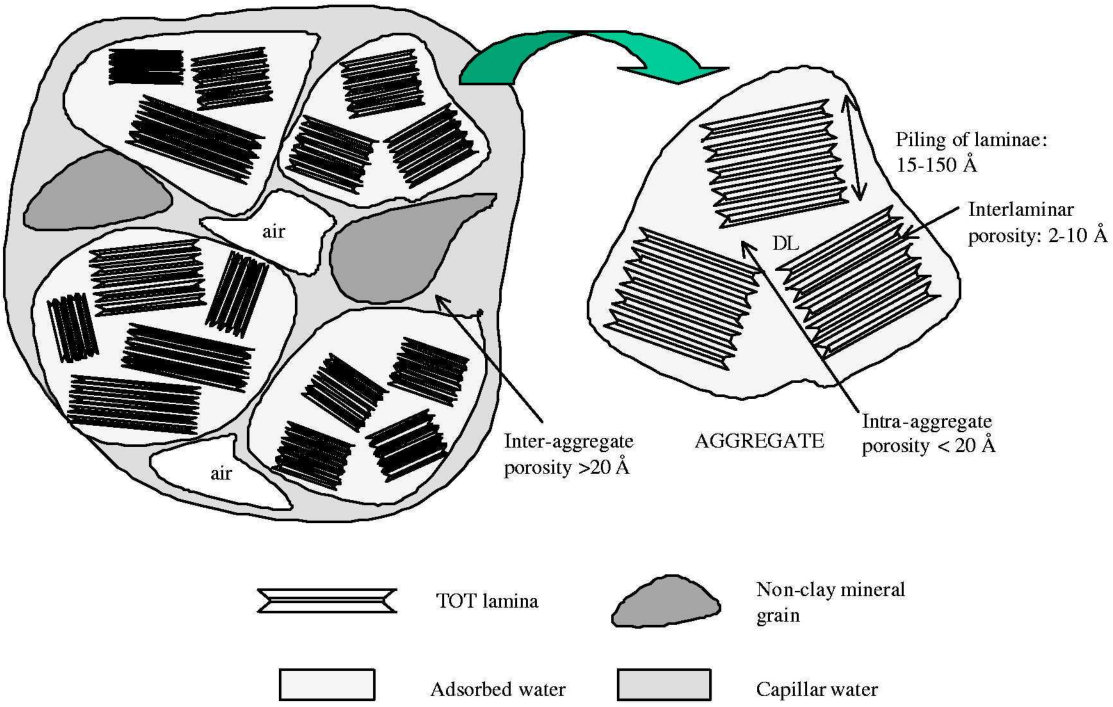

Figure 4: Schematic diagram of the organisation of a clay material: types of porosity and water (Villar 2002)

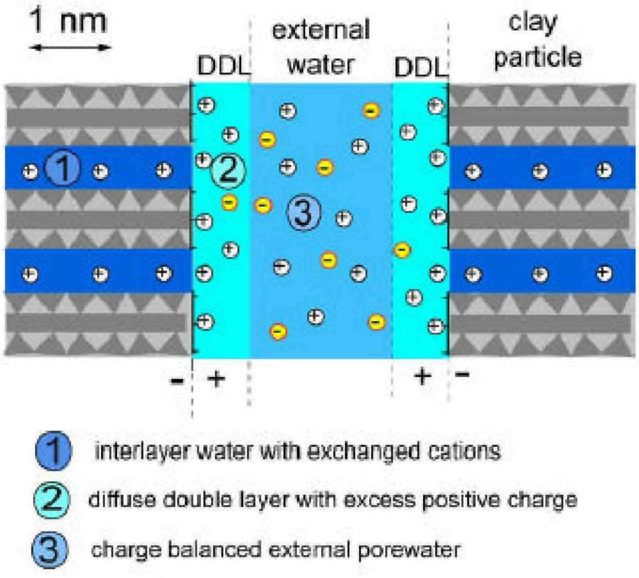

Figure 5: Water types in bentonite (Wersin *et al.* 2004)

For the MX-80 bentonite (a Na bentonite described below), Sauzeat *et al.* (2001) propose the multi-scale hierarchy of texture shown in Figure 6.

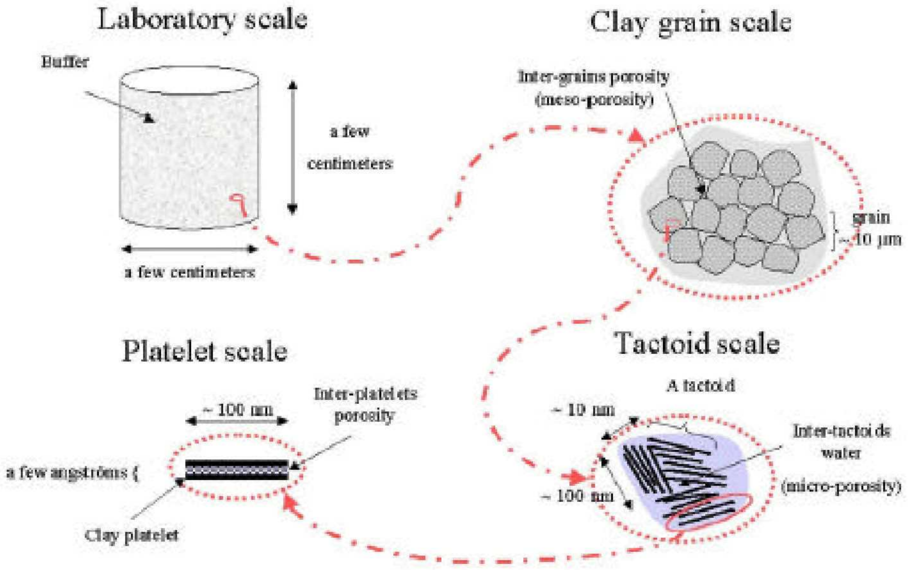

Figure 6: Multi-scale structure of a Na bentonite (Sauzeat *et al.* 2001)

The contribution of each kind of porosity (and water) to the total porosity (and water) depends on several factors, such as clay density, salinity of the pore water and type of exchangeable cation in the interlayer. Thus, since the particles in Na smectites are composed by less laminae than in Ca smectites, the interlaminar porosity will be smaller in the former, while the number of stack contacts will be higher and consequently the locations available for the development of diffuse layers (Pusch *et al.* 1990, Figure 7).

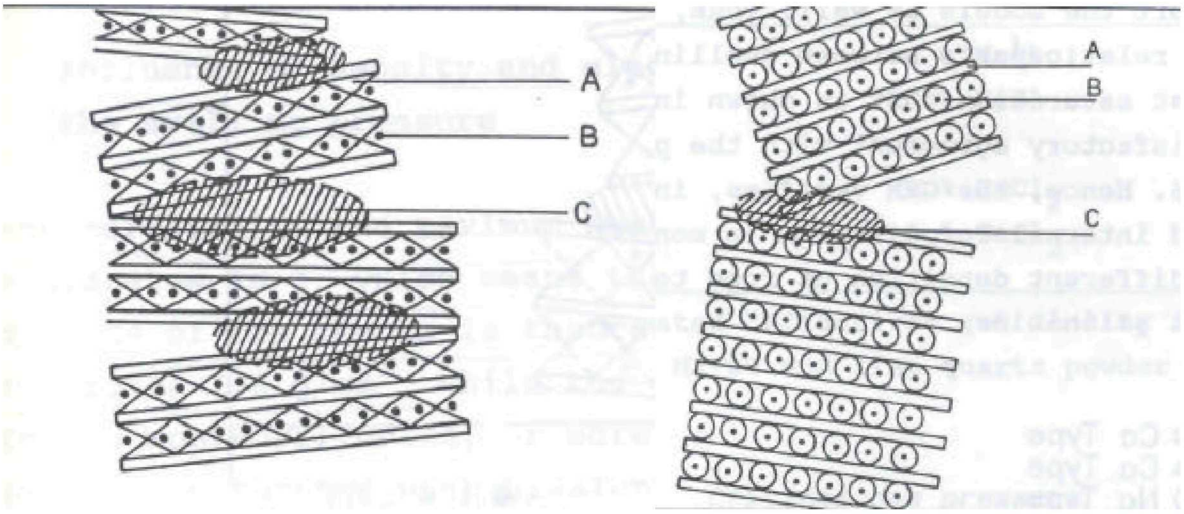

Figure 7: Schematic picture of the stack assemblage in Na (left) and Ca (right) montmorillonite (A: lamina, B: interlaminar space, C: stack contact with interacting DDL) (Pusch *et al.* 1990)

On the other hand, the retention of water in the soil occurs depending on different mechanisms, with one or another predominating depending on the water content of the soil (*i.e.* on the activity of the water): for low water contents the prevailing mechanism is adsorption, which depends on the specific surface and takes place in the micropores, while for higher water contents it is capillary condensation, which depends on the shape and arrangement of the particles and takes place in the mesopores and macropores (Everett & Haynes 1973). Adsorption creates pillars around the exchangeable cations and a film of water at the external surfaces of the particles or aggregates, both of these processes leading to an increase in the spacing between the laminae and the distance between particles, in other words, to swelling. The adsorption forces that induce swelling are balanced by the capillary condensation forces (Prost *et al.* 1998).

The water contained in the different porosities is known overall as hydration water. The dehydration of a mineral is a process that involves reactions having thermodynamic (temperature) and kinetic (time) conditioning factors (Cuadros *et al.* 1994). Thus, for example, hydration water is lost with heating to 110 °C if this is prolonged over 24 hours. In thermogravimetric studies, however, in which heating of the clay takes place at high speed (of the order of 10 °C/min), it is generally accepted that the hydration water exits the clay when it is heated to temperatures below approximately 300 °C, with the greater part being lost below 200 °C. Furthermore, thermogravimetric studies performed on smectites show a more or less continuous loss of mass at higher temperatures, with the temperatures of the hydration water loss overlapping those at which the loss of structural water occurs, *i.e.* the temperatures at which dehydroxilation (loss of OH groups) takes place, these being between 500 and 700 °C.

1.4 Bentonites dealt with in the report

Overall, the countries with a HLW disposal programme have selected a candidate or reference bentonite to be used as sealing material either alone or mixed with different additives. Usually, the material selected comes from the country itself, except in those with no natural occurrences of bentonite materials. In the latter case, commercial well-known products are selected, as it is the case of the MX-80 bentonite. This report is mainly focused on the bentonites most widely studied in Europe up to now in the context of HLW disposal:

- The **FEBEX** bentonite. It comes from the Cortijo de Archidona deposit (Almería, Spain), from which the S-2 bentonite was also extracted several years before. Both were selected by ENRESA (Spain) as suitable material for the backfilling and sealing of HLW repositories. A comprehensive characterisation of it exists (ENRESA 2000, Villar 2000, 2002, Fernández 2003, among others). The processing at the factory consisted in disaggregation and gently grinding, drying at 60 °C and sieving by 5 mm. The smectite content of the FEBEX bentonite is higher than 90 percent (92±3 %). The smectitic phases of the FEBEX bentonite are actually made up of a montmorillonite-illite mixed layer, with 10-15 percent of illite layers. Besides, the bentonite contains variable quantities of quartz (2±1 %), plagioclase (2±1 %), K-felspar, calcite and opal-CT (cristobalite-trydimite, 2±1 %). The less than 2 μm fraction of this bentonite is 67±3 % of the total. The CEC varies from 96 to102 meq/100g, and the major exchangeable cations are: Ca (35-42 meq/100g), Mg (31-32 meq/100g), Na (24-27 meq/100g) and K (2-3 meq/100g). The liquid limit of the bentonite is 102±4 %, the plastic limit 53±3 % and the specific weight is 2.70.

- The **MX-80** bentonite. It is extracted from Wyoming (USA), and has been selected in many disposal concepts as backfilling and sealing material (Sweden, Finland, Germany, France). It is a worldwide known material supplied in the form of powder homoionised to sodium (SKB reports, Müller-Vonmoss & Kahr 1983). The MX-80 bentonite is composed mainly by montmorillonite (65-82 %) and it also contains quartz (4-12 %), feldspars (5-8 %), and smaller quantities of cristobalite, calcite and pyrite. The less than 2 μm fraction of this bentonite is 80-90% of the total. The CEC is 74 meq/100g, and the major exchangeable cations are: Na (61 meq/100g), Ca (10 meq/100g) and Mg (3 meq/100g). The liquid limit of the bentonite is 350-570 %, the plastic limit 70 % and the specific weight is 2.82.

- The **FoCa** bentonite. It is a sedimentary clay coming from the Paris Basin (extracted in the Vexin region, Fourges-Cahaignes) that was the reference sealing material in France and Belgium for several years (CEA reports, Tessier *et al.* 1998). The major component (85-90 %) is an interstratified clay of 50 % Ca beidellite and 50 % kaolinite. It also contains quartz (6 %), goethite (6 %), calcite (2 %), kaolinite, hematite, and gypsum. The less than 2 μm fraction is 75 percent of the total. The industrial process consists in drying, grinding and sieving (max. grain size: 2 mm). The CEC is 57-71 meq/100g, and the major exchangeable cations are Ca (50 meq/100g), Mg (5 meq/100g), Na (2 meq/100g) and K (0.6 meq/100g). The liquid limit of FoCa is 98-112 %, the plastic limit is 33-50 %, and the specific weight is 2.68.

2 THERMO-HYDRO-MECHANICAL CHARACTERISTICS OF BENTONITES

As mentioned in the previous sections, the material constituting the barrier is a high density compacted bentonite. Consequently, it is a highly expansive remoulded material. The emplacement of these blocks under the conditions of a disposal facility will subject them to thermal and hydraulic gradients. The fact that the blocks are not initially saturated, along with the low permeability of the bentonite, means that a relatively long transient period is foreseen, during which a part of the material will be unsaturated.

Consequently, the characterisation of the barrier material implies, among others, the determination of thermal, hydraulic and mechanical properties of compacted bentonite, which requires the use of experimental techniques adapted to the study of highly expansive soils, in both the saturated and unsaturated states. The following sections review the state of the art in these areas and summarises the knowledge on the THM properties of bentonites.

2.1 Thermal properties

For the thermal analysis of the barrier, the following thermal properties must be known: specific heat (heat capacity), thermal conductivity and thermal expansion coefficient. The first two are easy to determine, while the measurement of the latter presents technical difficulties not yet solved. These are necessary parameters to determine the dissipation of the heat generated by the waste, and consequently they have an influence in the design of the repository concerning the separation between canisters and galleries.

For the FEBEX bentonite, a relation between specific heat and temperature exists in the temperature range between 45 and 150 °C (ENRESA 2000). The linear thermal expansion coefficient of the FEBEX bentonite compacted to dry densities of 1.60 and 1.70 g/cm^3 with water contents between 12.5 and 25.0 percent has been measured for temperatures between 25 and 70 °C. It has been seen that this coefficient varies in heating/cooling cycles (Lloret *et al.* 2002).

However, the most important parameter to explain heat transport in the barrier –which takes place mainly by conduction, specially in the saturated stages– is the thermal conductivity. Thermal conductivity is the capacity of a soil to conduct a quantity of heat in the presence of a temperature gradient. In homogeneous soils uniformly moist, heat flow may be assumed to be conductive and equal to a thermal conductivity times a temperature gradient (Fourier equation), where it is implicitly assumed that thermal conductivity includes the thermal vapour flux induced by the temperature gradient (Buchan 1991).

Thermal properties in general, and thermal conductivity in particular, depend on the physical composition of the soil, especially on the water content, and to a lesser extent on the density, since both properties condition the quantitative relationship between the different phases of the soil (solid, water and air) and each of these phases contributes to the macroscopic thermal conductivity. The increase in thermal conductivity with the water content for a given density (and consequently porosity) is due to the fact that the thermal conductivity of water (0.58 W/m·K) is 23 times higher than that of air (0.025 W/m·K), as a result of which the increase in the water content of the pores causes an increase in the overall thermal conductivity. Furthermore, this increase is not linear. This is because below a given value of water content, water is adsorbed or forms a film over the mineral particles. For higher water contents, filling of the pores begins, with water "bridges" being formed between the voids filled with air, on account of which the thermal conductivity increases in a significant amount. Above a certain value of water content, the increases in water content have a slight effect on the value of conductivity (Tindall & Kunkel 1999). The increase in density contributes also to the increase in thermal conductivity, due to the fact that the decrease in porosity implies a reduction of the space occupied by water or air, components that both have a much lower conductivity than the mineral particles. On the other hand, the thermal conductivity of the solid is determined by its mineral composition. Since there are no major differences among the thermal conductivity of various smectites, the main changes in the thermal conductivity of the solid phase of a bentonite may arise as a consequence of the quantity and type of accessory minerals present and of the use of additives such as quartz and graphite, both of them increasing the thermal conductivity.

With respect to the influence of dry density and water content on the thermal conductivity, there are well established empirical relationships relating these parameters (or porosity, degree of saturation), both for the FEBEX (Figure 8, Villar 2002), the MX-80 (Figure 9, SKB 1999) and the FoCa (Figure 10, Beziat *et al.* 1988) bentonites. In these relationships the thermal conductivities of the dry and the saturated bentonite are usually present.

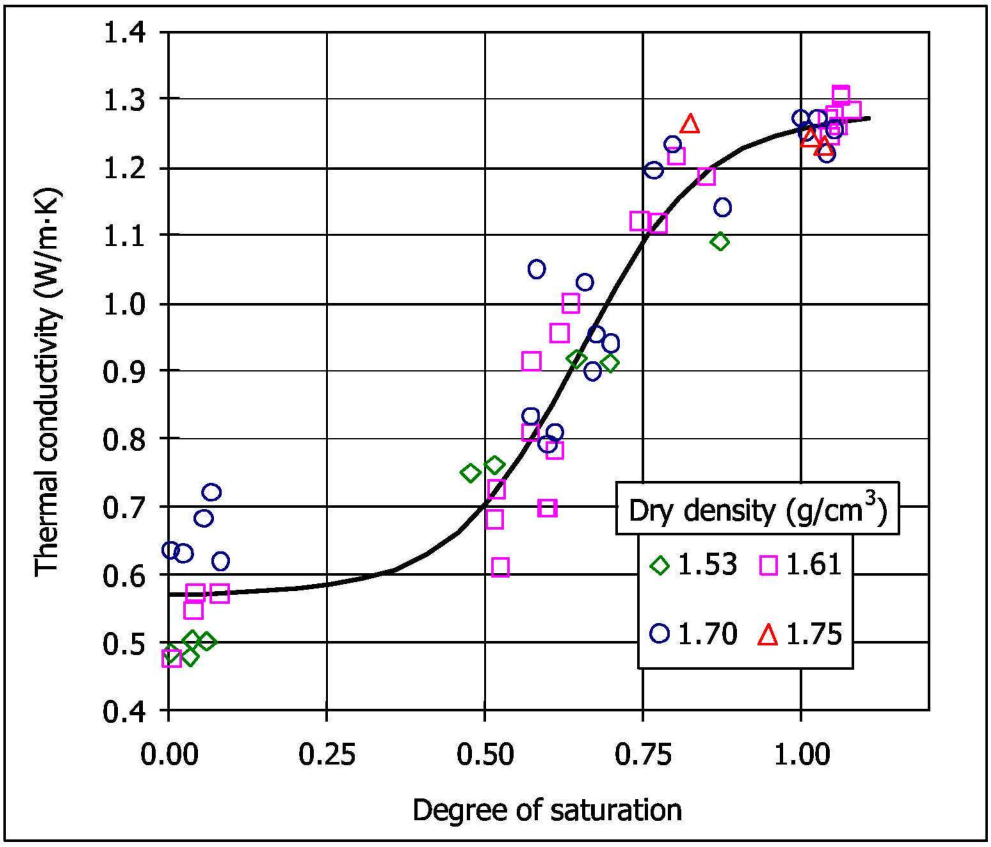

Figure 8: Thermal conductivity of compacted FEBEX bentonite as a function of the degree of saturation (Villar 2002)

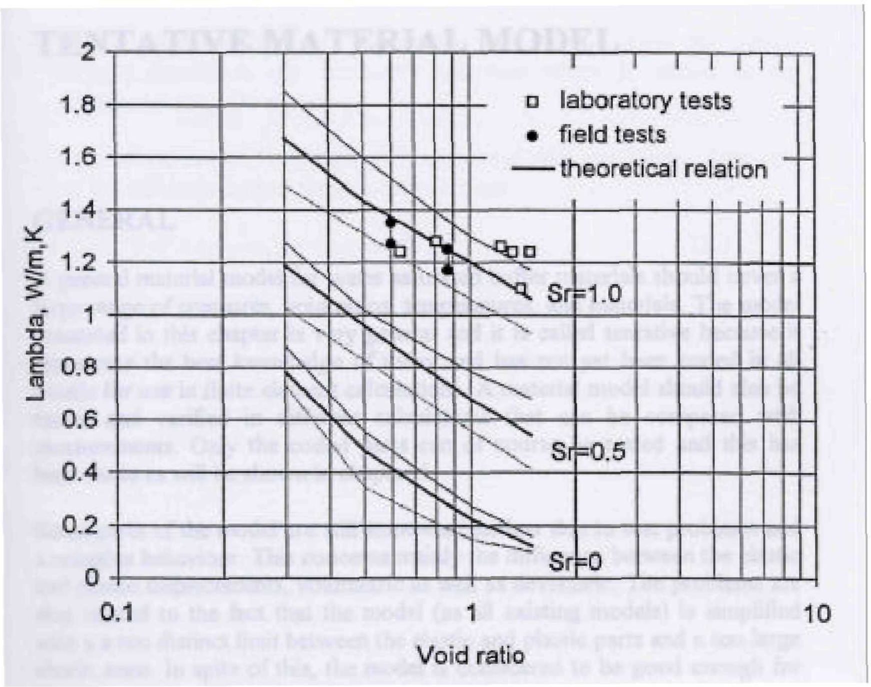

Figure 9: Measured thermal conductivity of the MX-80 bentonite with a degree of saturation higher than 90% as a function of the void ratio (Börgesson *et al.* 1995)

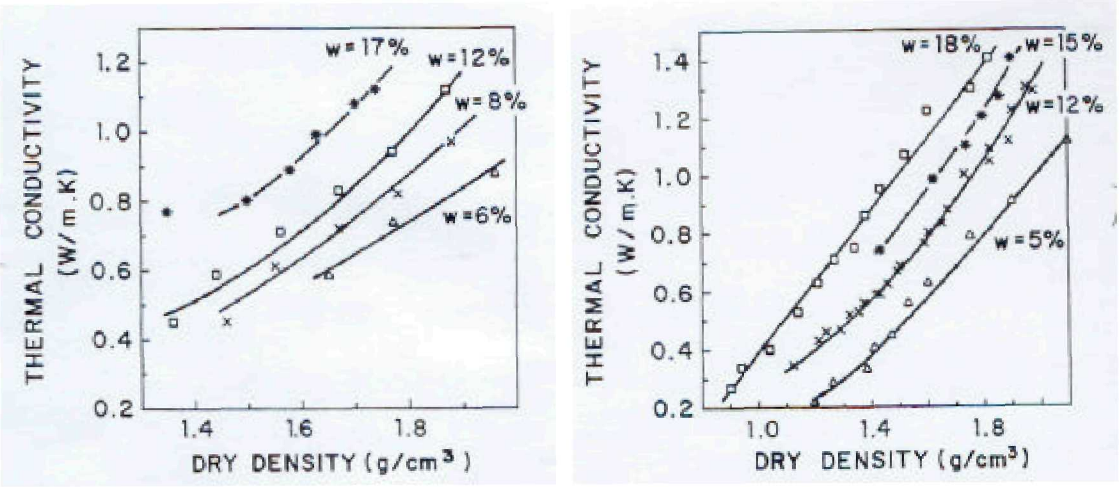

Figure 10: Thermal conductivity measured at laboratory temperature of samples of Na smectite (left) and Ca smectite (FoCa clay, right) compacted to different dry densities and water contents (Beziat *et al.* 1988)

Likewise, there is a certain dependence of thermal conductivity on temperature, although this is insignificant within the range of temperatures typical of soils. Beziat *et al.* (1988) found that the increase in the thermal conductivity of a calcic smectite on increasing temperature from 50 to 188 °C is around 10 percent, much smaller than that observed when the water content or the density is varied (Figure 11). Börgesson *et al.* (1994) claim that the slight increase of the thermal conductivity of the MX-80 bentonite with increasing temperature is not sufficiently verified.

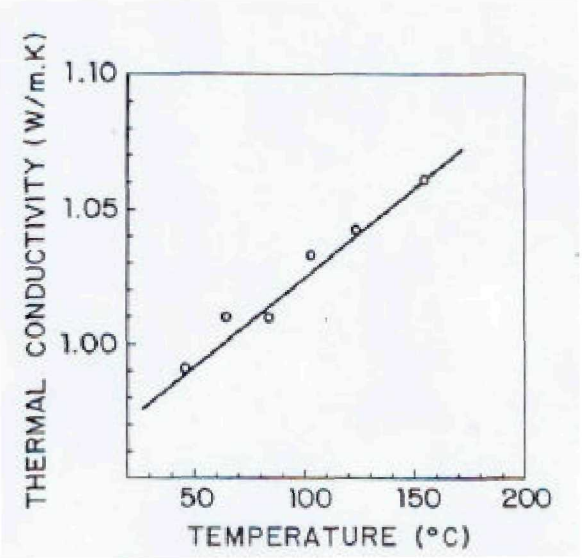

Figure 11: Thermal conductivity of a Ca smectite as a function of temperature ($\rho_d = 1.99$ g/cm^3, w = 0%) (Beziat *et al.* 1988)

On the other hand, measurements performed in compacted FEBEX bentonite have shown that its thermal conductivity is independent of the direction of measurement, parallel or perpendicular to compaction (Villar 2002). Other forms of preparation of the bentonite barrier, such as the use of high density pellets, have been scarcely explored with respect to thermal properties. A recent work by Sugita *et al.* (2003) presents measurements of the thermal conductivity of Kunigel V1, a japanese Na bentonite, prepared as compacted blocks and as pellets (Figure 12). The thermal conductivity of the pellets is slightly lower.

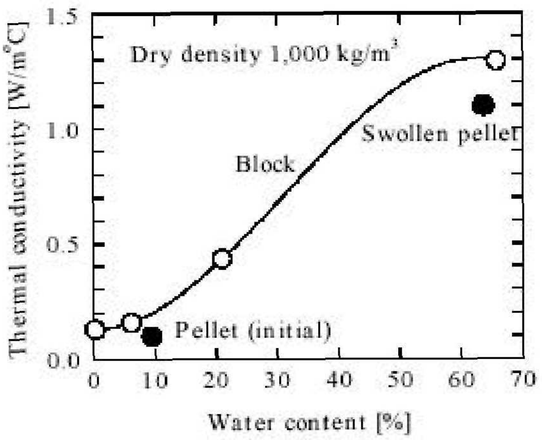

Figure 12: Thermal conductivity of the Kunigel V1 bentonite in the form of blocks and pellets (Sugita *et al.* 2003)

2.2 Mechanical properties: volume change

The mechanical properties of the bentonite determine the strength and deformability of the barrier and have important implications on the design of the repository. Among the properties that must be taken into account are the compressive and tensile strength, swelling, compressibility and elastic shear modulus. All the mechanical properties depend on the dry density and the water content and, hence, they are determined in compacted blocks of different dry density and water content. The compressive strength must be known to predict the behaviour of the blocks during the construction of the barrier. But the most outstanding mechanical characteristic of bentonites is their capacity to change volume, the laminar constitution of smectites described in section 1.3 being responsible for the peculiar hydro-mechanical behaviour of the soils in which they are contained. Since the mechanical behaviour of the installed barrier is mainly determined by its volume change characteristics, this section is devoted to them.

It is generally accepted that various mechanisms intervene in the expansion of a soil, and they can be classified in three main interrelated groups (Mitchell 1976): attraction of water by the clay particles due to the negative charge of their surface, cation hydration, and osmotic repulsion. The main factors that affect the expansion of a soil may be divided into three groups. The first group concerns the intrinsic properties of the particles and the conditions of the soil mass, and consequently includes the type of clay mineral and its proportion, dry density, water content, the orientation of the particles and the type and quantity of cations adsorbed (Pusch 1979). The second group refers to the composition and properties of the saturation water. Finally, external factors such as the soil stress history, temperature and the availability of water also influence swelling.

If an expansive material is saturated at a constant volume while its deformation is prevented, the particles will exercise a pressure on the confining structure. This is known as swelling pressure and reaches its equilibrium value when the sample is completely saturated. Since bentonite is placed in the repository in unsaturated conditions, and due to its high suction pressure (see section 'Water retention"), it will take water from the host rock and swell. If bentonite is unable to expand freely, as is the case in most of the barrier, a swelling pressure develops. Swelling in turn modifies the porosity of the clay and consequently its hydraulic and diffusion properties, as discussed in section 2.3.1.1.

On the other hand, compression (*i.e.* the application of a load) causes an excess air and water pressure in the pores that is dissipated through consolidation, which induces the volume of a soil to decrease as the water is expelled from the pores. The movement of the water exiting from the internal pores of the soil to the exterior is controlled by Darcy's law, and causes a reduction in volume that depends on the interaction between the total and effective stresses, pore pressure, flow, hydraulic gradient and permeability.

The volume change characteristics of a soil may be studied by means of triaxial or oedometric tests. The limitation of oedometric testing is that it does not allow complete insight to be gained into the stress state of the material, since there is no measure of the horizontal confining stress. As a result, difficulties are introduced in the interpretation of the results and in the comparison with the predictions of the models. In the case of unsaturated soils, there is also uncertainty regarding the degree of saturation of the soil, which is always unknown. However, the most significant trends and features of soil behaviour may be followed with this type of tests (Gens & Alonso 1992), the performance of which is far less complex than that of triaxial tests.

The swelling capacity of a material may be indicated by two parameters: the swelling pressure developed under confined conditions and the expansion (or strain) upon saturation. Both are determined in oedometer equipments under different stress conditions in samples compacted to different dry density and with various water contents. Different types of water may be used to saturate the specimens, and the determinations can be carried out at various temperatures, thus exploring the volume change behaviour of the bentonite under conditions representative of those in a repository.

2.2.1 Effect of intrinsic properties of the bentonite on volume change

The swelling capacity and the swelling pressure of a bentonite depend greatly on its content of smectite (Figure 13) and the type of adsorbed cations (Figure 14). For a given bentonite, the strain upon saturation depends mainly and inversely on the load applied during saturation, with which logarithmic relationships may be established (see Figure 15 for FEBEX bentonite). Besides, the increase of the dry density rises the swelling deformation of the bentonite exponentially (see the same Figure) and the swelling pressure (see Figure 16 for FEBEX bentonite and Figure 17 for MX-80 bentonite), although the increase of the former is not as remarkable.

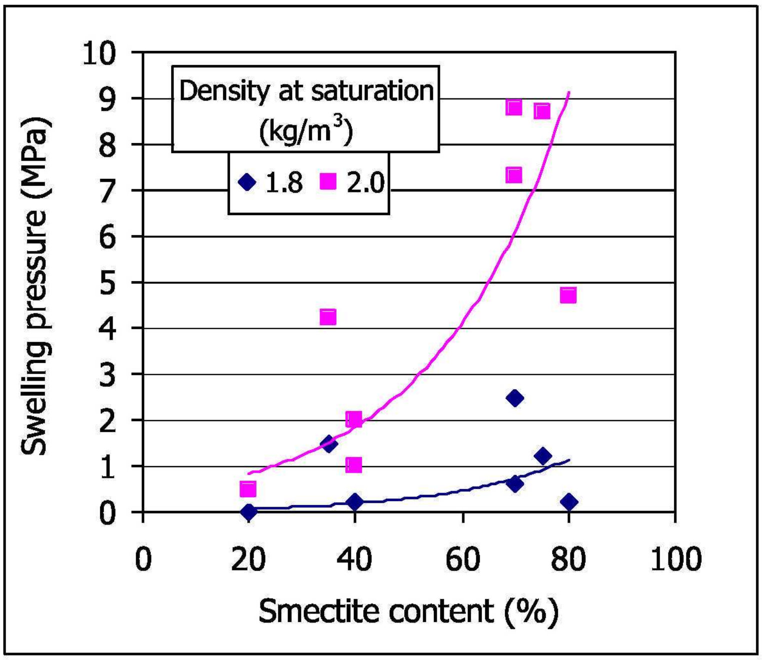

Figure 13: Swelling pressure as a function of the smectite content for various bentonites and bentonite/sand mixtures (data from Pusch 1995)

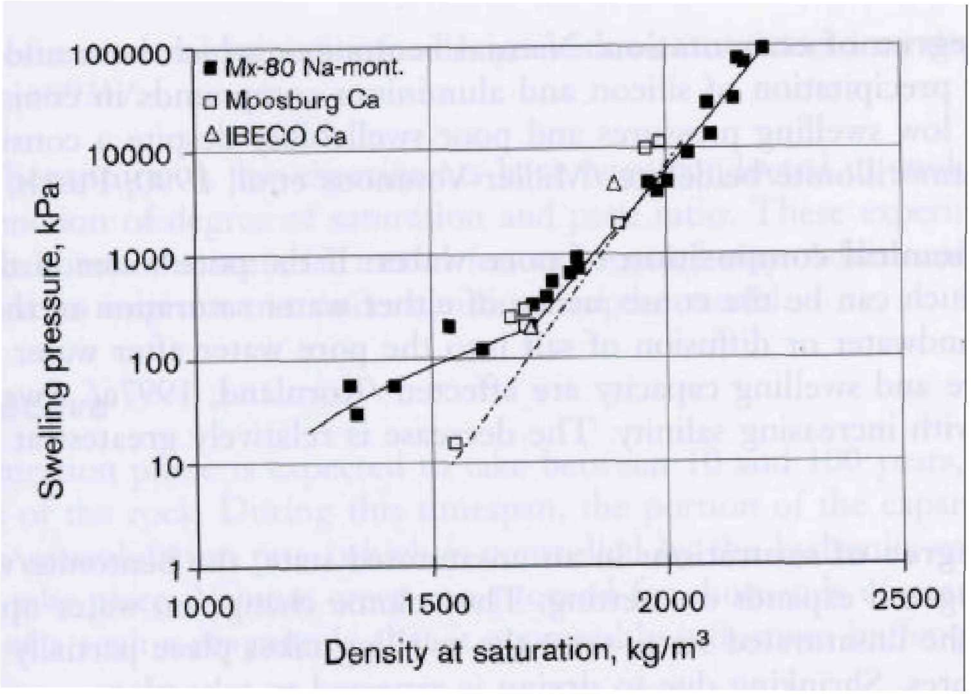

Figure 14: Dependence of swelling pressure on density and adsorbed ion species (SKB 1999)

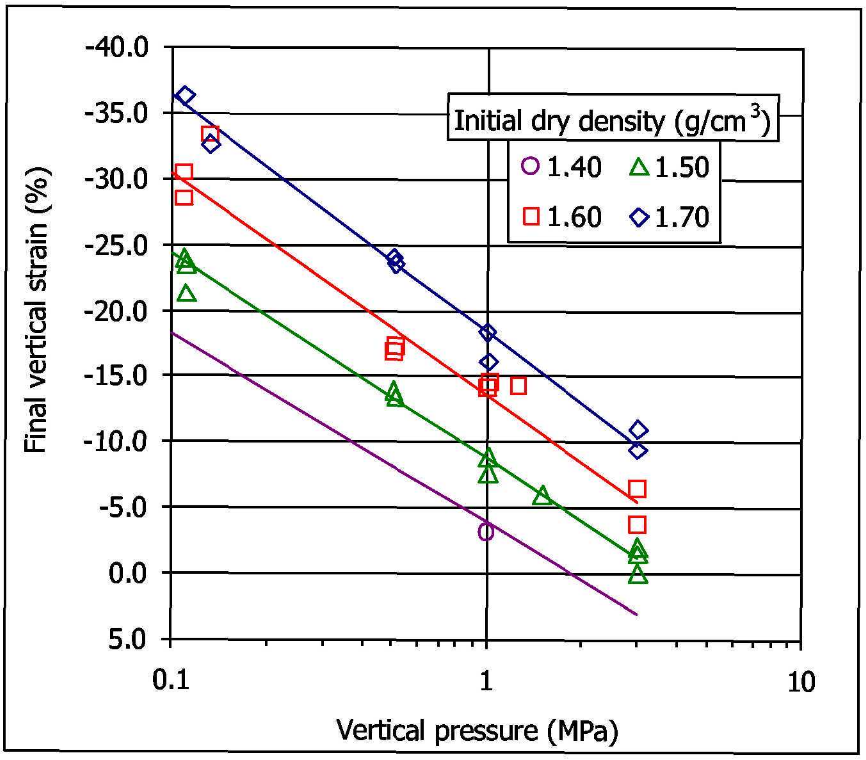

Figure 15: Vertical strain after saturation with deionised water under different vertical loads for FEBEX bentonite compacted with its hygroscopic water content at various dry densities (Lloret *et al.* 2004)

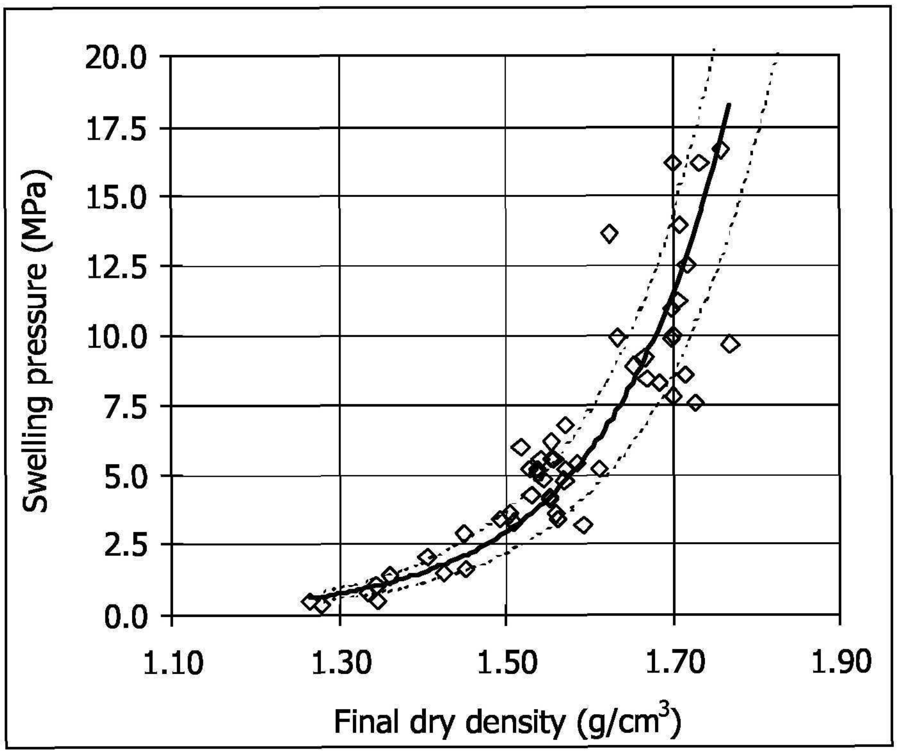

Figure 16: FEBEX bentonite swelling pressure versus dry density at the end of the test, obtained for saturation with deionised water, and exponential fit (Villar 2002)

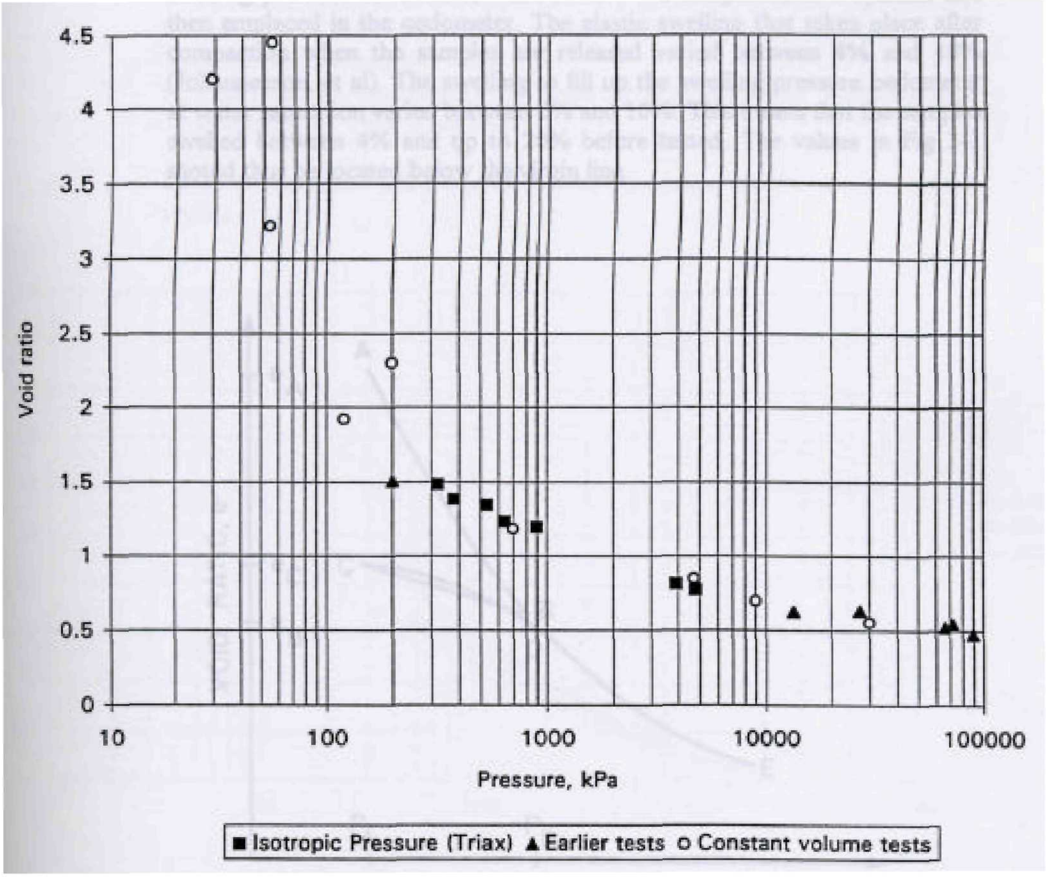

Figure 17: Measured relation between swelling pressure and void ratio for MX-80 (Börgesson *et al.* 1995)

Conversely, the swelling pressure does not seem to be affected by the initial water content of the clay, a fact that was checked for the FEBEX Ca bentonite and other less plastic clays (Sridharan *et al.* 1986). On the contrary, the swelling deformation for a particular dry density after saturation under a fixed vertical pressure decreases with the initial water content of the clay (Figure 18). However, the influence of the initial water content on the swelling capacity of the FEBEX bentonite is smaller than the influence of the initial dry density. Moreover, the influence of the initial water content on the swelling deformation becomes negligible above a given vertical pressure, which is higher the higher the dry density. This value of vertical pressure is very close to the swelling pressure value. Komine & Ogata (1994) found that the maximum swelling deformation and swelling pressure of a compacted sodium bentonite are almost independent of the initial water content but that they increase in proportion to the initial dry density, as can be observed in Figure 19 for the swelling pressure.

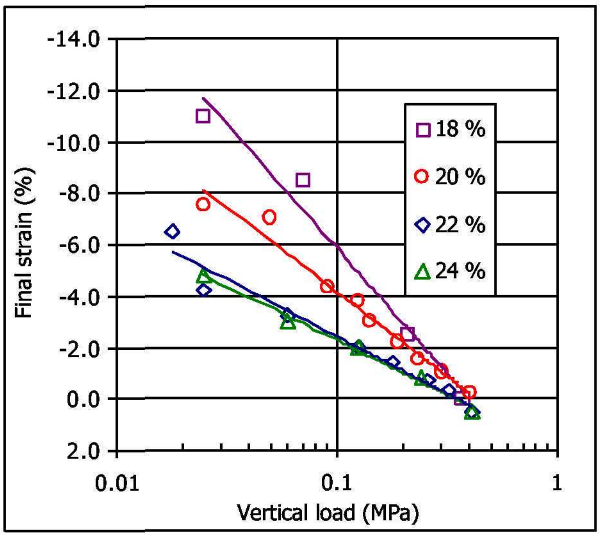

Figure 18: Final strain of soaking tests performed with a plastic clay compacted to an initial dry density of 1.5 g/cm^3 with different initial water contents (data from Kassiff *et al.* 1973)

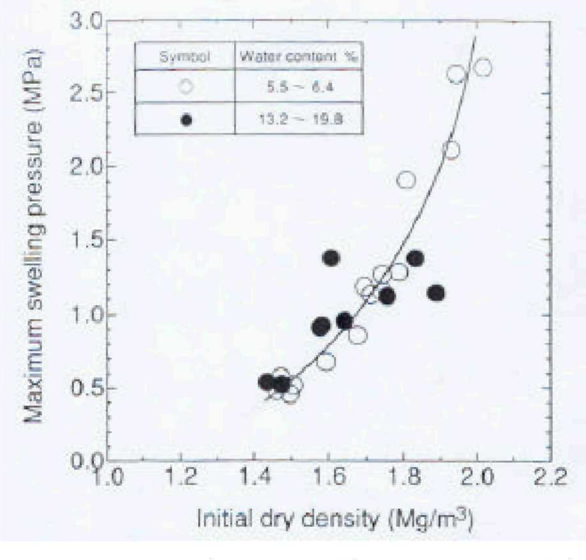

Figure 19: Relationship between maximum swelling pressure and initial dry density as a function of the initial water content for a Na bentonite (Komine & Ogata 1994)

On the other hand, the kinetics of the development of the swelling pressure is not well known, and it may vary with the density of the bentonite and with manufacturing aspects of the barrier, such as the use of compacted blocks or pellets. In the context of the RESEAL I and II Projects, some of these aspects have been studied by means of infiltration tests performed in specially designed oedometers. The development of the swelling pressure in pellets/powder mixtures shows three phases (Figure 20): a first one with a quick swelling pressure increase, a second one with either a quasi-constant level or even a decrease of the swelling pressure, and the last one with a new increase of the swelling pressure. The interaction between micro and macrostructure accounts for this pattern. At saturation and for equal density, precompacted samples and pellets/powder samples display the same swelling pressure (Volckaert *et al.* 2000, Van Geet *et al.* 2003).

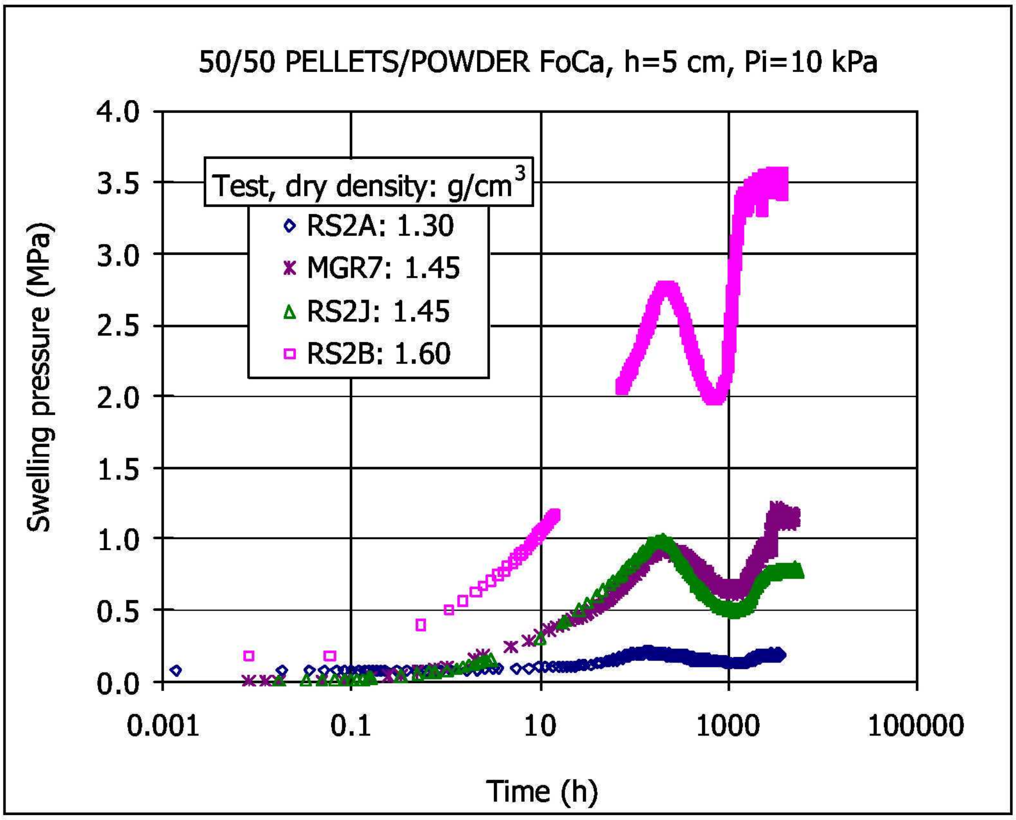

Figure 20: Evolution of the swelling pressure in tests performed with pellets/powder mixtures (50/50 wt %) of FoCa clay by CEA and CIEMAT in the context of RESEAL II Project

2.2.2 Effect of the water salinity on volume change

Swelling tests performed with different kinds of water have shown that the swelling capacity of the bentonite saturated with saline water reduces as the salinity of the solution increases, and osmotic consolidation was even observed. Although it is generally accepted that the swelling pressure decreases with the saline content of the solution, for montmorillonite compacted at high density the influence of the salinity of the water on the value of the swelling pressure is negligible (Pusch 1994). Indeed, Karnland *et al.* (2003) have measured reductions of the swelling pressure of MX-80 bentonite with salinity which are relatively lower as the clay density is higher (Figure 21). The same has been observed by Sugita *et al.*

(2003) in Kunigel V1 bentonite (Figure 22). However, if the salinity of the solution is very high, the decrease of the swelling may be noticeable even for high bentonite densities: Figure 23 and Figure 24 show the effect of highly saline solutions on the swelling pressure and swelling strain, respectively, of compacted FEBEX bentonite. The same bentonite develops slightly lower swelling strains upon saturation with low-concentrated saline water (0.8 percent) than with deionised water, although this difference becomes less evident for the higher vertical loads (Villar *et al.* 2002). The swelling pressure of less expandable materials (like Friedland Ton clay, with 45 percent smectite) is insensitive even to high salt content at relatively high clay densities (Figure 25, Pusch 2001).

Besides, the type of exchangeable cations in the exchange complex has an influence on the effect of salinity on swelling properties. Thus, in Na bentonites, in which the number of stack contacts and therefore of interacting electrical double layers is much larger than in Ca bentonites (see section 1.3), there is a stronger influence of pore water salinity on swelling, since the contribution of osmotic swelling is higher in Na bentonites than in Ca bentonites, in which swelling has a predominant interlaminar component (Pusch *et al.* 1990).

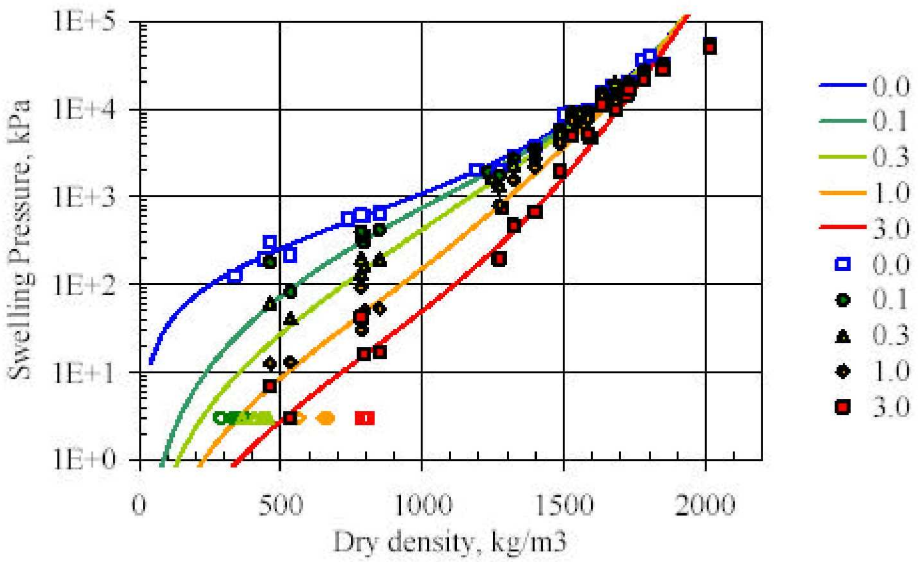

Figure 21: Measured (dots) and calculated (lines) swelling pressure *vs*. clay dry density. Legend shows external solution concentration in mol/L (Karnland *et al.* 2003)

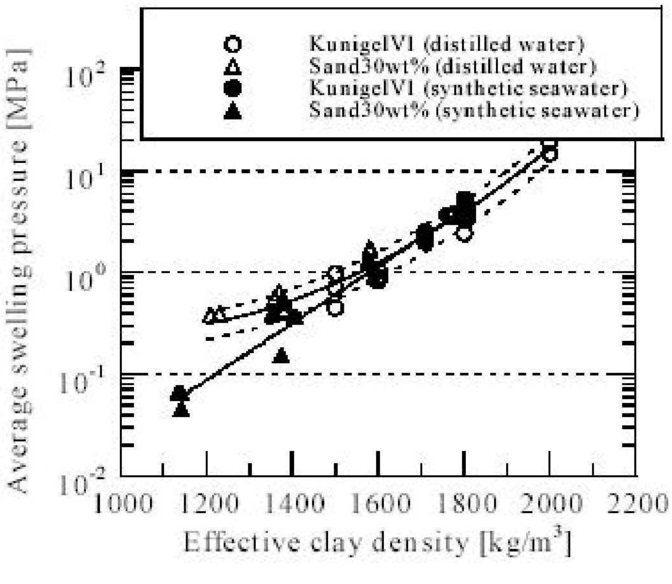

Figure 22: Variation of swelling pressure with dry density for Kunigel V1 bentonite saturated with distilled and saline water (Sugita *et al.* 2003)

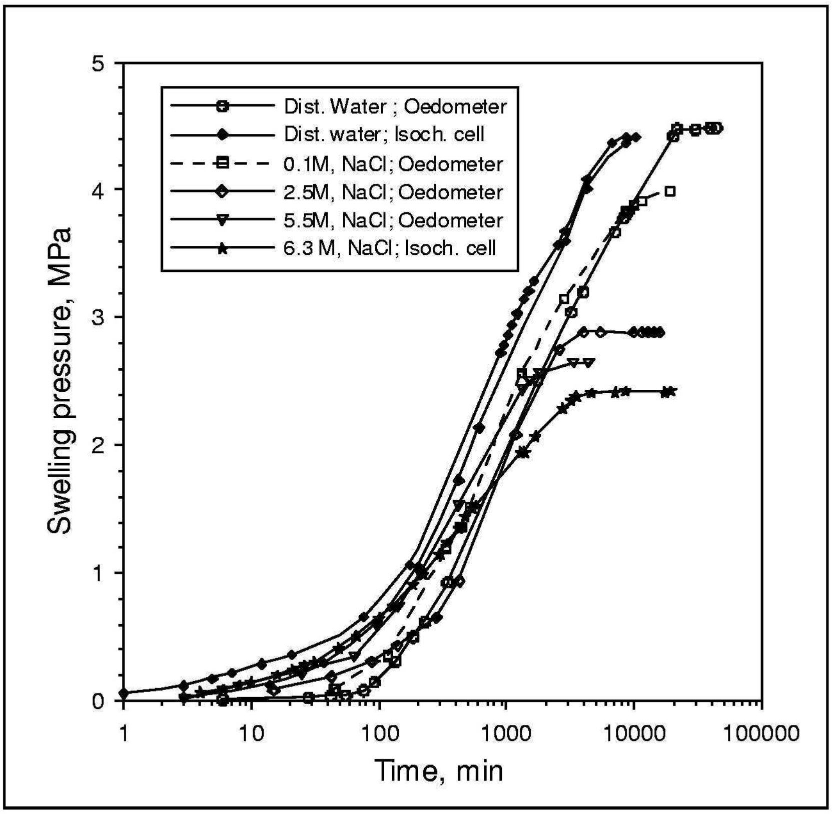

Figure 23: Evolution of swelling pressures measured in FEBEX bentonite compacted at dry density 1.65 g/cm^3 saturated with water with different concentrations of NaCl (Lloret *et al.* 2004)

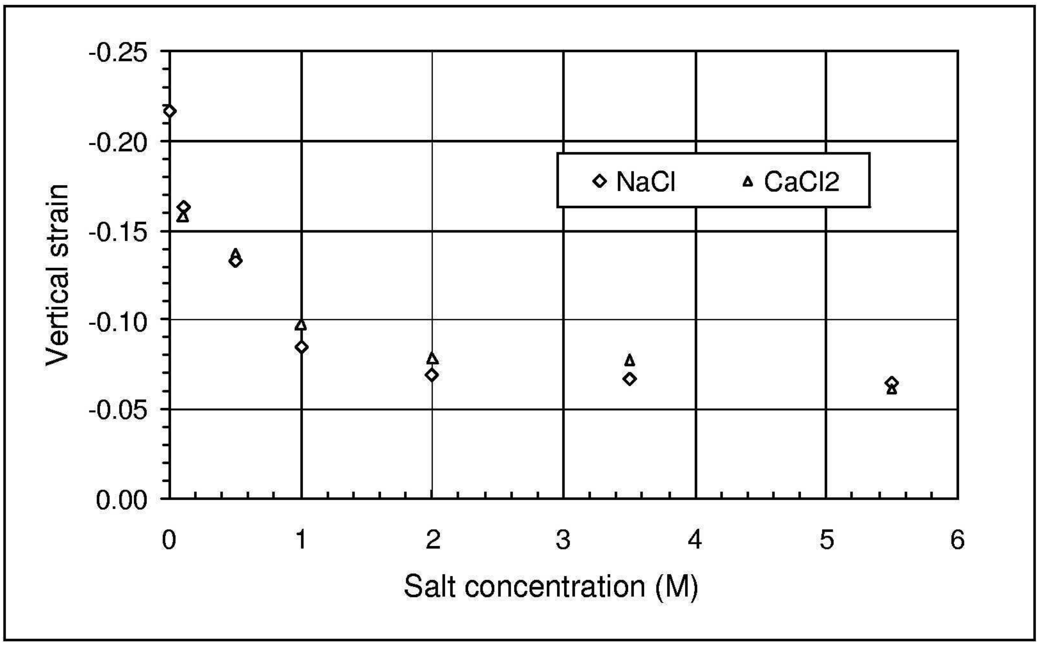

Figure 24: Variation of swelling strains due to wetting of FEBEX bentonite with different solutions under a vertical stress of 0.5 MPa. Initial dry density 1.65 g/cm^3 (Lloret *et al.* 2004)

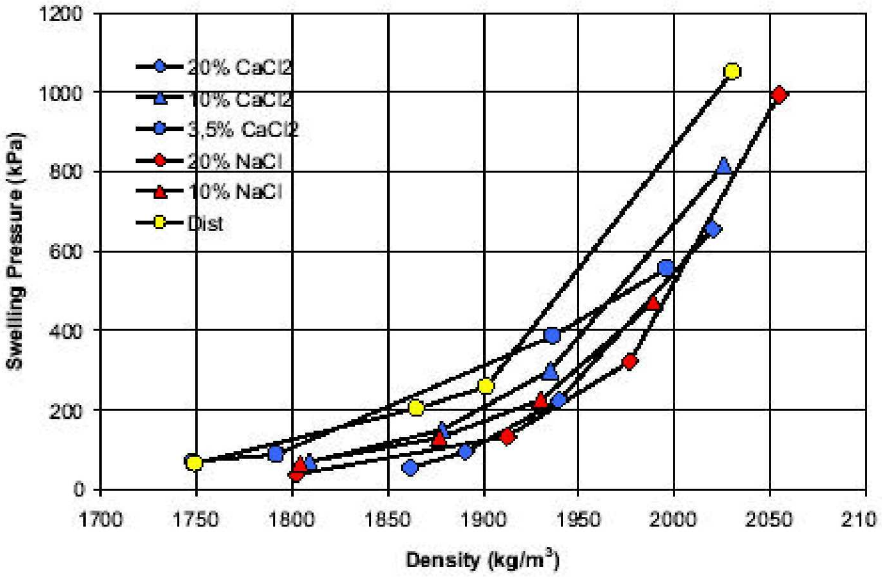

Figure 25: Swelling pressure of samples of Friedland Ton clay saturated with different solutions (Pusch 2001)

Compressibility is also affected by the salinity of the solution. For the FEBEX bentonite it has been observed that the specimens saturated with high-concentration solutions are less deformable and consolidate more quickly than those saturated with low-concentration solutions (Lloret *et al.* 2004). However, for granite/bentonite mixtures (70/30 weight percentage) no influence of the salinity (for salinities of up to 16 g/L) on the mechanical response of the material has been observed (Mata & Ledesma 2003).

2.2.3 Effect of the suction on volume change

One of the main external factors affecting the volume change behaviour of the bentonite is the availability of water, which in turn is related to the suction level. The mechanical behaviour of the unsaturated compacted FEBEX bentonite has been investigated by the performance of a testing programme carried out in suction-controlled oedometers capable of applying high suctions and large vertical loads (Villar 2002, Pintado 2002, Lloret *et al.* 2002). Two types of tests have been performed: i) tests with simultaneous control of vertical stress and suction, and ii) suction-controlled swelling pressure tests. The stress paths followed were selected with the dual objective of simulating the conditions of different parts of the clay barrier in the disposal facility and of obtaining parameters quantifying its behaviour. The examination of the results has revealed significant features of behaviour such as yield phenomena, dependency of swelling strains on applied stresses, stress path dependency of strains, and compound stress paths in swelling pressure tests. It has been observed that saturation induces a decrease in the apparent preconsolidation pressure (Figure 26) and that the samples with hygroscopic water content (suctions around 100 MPa) are very stiff for vertical pressures of up to 8 MPa. It has also been observed that the swelling pressure depends greatly on the suction level, *i.e.* on the clay water content, and it sharply increases as suction decreases, specially at the beginning of hydration, when suction is high (Figure 27).

In spite of the complexity of the behaviour observed, a good understanding of it is possible by considering explicitly the fabric of the soil that has been independently studied revealing a dual arrangement with microstructural and macrostructural components. A deeper insight of the behaviour of the compacted bentonite, and of the basic mechanisms controlling it, has been achieved using an elasto-plastic model that incorporates the interplay between microstructural and macrostructural fabric levels in a simplified manner. Most of the main features of behaviour are correctly reproduced by the model allowing a more detailed examination of the role that the different variables play on the explanation of the overall behaviour of the soil (Lloret *et al.* 2003).

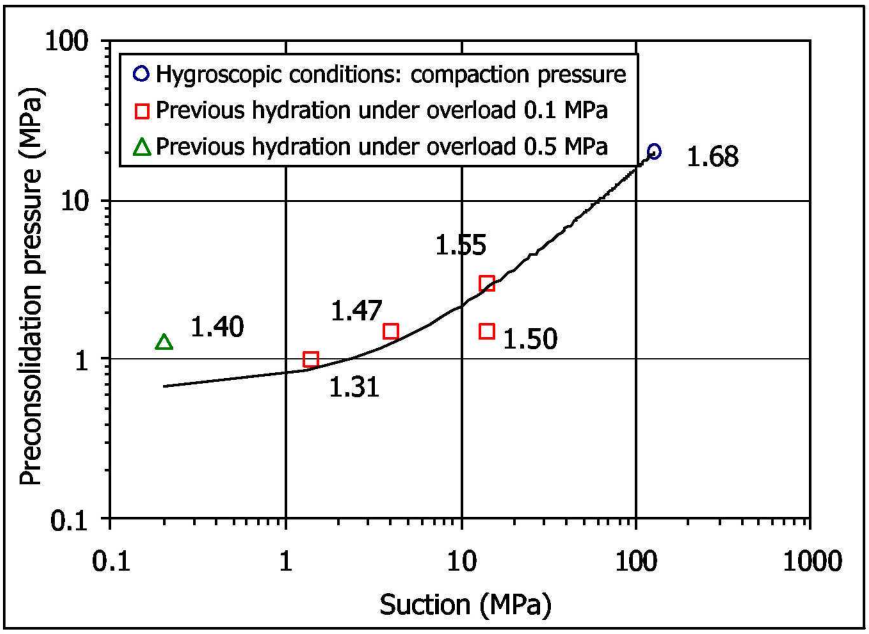

Figure 26: Preconsolidation pressure for different tests of loading under constant suction performed with FEBEX bentonite, indicating dry density at the end of the first loading step in g/cm^3 (Villar 2002)

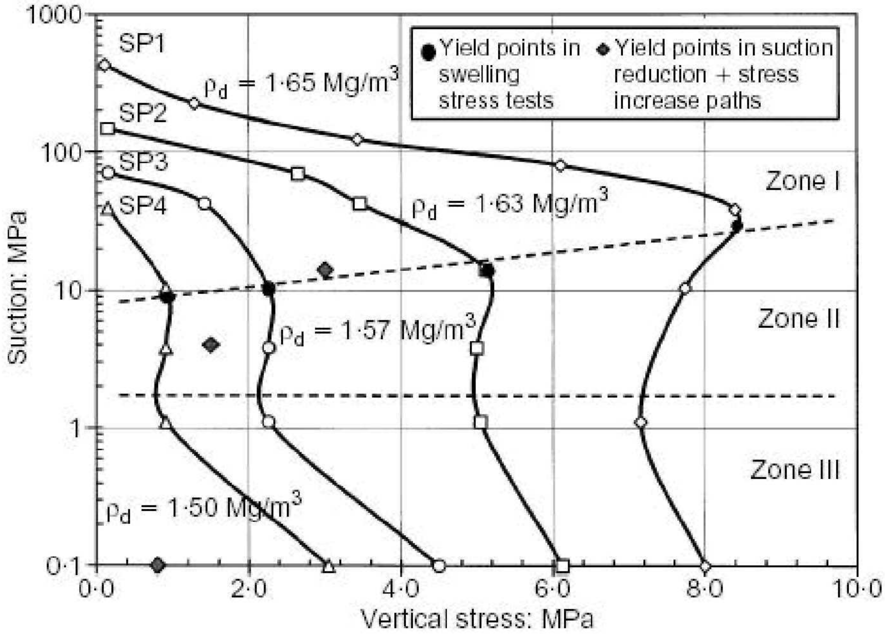

Figure 27: Generalised stress paths observed in the swelling pressure tests with FEBEX bentonite (Lloret *et al.* 2003)

On the other hand, consolidation tests performed in suction control oedometers with a silt/bentonite mixture showed that the parameters $\lambda(s)$ and $p_0(s)$ cannot be considered constant as suction increases from 1.2 to 152.9 MPa, while κ remains constant in the same range of suction (Cuisinier & Masrouri 2001).

2.2.4 Effect of the temperature on volume change

The mechanical response of the material is also affected by temperature (swelling pressure, swelling and collapse, thermal dilatation and contraction, compressibility, yielding, effects on time dependent behaviour). The influence of temperature on the mechanical behaviour of a geological material depends on its overconsolidation ratio, plasticity, expansibility and compaction water content. According to this, the temperature may have different opposite effects: swelling/retraction, softening/stiffening, decrease of elastic domain/overconsolidation. These effects may in turn be reversible or irreversible. Besides, the volume change induced by the temperature increases with the plasticity of clays (Demars & Charles 1981). In the last decade, equipment and experimental techniques were developed to test soils under different temperatures (Belanteur *et al.* 1997, Romero 1999). In relation to radioactive waste disposal, a number of laboratory results referring to thermal effects on saturated expansive soils have been presented in the last years (Towhata *et al.* 1993, Sultan *et al.* 2002, Burghignoli *et al.* 2000). Results focused on the thermal influence on volume change behaviour of unsaturated soils are still limited (Wiebe *et al.* 1998, Romero *et al.* 2003). In particular, information concerning the temperature effects on the mechanical behaviour of highly expansive clays in unsaturated conditions is still scarce (Lingnau *et al.* 1996, Romero *et al.* 2001a, Villar & Lloret 2003).

According to Pusch *et al.* (1990), the effect of heating on the swelling pressure of saturated montmorillonite differs depending on the predominant cation in the exchange complex: the swelling pressure increases in Na bentonite while it decreases in Ca bentonite. The reason invoked is that the net effect of two mechanisms –lattice contraction due to dehydration of the interlamellar space and increase of the osmotic pressure at stack contacts– is different for the two types of clay. Lattice contraction predominates in Ca bentonite, while the increase in osmotic pressure predominates in Na bentonite, where the number of stack contacts is higher (see section 1.3). The same authors pointed out that the influence of heating on the swelling capacity decreases as the confining stress increases. They observed that heating is not important for the development of swelling pressure at very high bulk densities, while it is rather significant at low bulk densities. On the other hand, the contribution of mineral thermal expansion phenomena to the overall volume change upon saturation of bentonite is minor.

In particular, at high temperatures the swelling capacity of the FEBEX clay –a Ca bentonite– decreases (Figure 28), and a clear decrease of swelling pressure as a function of temperature (down to 2.5 MPa at 80 °C) has been observed (Figure 29).

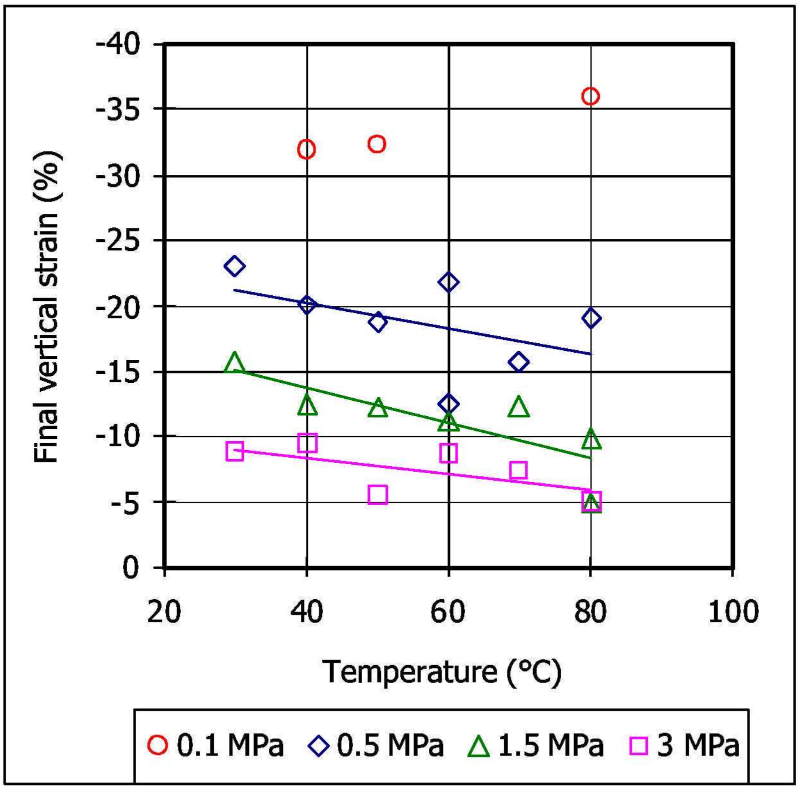

Figure 28: Final strain of FEBEX samples compacted to dry density 1.60 g/cm^3 saturated with deionised water under different vertical pressures and temperatures (Lloret *et al.* 2004)

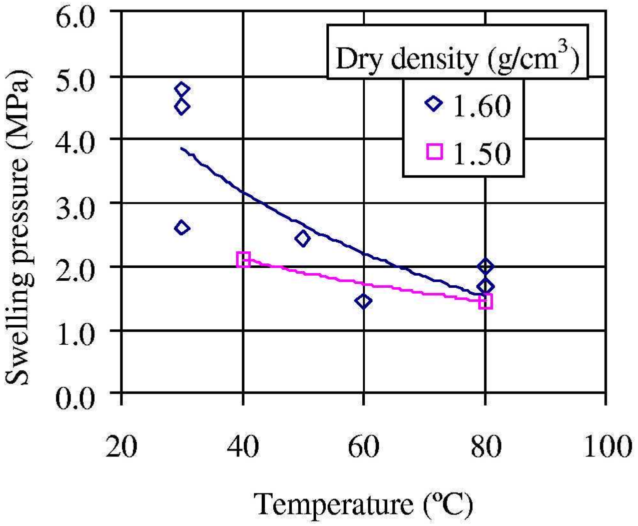

Figure 29: Effect of temperature on swelling pressure of FEBEX bentonite for two different dry densities (Villar & Lloret 2003)

Decreases of swelling pressure with temperature were also measured in other materials: beidellite (Karnland *et al.* 1994), sand/bentonite mixtures (Lingnau *et al.* 1996) and Boom clay (Romero *et al.* 2003). On the other hand, increases in swelling pressure with temperature were found for saponite and Na montmorillonite (Karnland *et al.* 1994, Figure 30).

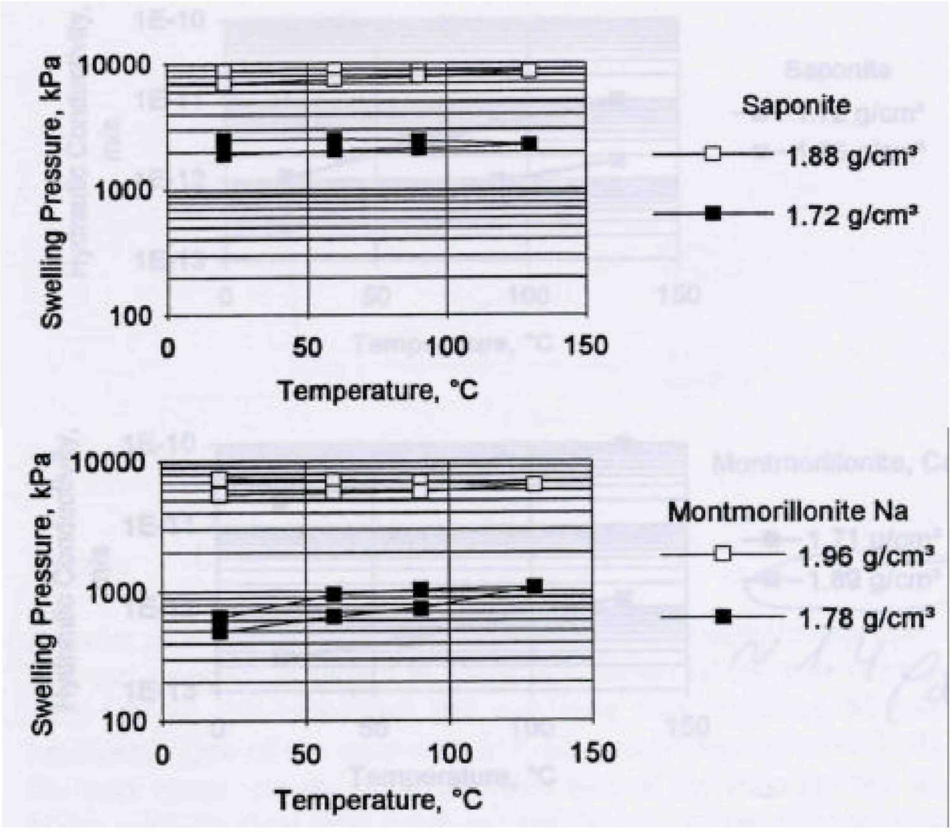

Figure 30: Measured swelling pressure at different temperatures and bulk densities (Karnland *et al.* 1994)

Other aspects of the mechanical behaviour of bentonite regarding temperature have been studied through suction controlled and temperature controlled oedometric tests. The oedometric tests performed with the FEBEX bentonite have shown that the decrease in the apparent preconsolidation pressure induced by saturation (see previous section) is independent of the temperature of the tests (Villar & Lloret 2003, Lloret *et al.* 2004). The high stiffness upon loading of samples compacted with hygroscopic water content is also independent of temperature, which indicates that the effect of the temperature on the elastic behaviour of the bentonite is small. On the other hand, the temperature increases the compressibility of the bentonite (Figure 31), both saturated and unsaturated, what would imply that a certain reduction in the size of the elastic domain takes place with temperature. Nevertheless, it seems that the deformation of the bentonite is more dependent on the stress and suction path than on temperature. In tests with suction reduction, the structure changes due to hydration are more relevant in the subsequent mechanical behaviour of the bentonite than the effects of temperature.

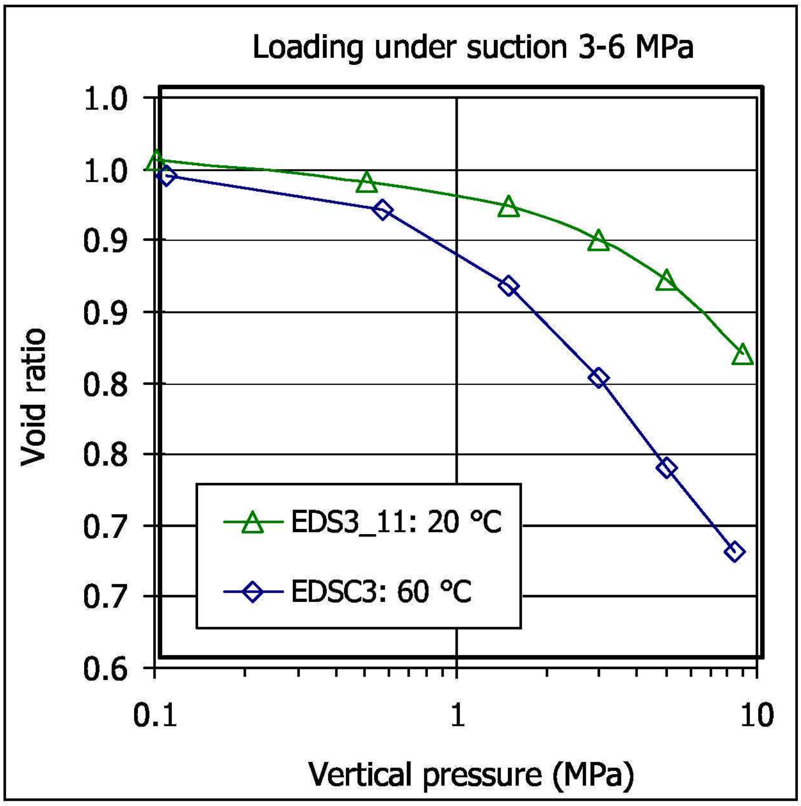

Figure 31: Evolution of void ratio in the different steps of loading paths performed at different temperatures with FEBEX bentonite compacted to initial ρ_d 1.70 g/cm^3 (modified from Lloret *et al.* 2004)

2.3 Hydraulic properties

The hydraulic properties control the movement of water inside the buffer, which, according to Darcy's law, is due to the head gradient. The hydraulic gradient in the repository can be imposed by the regional differences in hydraulic head or by the suction pressure exerted by the unsaturated bentonite. Consequently, the main hydraulic properties to be considered are the hydraulic conductivity and the water retention capacity (suction / water content relationship).

2.3.1 Permeability

The clay blocks of an engineered barrier are –in most repository concepts– initially unsaturated, and an important percentage of their pores (40-50 %) will be filled with air. Thermal and hydraulic gradients will be generated in the repository, due to the heat emitted by the wastes and to the resaturation of the near field. Consequently, a combined movement of gas and water will take place in the clay barrier.

The flow of water is governed by Darcy's law, the coefficient of permeability depending on the degree of saturation. In any soil, permeability to water increases with the degree of saturation. Taking this into account, the relative permeability is defined for a given porosity and degree of saturation as the ratio between the unsaturated permeability for this degree of saturation and the saturated permeability (or hydraulic conductivity) for the same porosity; therefore, it takes a value between 0 and 1. It is generally accepted that relative permeability is related to the degree of saturation in accordance with a potential law.

On the other hand, air flow is governed by the Fick's law which, when the air is considered to be a perfect gas, may be also assimilated to Darcy's law, with a coefficient of permeability to gas. Given that there are no important interactions between the air and the other constituents of the soil, air flow depends only on the air pressure gradient and on the volume of air in the soil, which, in turn, depends on the degree of saturation. The flow of air requires the continuity of the gas phase. Water-filled pores block gas flow, consequently, gas permeability decreases as soil water content increases.

There is also the intrinsic permeability, which depends only on soil structure. The value of the intrinsic permeability is generally associated to the pore diameter and pore size distribution. Qualitatively, the effect of the pore size and total porosity on the intrinsic permeability may be evaluated through Poiseuille's equation. Ideally, the intrinsic permeability has the same value for gas and liquid flow. This value can be obtained either from gas flow tests in the totally dry soil, or from water flow tests in the saturated soil, and should be the same regardless the fluid employed in its determination. However, if fluid-media interactions alter the medium structure, the intrinsic permeability can be greatly altered. This is the case of expansive soils like bentonite, in which water reacts with clay minerals causing the swelling of the clay lattice, thereby reducing the pore space available for flow (Tindall & Kunkel 1999).

It is generally accepted that the main factors having a significant influence on the permeability value are the liquid degree of saturation, the void ratio, the temperature and the salinity of the permeant in the case of liquid fluid. There is also the discussion about the possible influence of the hydraulic gradient applied to determine permeability on the results obtained. All these aspects are discussed in the following sections.

2.3.1.1 Variation of the permeability with the void ratio

Given that intrinsic permeability, and consequently hydraulic conductivity, is related to the pore diameter of the material by the Poiseuille equation, the decrease in the average diameter of the pores as the dry density of a material increases, gives place to a permeability decrease that can be described through the Kozeny's expression. However, in expansive materials this decrease of permeability is higher than predicted through this expression, as is represented in Figure 32 for the FEBEX bentonite. The increase of the dry density of the bentonite gives rise to an exponential decrease in saturated water permeability (and of intrinsic permeability,) this being due to the fact that, as the swelling capacity of the clay increases, there is an additional reduction in the size of the flow channels and, therefore, in effective porosity and permeability. In the FEBEX bentonite, the decrease in hydraulic conductivity with increasing density is sharper for high dry densities (Villar 2002). The existence of two intervals of density for which the decrease in hydraulic conductivity with increasing density is different was also found by Villar & Rivas (1994) for the S-2 bentonite (from the same deposit than the FEBEX clay). This was interpreted as being caused by the sharp increase in swelling pressure for porosities below a given value (Figure 16), which leads to a more significant decrease in the size of the flow paths from porosities below this threshold value (Figure 32).

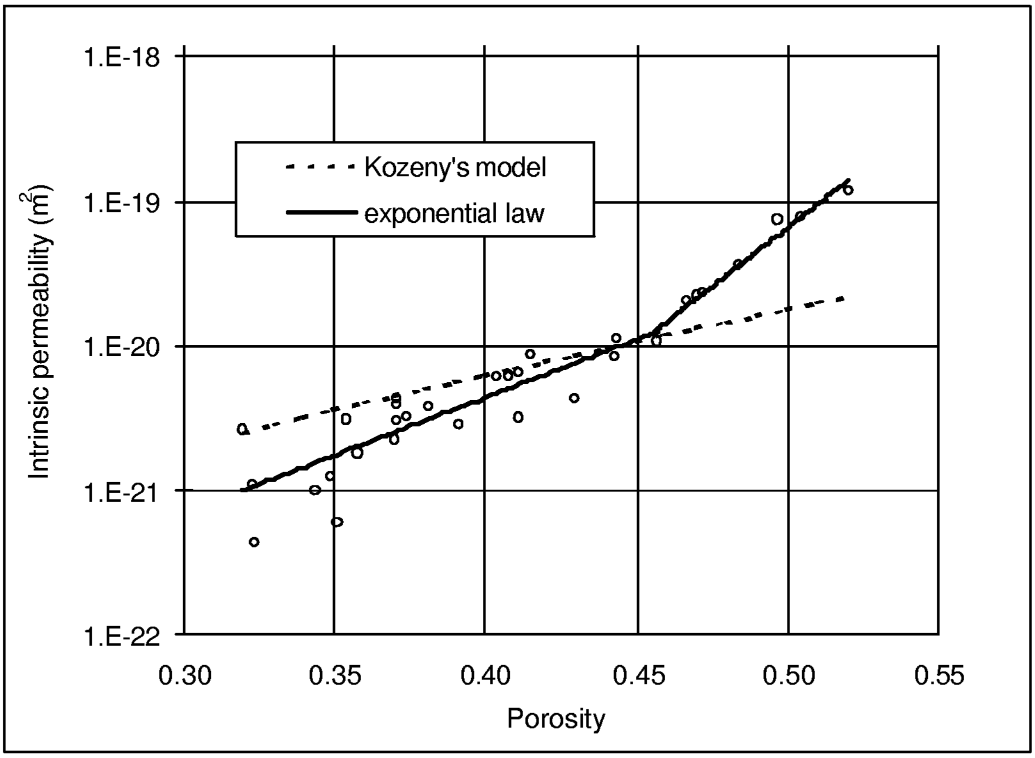

Figure 32: Intrinsic permeability of saturated FEBEX bentonite for different porosities (Lloret *et al.* 2002)

Pusch (1979) explains the extremely low permeability of the bentonite compacted at high density in terms of the low water contents required to saturate the highly compacted clay, since under these conditions, the thickness of the film of interlaminar water is very small (3-5 Å) due to its high specific surface. This means that the molecules of water are strongly adsorbed onto the surfaces of the clay minerals, leaving only very tortuous interparticle channels for the transport of water. Besides, in the Ca montmorillonite compacted at dry densities in excess of 1.60 g/cm^3, more than 90 percent of the total volume of the pores is occupied by this interlaminar water of restricted mobility (Pusch *et al.* 1990, Figure 33). Nakaoka *et al.* (2004) claim that the higher water viscosity in the vicinity of the clay surface significantly contributes to the relation between void ratio and permeability in bentonite, and evaluated this influence through a molecular dynamics model.

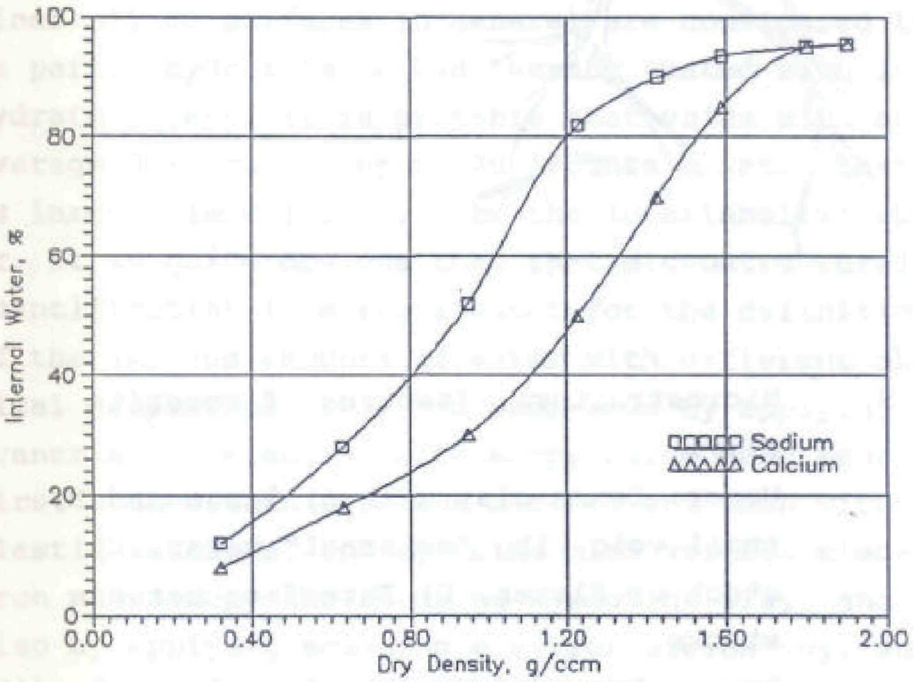

Figure 33: Theoretical relation between the dry density of smectite and the content of "internal" water expressed as percent of the total pore volume (Pusch *et al.* 1990)

2.3.1.2 Variation of the permeability with the degree of saturation

The variation of permeability with the degree of saturation can be obtained by gas permeability tests. For a FoCa bentonite (70%) /sand (30%) mixture, the air permeability decreases slowly as the degree of saturation increases, but when degrees of saturation higher than 85 percent are reached, the air permeability decreases drastically (Figure 34). The values obtained for the FEBEX bentonite show that, for each water content, the gas permeability decreases as dry density increases, following an exponential law (Figure 35). But the permeability to gas is mainly related to the accessible void ratio, which indicates the ratio between pore volume accessible to gas and the particle volume and can be expressed as the product of the void ratio (e) by the unit minus the degree of saturation (S_r). This factor, e (1-S_r), shows in fact a higher correlation with the permeability values than the dry density or the water content do (Villar & Lloret 2001).

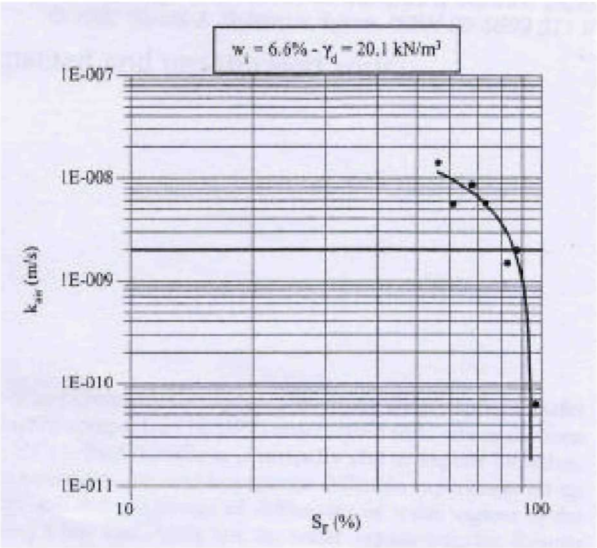

Figure 34: Air permeability *vs*. degree of saturation of a bentonite/sand mixture (Loiseau *et al.* 2002)

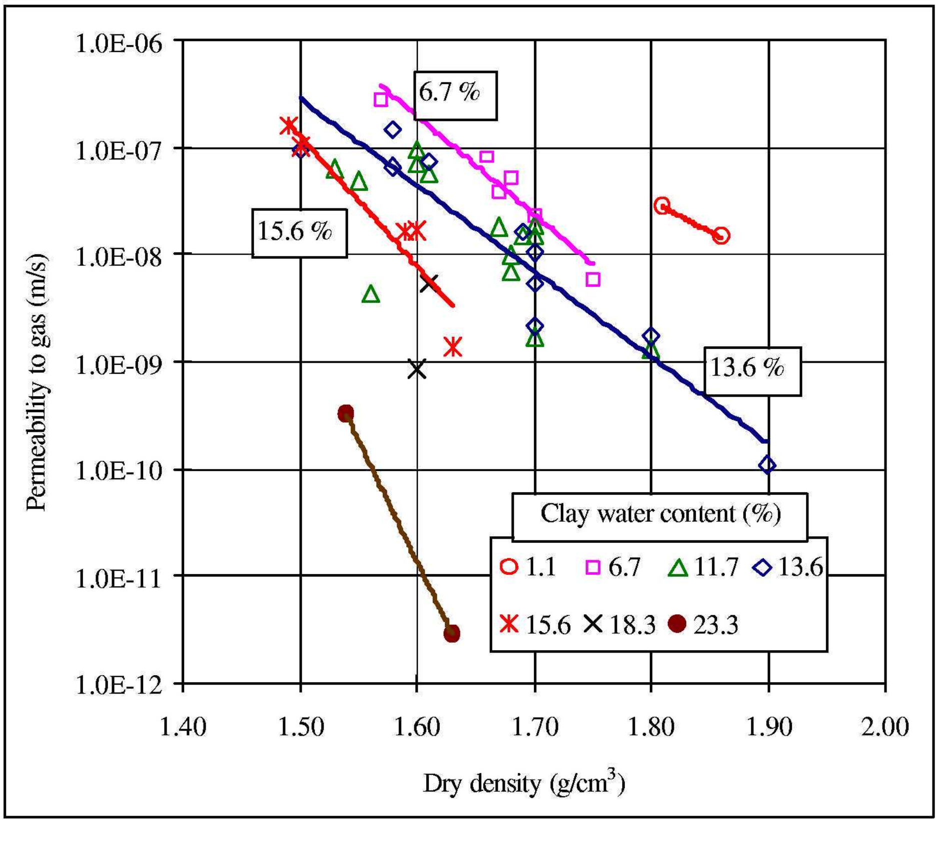

Figure 35: Permeability to gas versus clay density for FEBEX bentonite samples compacted at different water contents (Villar 2002)

The intrinsic permeability values derived from gas flow tests in FEBEX bentonite are plotted in Figure 36. This figure also shows the variation of the values of the intrinsic permeability obtained from water flow with the void ratio (a joint correlation for the two porosity intervals pointed out in Figure 32 is shown). For the same volume of pores, differences of about eight orders of magnitude in the intrinsic permeability values, for dry or saturated clay, can be observed. These observations suggest that a fundamental difference in the microstructural arrangement of the saturated and the unsaturated sample exists, due to the swelling of the clay as it saturates, which would not happen in non-expansive materials. The hydration of clay particles under constant volume reduces the size of the pores between the clay aggregates. In dry conditions, the diameter of macropores accessible to gas flow may be up to more than 10 µm. The clay hydrated in confined conditions presents the same global volume of pores than the dry clay, but the spaces between aggregates have been reduced or eliminated due to the swelling of the clay particles. That is to say, during the hydration of the clay at constant volume, the volume occupied by small mesopores and micropores increases, while the volume occupied by macropores decreases. In saturated conditions, accessible mean pore diameter is in the range of meso and micropores, *i.e.* more than three orders of magnitude smaller than the big interaggregate pores. The change in the mean pore diameter available to flow explains the big differences between the values of measured intrinsic permeability for dry and saturated clay (Villar & Lloret 2001, Thomas *et al.* 2003). Besides, most of the water in a bentonite saturated under constant volume will be in the interlamellar space (Pusch *et al.* 1990), and the water molecules constituting this film will be strongly adsorbed to the clay mineral surfaces, what leaves only narrow, tortous interparticle passages for water and ion transportation.

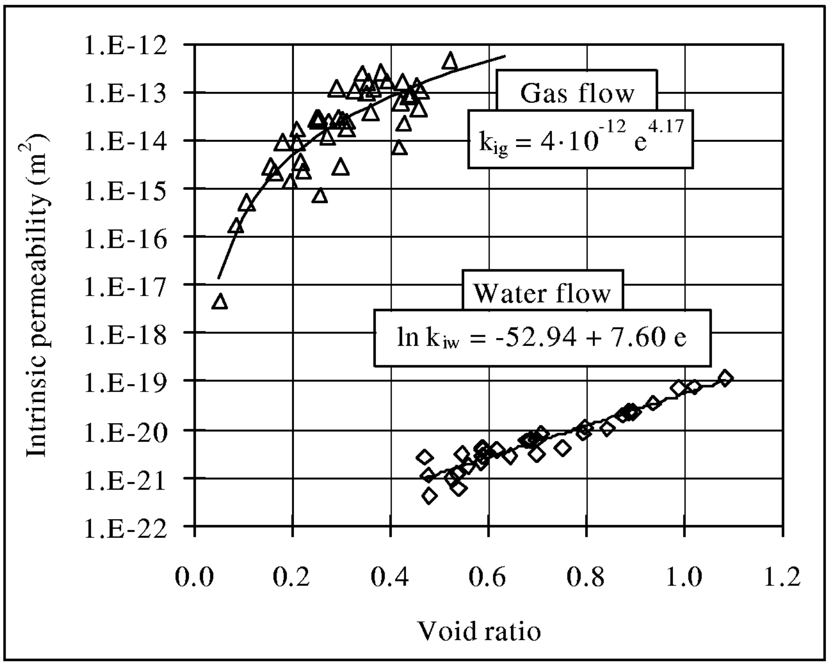

Figure 36: Intrinsic permeability of compacted bentonite obtained from saturated water flow and from unsaturated gas flow tests (Villar & Lloret 2001)

On the other hand, the methods used to determine permeability to water in the unsaturated state are based on the measurement or control of suction, this being necessary in order to determine the value of the gradient. Neither the steady-state nor the transient methods are easily applicable to measurement of unsaturated permeability of highly expansive materials. An alternative method to calculate the permeability of these materials is the back-analysis of infiltration tests performed under transient conditions, through the application of a theoretical model for solution of the inverse problem. To apply this technique, the water intake during the infiltration test must be measured, and it is also necessary to know the distribution of the water content inside the sample for different times after the infiltration process has started. Then, by means of parameter identification techniques similar to those used in groundwater engineering and geophysics, the intrinsic permeability and the exponent of the law used to obtain the relative permeability may be estimated. In addition, both the unsaturated water permeability and the tortuosity factor may be estimated from thermo-hydraulic experiments with prescribed heat and water flows.

Figure 37 shows the device used by UPC to identify by means of a backanalysis technique, the vapour tortuosity factor and the exponent of the unsaturated permeability law using water content measurements obtained at the end of a thermal flux test. Usually, the knowledge of the distribution of the water content in the clay is gained by means of destructive techniques (taking samples at various positions at the end of the infiltration test and determining their gravimetric water content by oven drying), which implies the performance of several tests of different duration (*e.g.* Fujita *et al.* 2001, Pintado *et al.* 2002). However, if a nondestructive technique is available, such as the dual-energy gamma ray attenuation, the spatial variation of water content and density of the clay can be measured "on line" all over a single experiment (*e.g.* Tabani *et al.* 2001, Figure 38). The latter technique is much less common, due to the experimental difficulties involved.

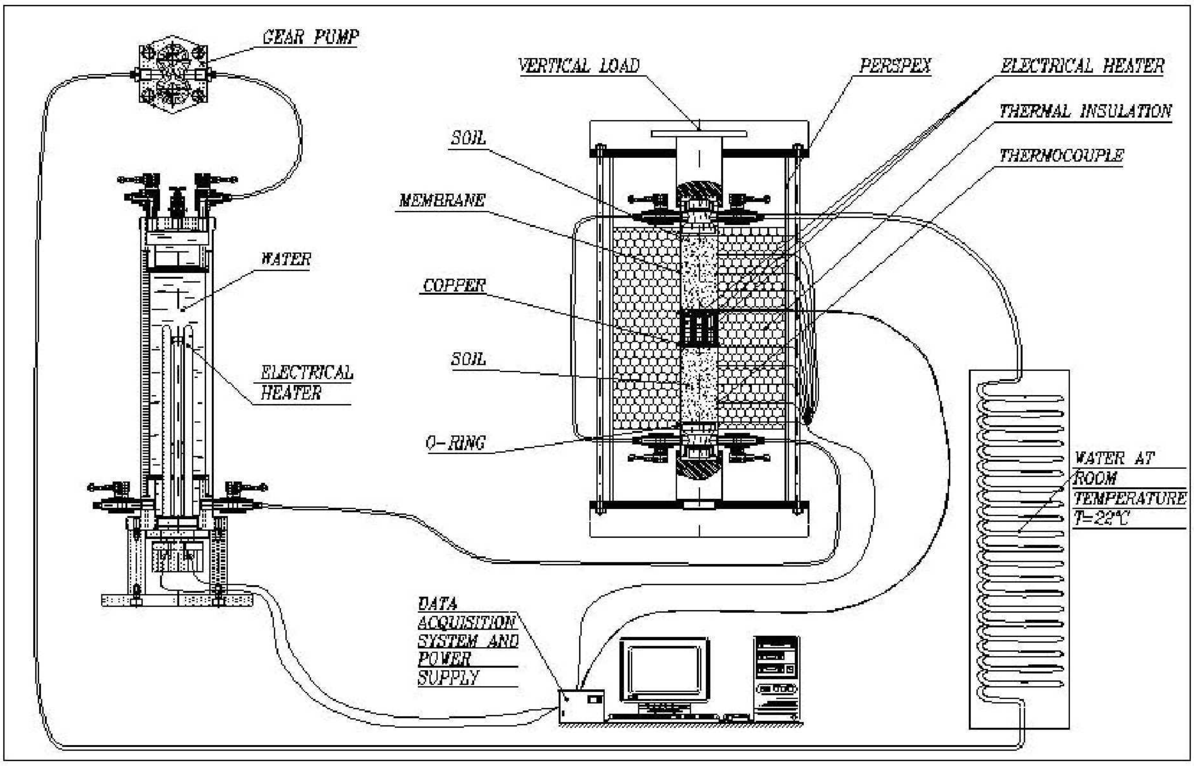

Figure 37: Scheme of the experimental device used to obtain the vapour tortuosity factor and the unsaturated water relative permeability exponent by backanalysis of a thermal flux test (Pintado *et al.* 2002)

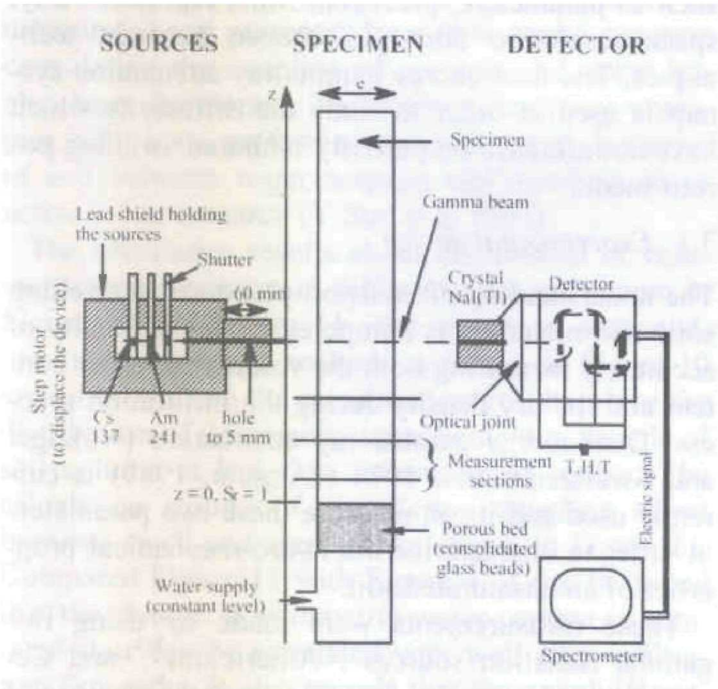

Figure 38: Experimental device for infiltration tests using dual-energy gamma ray attenuation (Tabani *et al.* 2001)

For example, infiltration tests with and without thermal gradient carried out by CIEMAT and UPC in compacted FEBEX bentonite have provided data allowing unsaturated hydraulic conductivity and its dependence on the degree of saturation to be obtained (ENRESA 2000, Lloret *et al.* 2002). In tests of different duration, the water content throughout the specimen was destructively measured at the end of the infiltration process. The effect of temperature on water viscosity and the effect of pore volume on intrinsic permeability allow to explain the different infiltration velocities observed in the infiltration tests performed under different temperatures and with different dry densities. As a result, the unsaturated permeability (k) can be expressed by the following expression:

$$k\ (S_r, T, \rho_d) = k_{iw}\ (S_r = 1, \rho_d = \rho_{d0})\ 10^{-3.6(\rho_d - \rho_{d0})}\ k_{rw}\ (S_r)\ (\gamma_w / \mu_w(T))$$

where k_{iw} is the intrinsic permeability obtained from water flow, T is the temperature, S_r is the degree of saturation, ρ_d is the dry density, ρ_{d0} is the reference dry density, μ_w and γ_w are the viscosity and specific unit weight of water and $k_{rw}(S_r)$ is the relative permeability. The values obtained for a porosity of 0.4 are: intrinsic permeability of $2 \cdot 10^{-21}$ m^2, exponent of the exponential law of 3 and tortuosity factor of 0.8.

On the other hand, the variation of the hydraulic conductivity of a Japanese bentonite as a function of the water content is shown in Figure 39 (Nakano *et al.* 1986). These results were obtained from the analysis of infiltration tests in the framework of a theory of water flow developed in a space coordinate system at rest, that assumes that the flux of the clay particles can be defined by the space gradients of potential responsible for the movement of clay particles, which is related to the gradients of water potential.

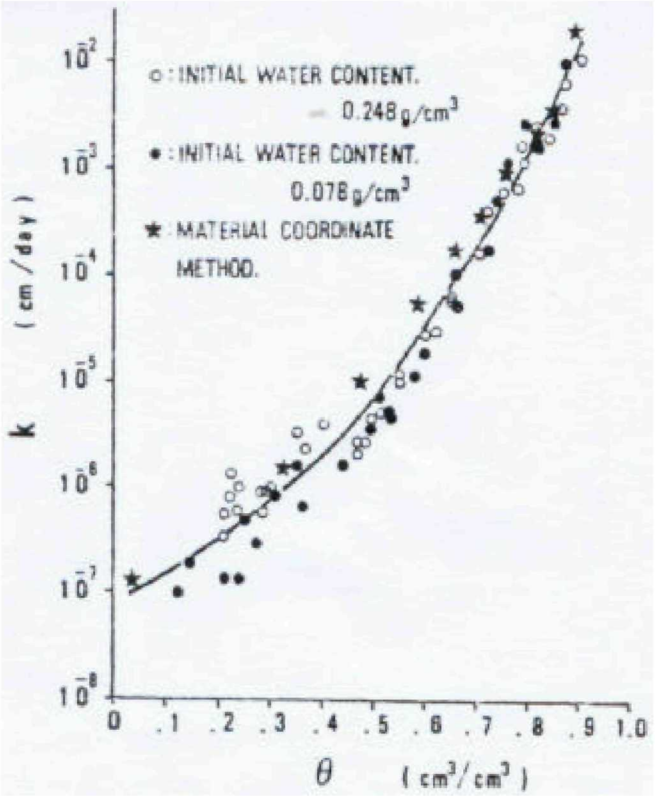

Figure 39: Hydraulic conductivity (k) as a function of volumetric water content (θ) for a Japanese bentonite (Nakano *et al.* 1986)

2.3.1.3 Influence of the hydraulic head on permeability – threshold gradient

The application of Darcy's law for the calculation of the permeability coefficient requires that the velocity of the flow be proportional to the hydraulic gradient, that is, the value of the coefficient obtained is independent of the hydraulic gradient applied during determination. Thus, the relation between flow and hydraulic gradient is linear, and that this linear relation passes through the origin. For different reasons, this condition may not be fulfilled, thus invalidating the use of Darcy's expression.

Pusch (1994) considers the hydraulic conductivity of bentonite to be independent of the hydraulic gradient applied to measure it. In fact, an analysis of the permeability tests performed with FEBEX bentonite (Villar 2002) has shown no dependence between the hydraulic gradient applied and the hydraulic conductivity obtained.

However, it has not been established whether or not the threshold hydraulic gradient pointed out by several authors exists, below which the relation between flow and gradient in clays deviates from linearity (Olsen 1962). For the FEBEX bentonite there seems to be a tendency to need gradients higher than 0 to have a measurable flow, which would point to the existence of a threshold gradient (Figure 40, Villar 2002, Villar *et al.* 2003). For sodium or lithium montmorillonite soils, no flow apparently exists below a threshold, possibly due to the non-Newtonian behaviour of the water in the small pores in such soils, since this water is adsorbed and quasi-solid and may require high shearing stresses in order to move (Jiménez & Justo 1971). In any case, the importance of this adsorbed water decreases as the water content of the soil increases, as a result of which the threshold would be lower for samples of lower dry density, the saturation of which is reached at higher water contents. Yong *et al.* (1986) find this threshold in their tests performed with mixtures of expansive clay and crushed granite, and explain it in terms of the existence of active surfaces that, in the presence of water, would cause interaction between particles, with which the flow would be restricted for hydraulic gradients below the apparent threshold. For larger hydraulic gradients, they observe a linear relation between flow and hydraulic gradient (Figure 41).

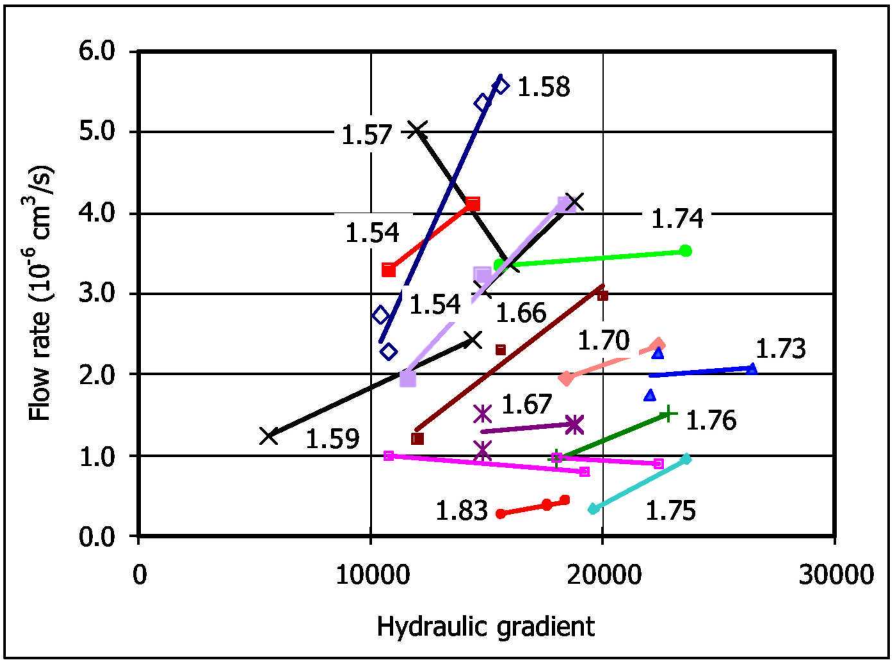

Figure 40: Variation in flow rate obtained for different FEBEX samples of high density (indicated in g/cm^3) versus the hydraulic gradient (points joined by lines correspond to the same sample) (Villar *et al.* 2002)

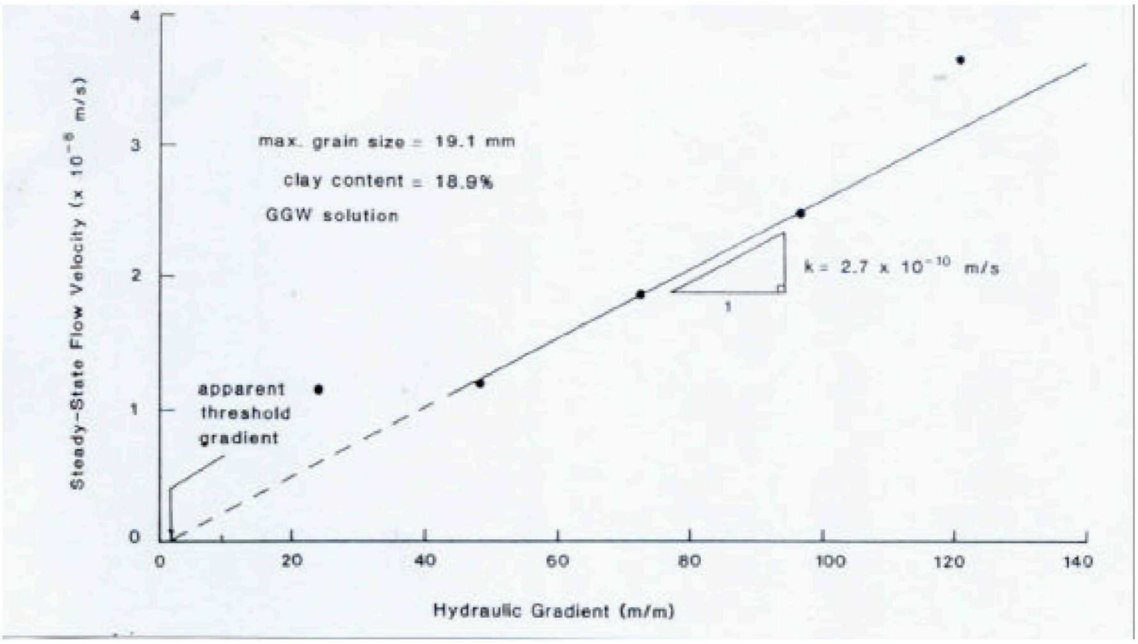

Figure 41: Flow velocity/hydraulic gradient relationship for a crushed granite/montmorillonite mixture (Yong *et al.* 1986)

2.3.1.4 *Influence of the temperature on saturated and unsaturated permeability*

The temperature effect on hydraulic conductivity of compacted bentonites or additive/bentonite mixtures has been studied by several authors, who generally observed an increase in permeability with temperature, mostly attributed to the thermal decrease of the kinematic viscosity. However, the changes of hydraulic conductivity with temperature can be above or below those expected on the basis of the change in water viscosity, depending on the kind of material and even on the type of cations in the exchange complex (Romero *et al.* 2001b, Villar & Lloret 2004).

Kharaka & Smalley (1976) reported hydraulic conductivity values of a Na bentonite and Mingarro *et al.* (1991) of a granite/Ca bentonite mixture, which increased with temperature (ranging from 20 to 100 °C) roughly according to estimates from the decreases in the kinematic viscosity of water. Likewise, Cho *et al.* (1999) found similar results for a Ca bentonite. On the other hand, Towhata *et al.* (1993) found a higher hydraulic conductivity increase with temperature than that calculated by using the properties of free and pure water in a kaolinite and a bentonite (Figure 42), and the same was found by Villar & Lloret (2004) for a saponite (Figure 43). However, the last authors found that the actual increase of hydraulic conductivity in two Ca bentonites (S-2 and FEBEX) is lower than predicted (Figure 43). Towhata *et al.* (1993) consider that the degeneration of adsorbed water into bulk pure water at elevated temperatures (Derjaguin *et al.* 1986) may result in an increase of the dimension of flow channels and therefore of the intrinsic permeability to liquid phase. This would explain the increase of hydraulic conductivity with temperature higher than predicted solely on the basis of the change of the water properties.

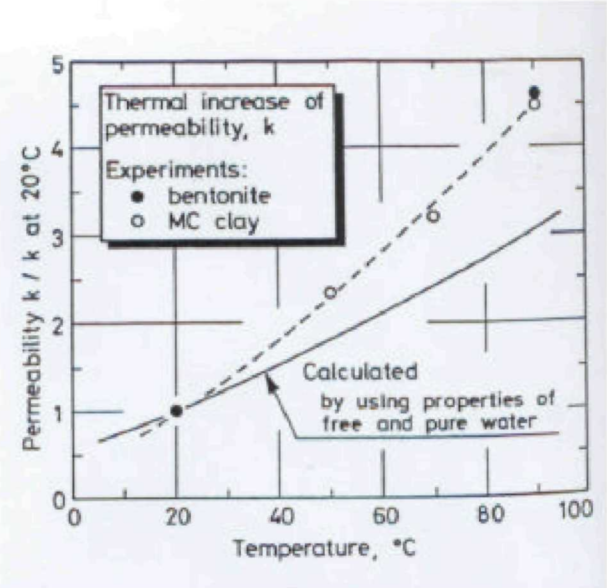

Figure 42: Predicted variation of the permeability upon heating compared with the experimental data (Towhata *et al.* 1993)

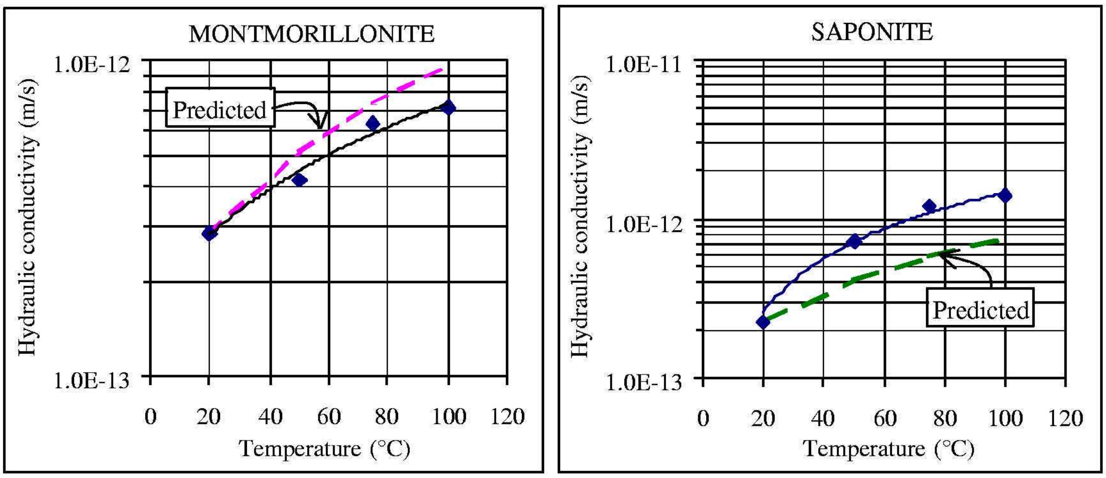

Figure 43: : Hydraulic conductivity of S-2 (montmorillonite) and MCA-C (saponite) bentonites compacted at dry density 1.59 g/cm^3 as a function of temperature: experimental values and predictions according to the water kinematic viscosity change (Villar & Lloret 2004)

2.3.1.5 Influence of the salinity of the permeant on permeability

It is an accepted fact that the type of water used as a permeating agent, and especially its salinity, may have an impact on the coefficient of permeability of a soil. The increase in permeability with the salinity of the permeating fluid has been underlined by various authors (Rolfe & Aylmore 1977) and attributed to: (1) alterations in the distribution of the pore dimension as a result of variations in the swelling pressure in the clay matrix, (2) variations in the mobility of the molecules of water associated with the exchangeable cations adsorbed on the surfaces or forming diffuse double layers (DDL), (3) alterations in the viscous behaviour of the structure of the water. As a result of these mechanisms, as the concentration of the electrolyte increases there is a reduction in the swelling pressure of the clay particles, the size of the flow channels increasing to the detriment of the number of small channels, this causing flow –and therefore, permeability– to increase. According to the DDL theories, the thickness of the DDL decreases as the saline concentration of the water in the pores increases. Consequently, for a given porosity, the effective porosity of the clay would increase with increasing concentration of the solution, with the corresponding increase in permeability. On the contrary, the increasing development of diffuse double layers on reduction of the concentration of the electrolyte causes a decrease in permeability. The reduction of effective porosity with the decreasing salinity of the permeating agent would be due to the fact that the pore space is occupied by the bound water (DDL), the viscosity of which is higher than that of free water.

However, the effect of salinity on the hydraulic conductivity of the compacted saturated bentonite is not very important, while the effect on the unsaturated permeability of less expansive granite/bentonite mixtures is more significant.

Figure 44 shows the values for the FEBEX bentonite permeated with granitic and saline water along with the fittings obtained for deionised water (Figure 32). The granitic water used has a salinity of 0.02 percent, while the saline water –which simulates the composition of the FEBEX pore water– has a salinity of 0.8 percent (see ENRESA 2000 for detailed chemical composition). No clear trend is observed concerning the variation of the values obtained with granitic water with respect to those obtained with deionised water. The values obtained with saline water are, however, 184 percent higher on average than those expected for a sample of the same density tested with deionised water, and in addition, they show greater dispersion. This higher permeability to saline water with respect to that expected for deionised water is more significant for low densities.

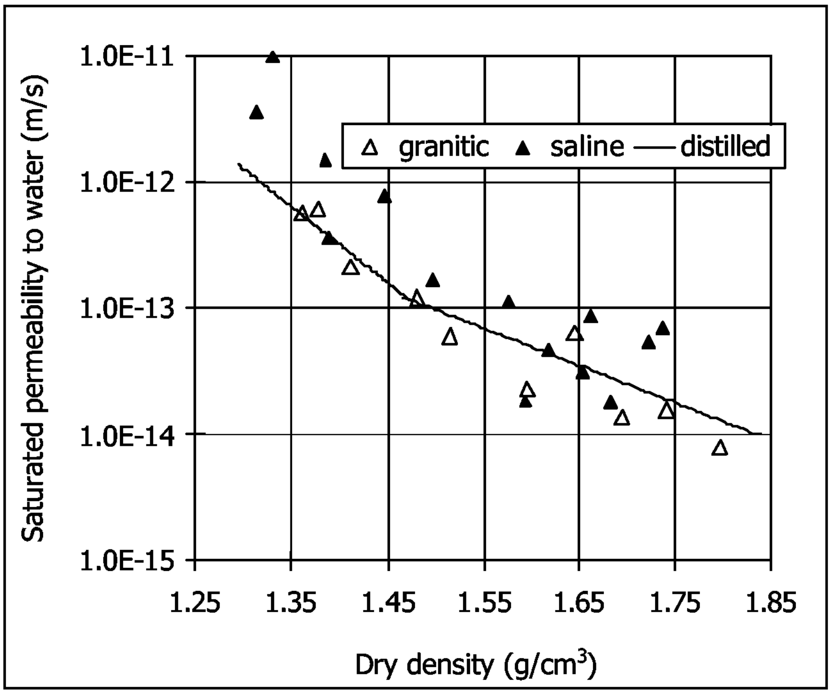

Figure 44: Hydraulic conductivity for granitic and saline water *vs*. dry density of the FEBEX clay and fitting obtained for deionised water (Villar 2002)

The hydraulic conductivity of the MX-80 bentonite has been measured using permeants of different salinities (Karnland *et al.* 1992). The results obtained are plotted in Figure 45 and show an increase of permeability of half an order of magnitude when the salinity of the permeant increases from 0 to 3.5 NaCl percent. In the context of the Prototype project, the same kind of tests has been performed at CIEMAT with the MX-80 bentonite permeated with deionised and saline water. The saline water used in this case was obtained in the laboratory by mixing $CaCl_2$ and NaCl at a relationship 32/68 percent to give rise to a solution of 0.5 percent salinity. The results obtained are plotted in Figure 46, in which the exponential correlations fitted between dry density and hydraulic conductivity are also shown. The hydraulic conductivity is 135 percent higher for saline water than for deionised water (Villar *et al.* 2003).

However, the hydraulic conductivity of less expandable materials (such as Friedland Ton clay, with 45 % of smectite) is more affected by the changes in the salinity of the permeant, increasing up to 2 orders of magnitude when salinity rises from 0 to 20 % (Figure 47, Pusch 2001). The figure also shows that the kind of electrolyte in the pore water has an influence on permeability too. Thus, sodium gives place to a higher increase of permeability than calcium.

On the other hand, Mata & Ledesma (2003), in the context of the Backfill & Plug Test project, calculated the hydraulic conductivity of a 70/30 weight percent Äspö granite / MX-80 bentonite mixture (Figure 48). The tests were carried out with permeants of different salinity and the results made clear that the chemistry of the pore fluid strongly influences the backfill hydraulic behaviour. The maximum difference of hydraulic conductivity was greater than 6 times when the salinity of the water used to saturate the specimens was increased up to 16 g/L. The high sensitivity of the backfill materials (mixtures) to high electrolyte contents of percolates is explained by Pusch (2002) as due to the fact that the density of the clay fillings in the voids between aggregate grains is rather low, even if the average density of the mixture is high.

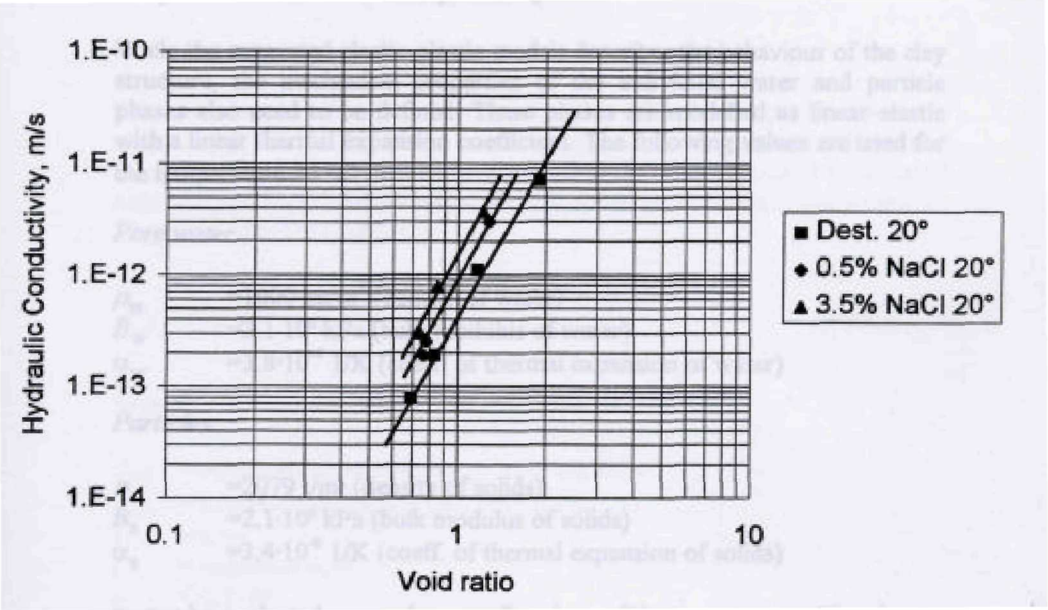

Figure 45: Hydraulic conductivity of MX-80 as a function of the void ratio at different salt concentration in the pore water (Karnland *et al.* 1992)

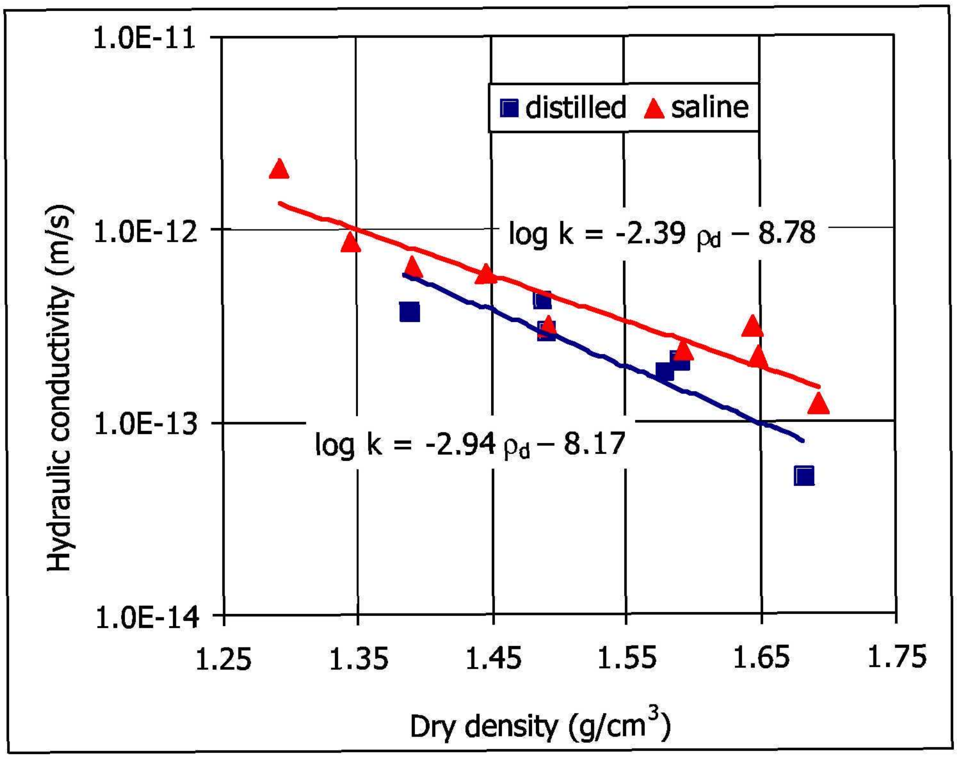

Figure 46: Results of the hydraulic conductivity tests with compacted MX-80 (Villar *et al.* 2003)

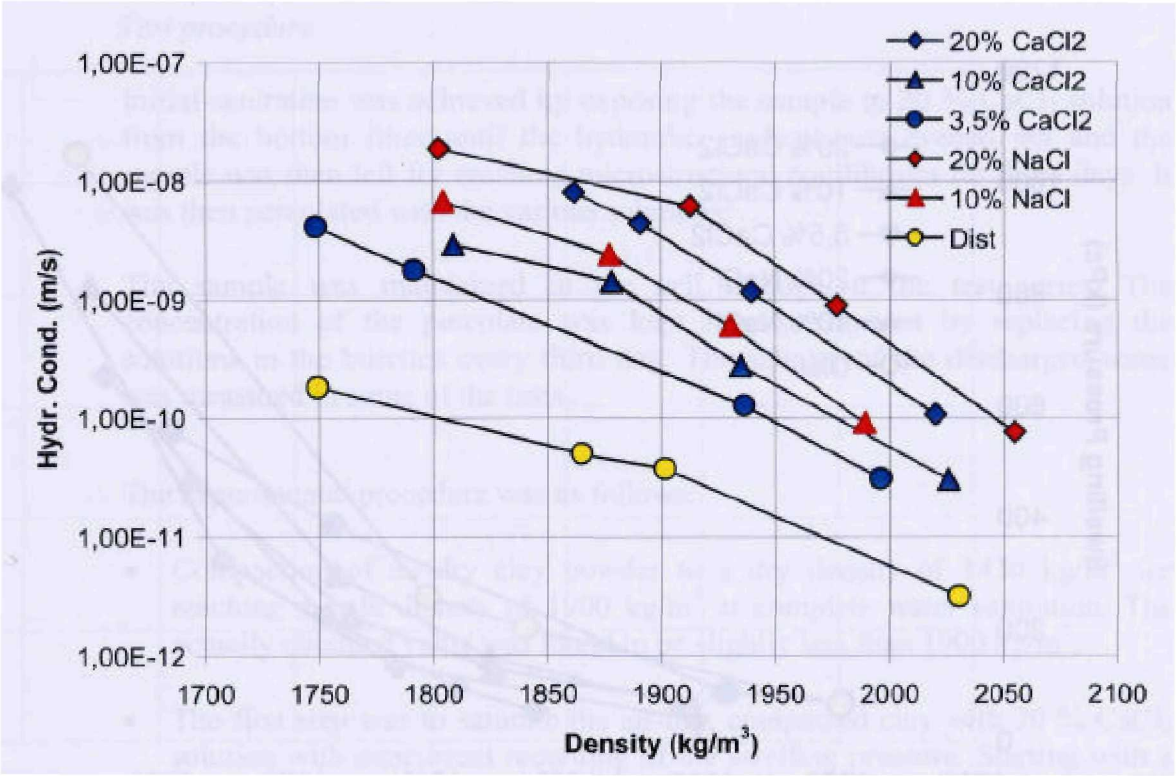

Figure 47: Hydraulic conductivity of saturated samples of Friedland Ton clay *vs*. bulk density as a function of the electrolyte concentration (Pusch 2001)

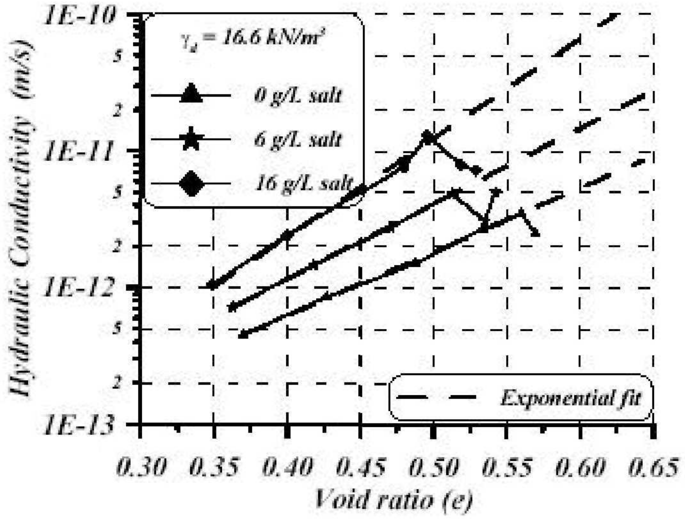

Figure 48:Estimated hydraulic conductivity of a granite/Na bentonite (70/30 weight percent) mixture (Mata & Ledesma 2003)

2.3.2 Water retention

The water retention curve (WRC; or soil water characteristic curve, SWCC) describes the evolution of the water content undergone by a sample when subjected to gradually increasing or decreasing suctions, *i.e.* to drying or hydration paths. It is a fundamental relation to estimate the evolution of the water content in the barrier and the prediction of the time needed for full saturation, since it determines the negative capillary pressure of the bentonite, which is the main driving force for water uptake in the initial stages of the repository.

The great avidity for water and high water retention capacity of bentonites, in other words, their high suction value, condition the behaviour of such materials, and consequently the techniques to be used for their study. The magnitude of the suction of the smectites when removed from the saturated state is such that it cannot be measured directly using conventional methods, such as tensiometers or psychrometers. When the aim is to study their unsaturated behaviour, it is necessary to fall back on the alternative of imposing suction instead of measuring it, that is to say, of subjecting the sample to a given and known suction that conditions its water content. Three methods are specially widely used: axis translation, control of relative humidity and the osmotic technique (Fredlund & Rahardjo 1993).

As discussed by several authors, the retention capacity of swelling materials is highly affected by the strain state, and it is higher for samples that can swell freely (unconfined samples) than for those kept at constant volume (Yahia-Aissa 1999, Romero & Vaunat 2000, Villar 2002). The comparison between curves determined under both conditions is shown in Figure 49 for the FEBEX bentonite (Villar & Lloret, 2004) and in Figure 50 for the FoCa clay (Yahia-Aissa *et al.* 2001), where the above-mentioned aspect can be observed. On the other hand, in the retention curves at constant volume determined for the FEBEX clay, at the beginning of hydration, the water content of the samples with a higher dry density is slightly greater for the same values of suction, but when lower values of suction are reached, the water content of the denser samples is lower. The repercussion of the initial dry density on the value of the water content reached is more obvious as suction decreases (Figure 51). The hysteretic behaviour of the clay has also become evident, the water contents reached during drying being higher than those obtained during the previous wetting (in confined samples) and the water contents in the wetting paths lower than those reached for the same suction in the previous drying (in unconfined samples).

Consequently, the retention curve of bentonites should be determined both in confined and unconfined samples, although the sealing material will be unconfined only in the outer part of the barrier, where a void exists between the compacted blocks and the host rock. The determination of the SWCC in unconfined samples is justified because, being much easier to determine, it will still be useful to identify the influence of other factors such as temperature or salinity. On the other hand, in drying paths it is not possible to maintain the volume of the sample constant, as it was seen in Figure 51.

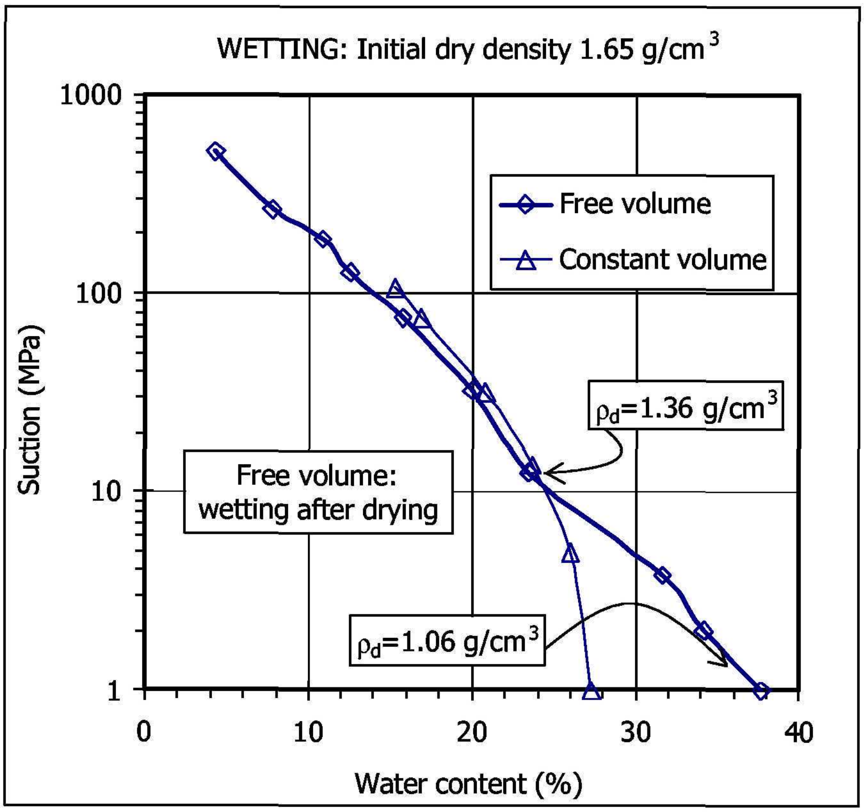

Figure 49: Comparison of the retention curves obtained for the FEBEX bentonite in confined and unconfined volume conditions (modified from Villar & Lloret 2004)

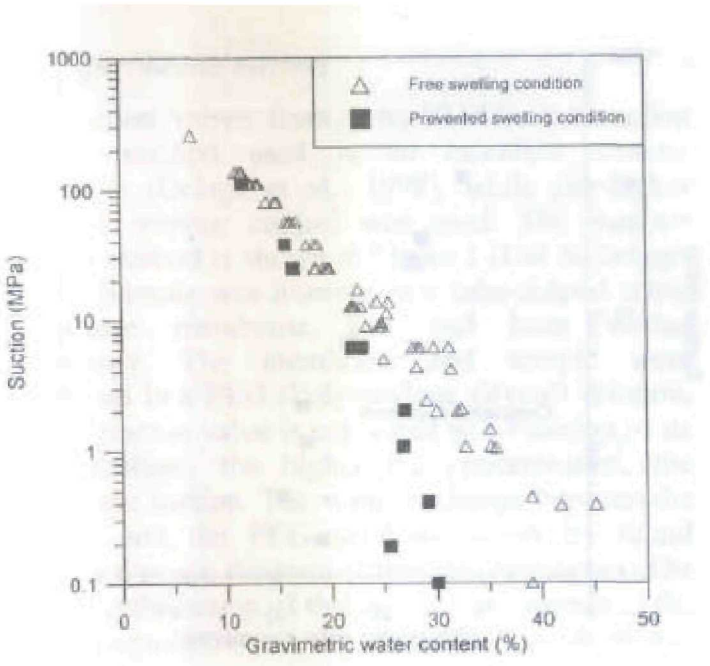

Figure 50: Suction / water content relation for compacted specimens of confined and unconfined FoCa clay (Yahia-Aissa *et al.* 2001)

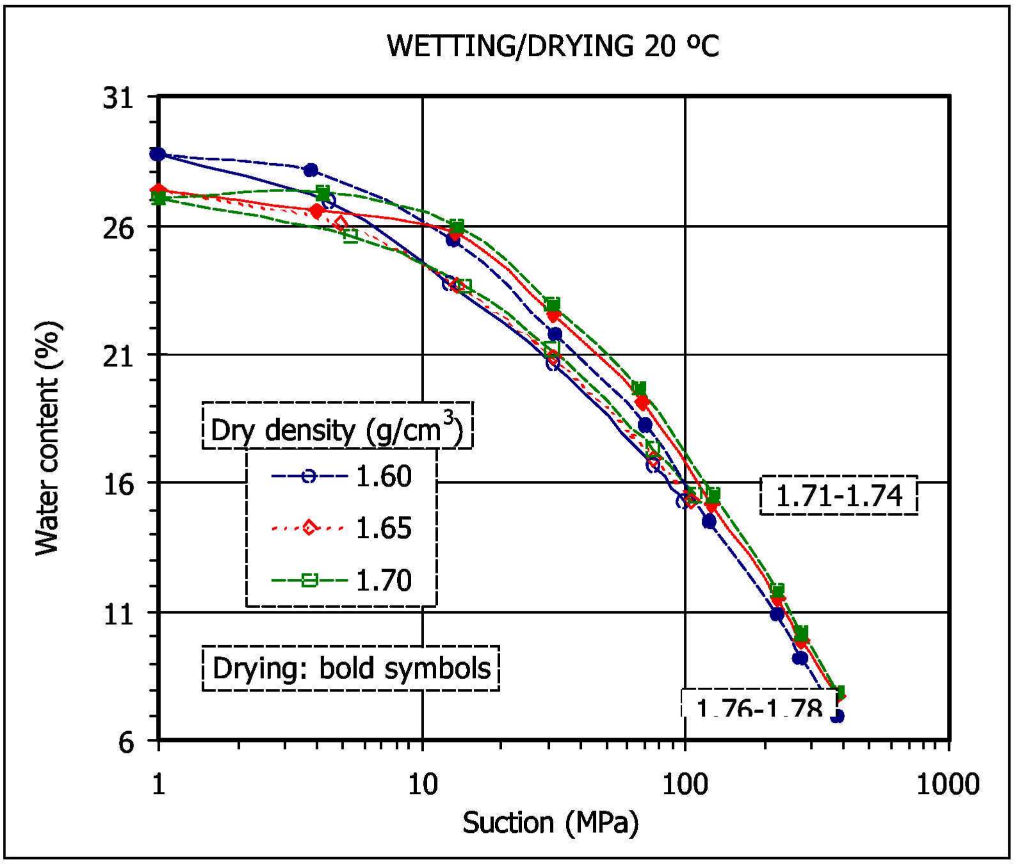

Figure 51: Retention curves at constant volume in a wetting/drying path for the FEBEX bentonite compacted at different dry densities. The dry densities at the end of drying are indicated in g/cm^3 (Lloret *et al.* 2004)

The tests performed with FEBEX bentonite have shown that the higher the temperature, the lower the retention capacity for the same dry density, especially for the lowest suctions (Lloret *et al.* 2004, Villar & Lloret, 2004): at a constant degree of saturation the higher the temperature of the sample the lower its suction, both in confined (Figure 52) and unconfined conditions. However, it seems that the effect of temperature on the retention capacity is different depending on the kind of material and the type of exchangeable cation of the bentonite. In fact, results obtained by CIEMAT for the MX-80 bentonite show the inverse tendency (Figure 53). However, there is not much information on this issue and the results available should be considered preliminary.

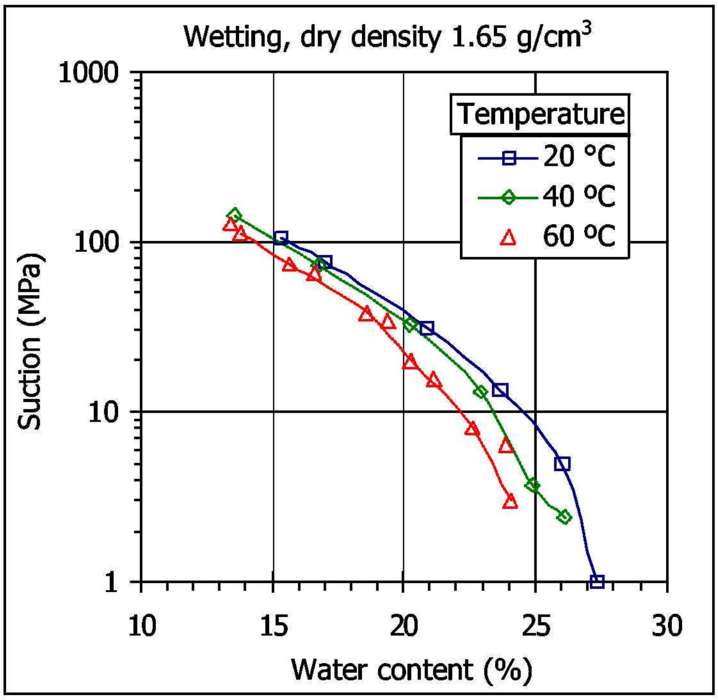

Figure 52: Retention curves at constant volume for different temperatures determined for FEBEX clay (Lloret *et al.* 2004)

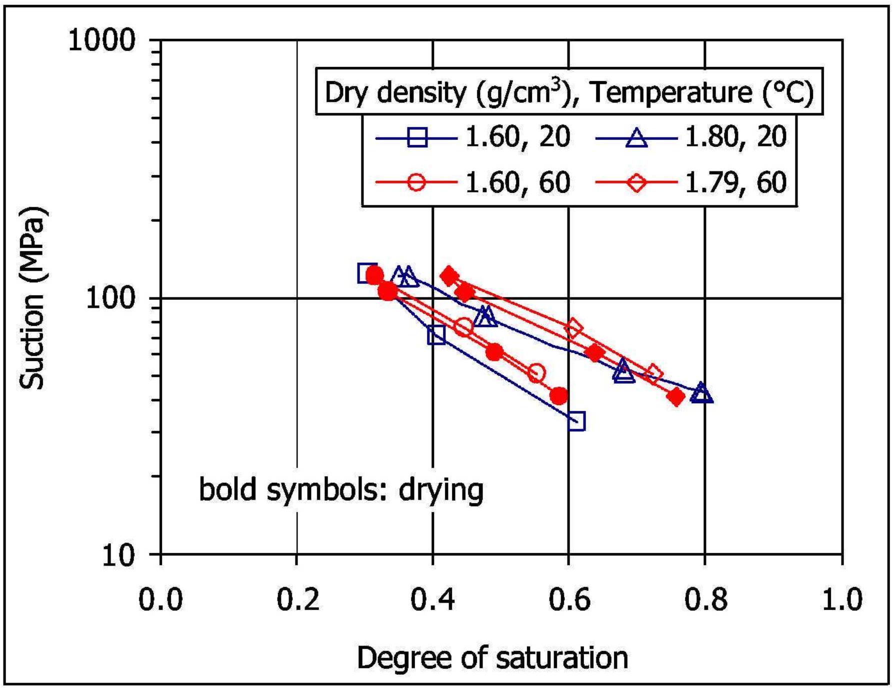

Figure 53: Retention curves for MX-80 clay at 20 and 60 °C (ENRESA's contribution to Äspö HRL report 2003, *in press*)

With respect to the influence of salinity on the retention capacity, the available information is still scarcer. Engelhardt (2003) presents data obtained for a crushed granite / Na bentonite mixture (70/30 wt %) using pure and saline water. The latter comes from the Äspö HRL. The results show that the air entry pressure is 39 percent lower when saline water is used (Figure

54). Besides, the results show that the influence of salinity on hydraulic parameters is more important than that of temperature.

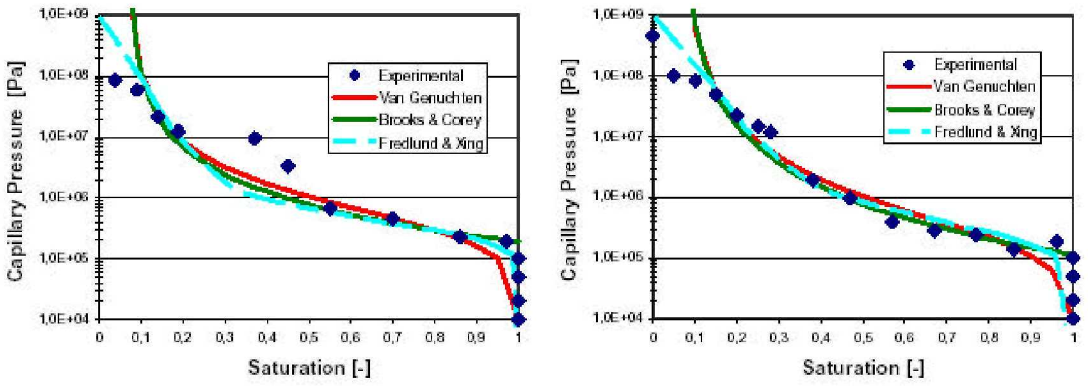

Figure 54: Measured capillary pressure curve of a Na bentonite/crushed granite mixture (30/70 wt %) compacted to dry density 1.6 g/cm^3 obtained using pure (left) and Äspö saline water (right) (Engelhardt 2003)

3 MODIFICATION OF THE BENTONITE DURING THE TRANSIENT STATE OF A REPOSITORY

The performance of a disposal facility is influenced by the changes occurring in the mechanical, physicochemical and geochemical properties of the engineered barriers and the host rock. These changes are generated by the combined effects of the heat produced by disintegration of the wastes, water movements and the geochemical composition of the near field. The design criteria and construction procedures used for the engineered barriers also influence the performance of a disposal facility.

The information about the modifications that occurred on the bentonite as a consequence of the interaction between thermal and hydraulic flows comes, apart from theoretical considerations (ENRESA *et al.* 2002), from the experience gained from laboratory and *in situ* tests. The laboratory tests basically consist in subjecting blocks of compacted clay, confined in hermetically sealed and non-deformable cells, to simultaneous heating and/or hydration on opposing fronts. After different periods of time –from days up to years– the bentonite is analysed (Villar *et al.* 1996, Cuevas *et al.* 2002, ENRESA 2000, Villar *et al.* in press). Among the *in situ* tests, several of them –performed according to different disposal concepts– have been dismantled and analysed (Villar & Lloret 2003):

- The pioneer test for simulation of the repository conditions in a granitic host rock was the Buffer Mass Test (BMT), carried out in the Swedish Stripa mine. The test consisted of several boreholes measuring some 3 m in depth and 0.76 m in diameter, housing electrical heaters surrounded by blocks of compacted, unsaturated sodium bentonite (MX-80). After different periods of testing of up to 39 months, the physical and chemical properties of the material that had been subjected to thermo-hydraulic gradients were determined in the laboratory (Pusch *et al.* 1985).

- More recently, two large-scale sealing experiments conducted at the Atomic Energy of Canada Limited's Underground Research Laboratory at Lac du Bonnet (Canada) were dismantled: the Buffer Container Experiment (BCE) and the Isothermal Test (ITT) (Dixon *et al.* 2002). Both were performed according to the Canadian deep geological repository concept, in which the buffer material –consisting of a 50:50 (by dry weight) mixture of quartz sand and sodium-rich bentonite– is placed in 5-m depth by 1.24-m diameter boreholes. The BCE had a central heater and was in operation for approximately 2.5 years. The ITT, performed under isothermal conditions, was monitored for 6.5 years before decommissioning.

- At present, in the underground laboratory of Äspö (Sweden), the "Long term tests of buffer material" (LOT) is being conducted. It is an *in situ* test based on the Swedish disposal concept but at a smaller scale, in which compacted blocks of sodic bentonite (MX-80) are placed in vertical boreholes (4-m depth, 0.3-m diameter). The foreseen duration of these tests is 1, 5 and 20 years, and the 1-year ones have already been dismantled and analysed (Karnland *et al.* 2000).

- The FEBEX is a project for the study of the near field for a HLW repository in crystalline rock according to the Spanish concept (ENRESA 1995, 2000), in which two heaters are placed horizontally in a drift of 2.3 m diameter at the Grimsel Test Site (GTS, Switzerland) and surrounded by a clay barrier constructed of highly compacted bentonite blocks (FEBEX bentonite). After five years of operation, heater 1 was switched off, the bentonite barrier was dismantled and the heater extracted, the rest of the test remaining in operation (Bárcena *et al.* 2003).

It must be taken into account that the results available up to now refer only to periods of thermo-hydraulic treatment smaller than five years and that the effect of radiation has not been evaluated in the *in situ* tests.

3.1 Physical properties

During the construction phase of the disposal facility, the galleries and shafts and all the other spaces will be filled with air at atmospheric pressure. Besides, since the barrier will be probably manufactured with the clay at its hygroscopic water content, the bentonite will initially have degrees of saturation much lower than 100 percent. Consequently, the largest pores will contain air, while the intra-aggregate pores and interlaminar spaces will be hydrated. This confers the bentonite a very high degree of suction, a characteristic that fundamentally conditions the performance of the barrier. This high initial suction promotes and conditions the saturation of the blocks, which will take water from the surrounding geological medium via the largest pores, the latter acting as capillaries from which the water will be distributed to smaller pores, *i.e.* in accordance with a mechanism of double porosity flow.

However, the low permeability of the bentonite, which is even lower when not saturated, will make the saturation process very gradual. The movement of water from the surrounding rock to the clay barrier may be facilitated to some extent by the increased permeability of the gallery walls, due to microfissures generated during the excavation. The tests performed in Stripa underground laboratory (Pusch *et al.* 1985), and the *in situ* test carried out at the Grimsel underground laboratory for the FEBEX Project (ENRESA 2000), have shown that the transport of water from the rock to the bentonite is highly uniform, and have also shown that the increase in the water content of the bentonite occurs in a spatially homogeneous and radial fashion, as it can be seen in Figure 55 and Figure 56. Besides, the water distribution is not affected by the existence of fractures or other hydraulically more active zones, due to the fact that the permeability of the rock is high compared to that of the bentonite, as a result of which the water flow is determined by the hydraulic properties of the latter (Villar *et al.* 2004). However, in tests in which a more permeable buffer has been used, a more irregular pattern of water distribution has been found. This is the case of the BCE test performed in Canada with a 50/50 weight percent mixture of Na bentonite and sand (Figure 57).

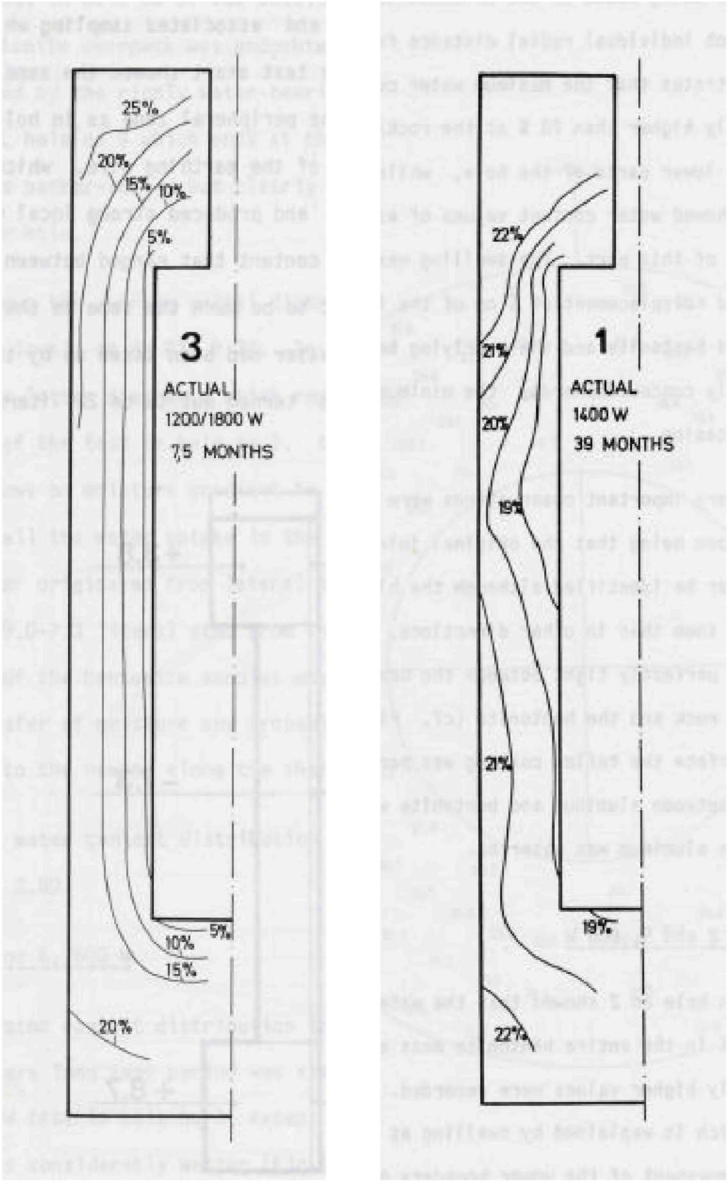

Figure 55: Moisture distribution in the bentonite from the heater holes of the Stripa Project after 7.5 and 39 months of experiment (Pusch *et al.* 1985)

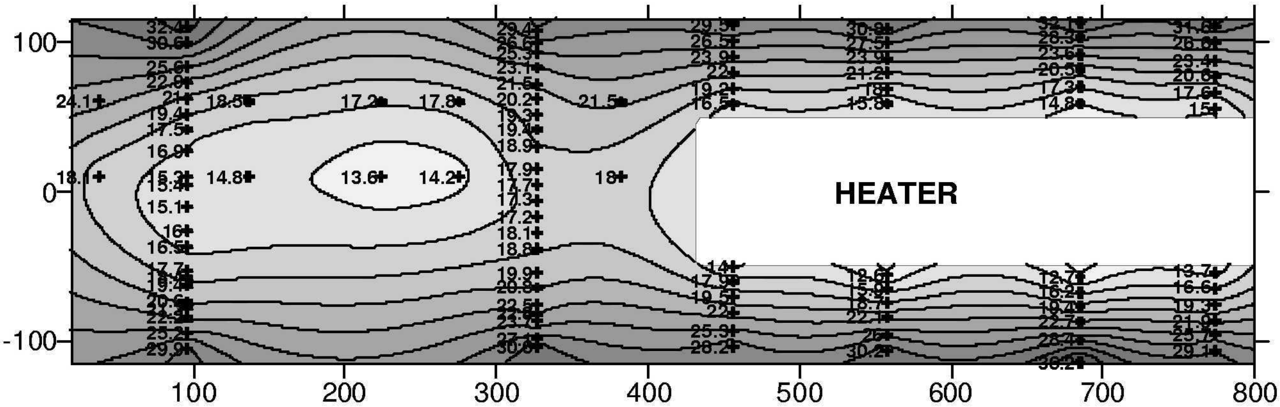

Figure 56: Longitudinal distribution of the water content (%) along the first section of the FEBEX gallery at GTS after five years of operation (Villar *et al.* 2004)

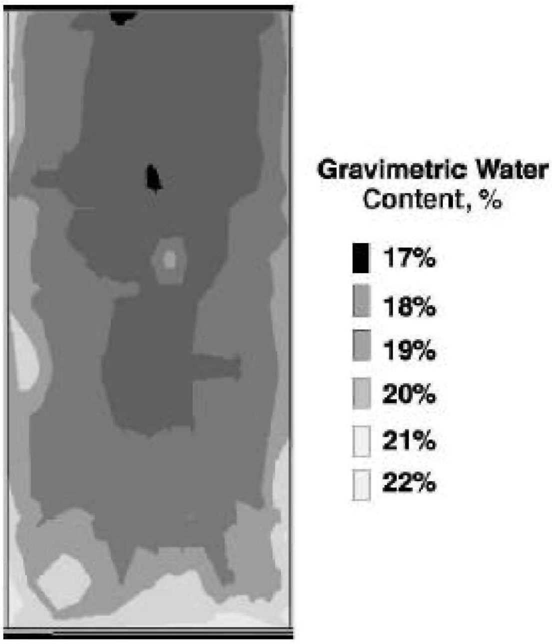

Figure 57: Moisture profile vertically through the isothermal test (ITT) buffer at the time of decommissioning (Dixon *et al.* 2002)

During the transient phase of the disposal facility, as the material in the clay barrier becomes hydrated, expansion occurs. In the areas closest to the surface of the gallery there will be a gap between the blocks and the host rock, due to the difficulties involved in ensuring a perfect fit between both. This gap, and the joints between the different blocks, may partially absorb the increase in the volume of the bentonite, as a result of which its density will decrease in these areas, compared to the manufacturing density of the blocks. In this way, the swelling and high level of suction of the bentonite prevent the joints between the blocks to become preferential channels for the flow of water, since water coming into contact with the bentonite will be immediately absorbed, causing the latter to expand and to seal any type of joint. Likewise, the gap between the bentonite and the wall of the drift will be closed by this same process. In fact, it has been shown that the high density compacted bentonite may penetrate the joints and the fractures in the rock. In the case of the BMT at Stripa –in which a Na bentonite was used as buffer (MX-80)– a clay gel formed between the rock and the bentonite, yielding a tight clay/rock contact (Pusch *et al.* 1985). Also, the blocks become sealed after saturation: in the FEBEX experiment the bentonite sections presented a consistent appearence upon exposure after five years of operation; although the joints between the blocks were clearly visible, all the construction gaps were sealed, even the big apertures hewn in the bentonite for the passing of the cable bunches (Figure 58).

Figure 58: Appearence of the bentonite barrier after dismantling of the first bentonite sections and extraction of the heater in the FEBEX experiment (Bárcena *et al.* 2003)

However, this reduction in the density of the bentonite will be local, since the overall barrier is confined, thus, when hydration occurs swelling pressures will be developed and, therefore, there will be an increase in the radial stresses in the barrier. During this transient state, these stresses may cause the internal parts of the barrier, which have not yet been reached by the water, to compress, thus increasing their density. In this way, a density gradient is established in the system, with lower densities in the areas affected by water, where the bentonite will have expanded (Figure 59). The inverse relationship established in the barrier between dry density and water content can be seen in Figure 60 from the BCE (Dixon *et al.* 2002). These density modifications affect increasingly larger areas as hydration progresses and are enhanced by the effect of heating (Villar 2002). The same kind of coupled density and water content gradients have been observed in infiltration laboratory tests performed with compacted MX-80 bentonite and tracked by γ-ray attenuation measurements, a nondestructive method (Lemaire *et al.* 2004).

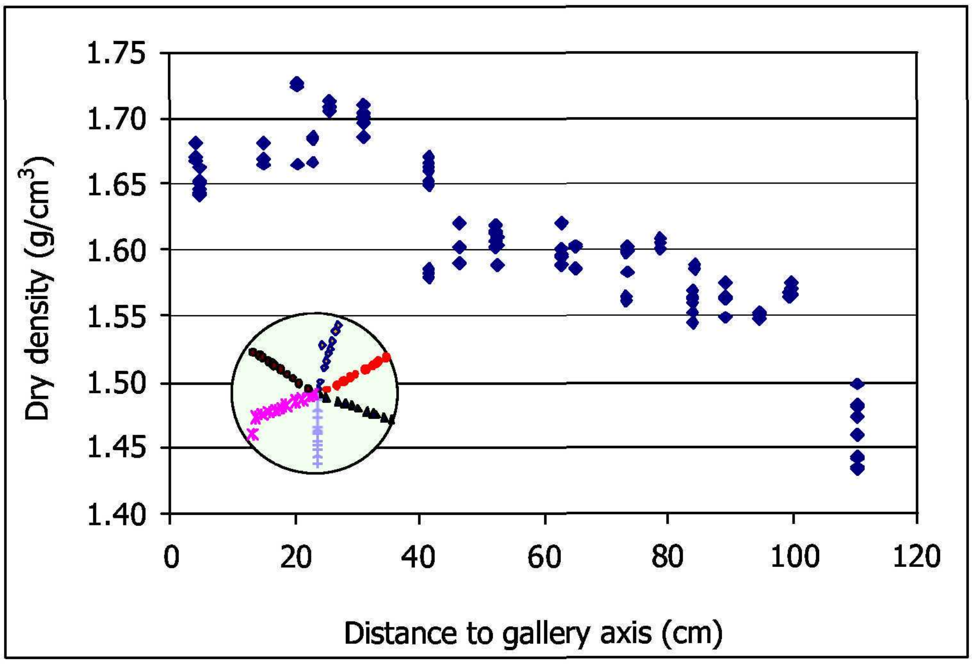

Figure 59: Dry density measurements along different radii in a vertical section of the bentonite without the heater from the FEBEX gallery at GTS. The initial ρ_d of the blocks was 1.70 g/cm^3 (Villar *et al.* 2004)

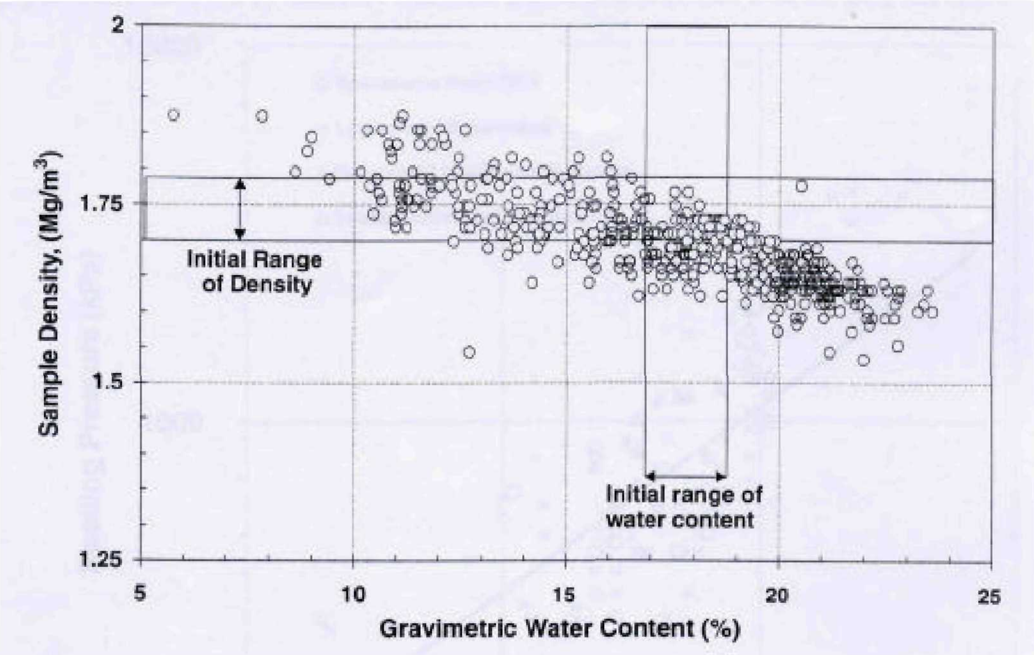

Figure 60: Dry density and water content of samples from the BCE at the time of decommissioning (Dixon *et al.* 2002)

On the other hand, in the internal part of the barrier the heat generated by the waste canister causes an increase in temperature that is transmitted outwards by conduction, convection and radiation, generating a thermal gradient. The evaporation of the water, induced by the high temperatures close to the canister, causes desiccation of the bentonite and its shrinkage, this contributing to the decrease of the porosity of the blocks in the internal areas of the barrier, as can be seen in Figure 61 from the FEBEX *in situ* experiment (Daucausse & Lloret 2003, Villar *et al.* 2004). Furthermore, the thermal expansion of the bentonite is insufficient to compensate for this decrease in porosity (UPC 1998). However, the drying of the bentonite close to the canister due to increasing temperature should not cause any significant reduction in volume –as long as water content does not decrease below 4-6 percent in the case of the FEBEX bentonite–, since above the value of suction corresponding to hygroscopic water content the sample becomes very stiff (Villar 2002). This will prevent the formation of shrinkage cracks and preferential flow channels from becoming important.

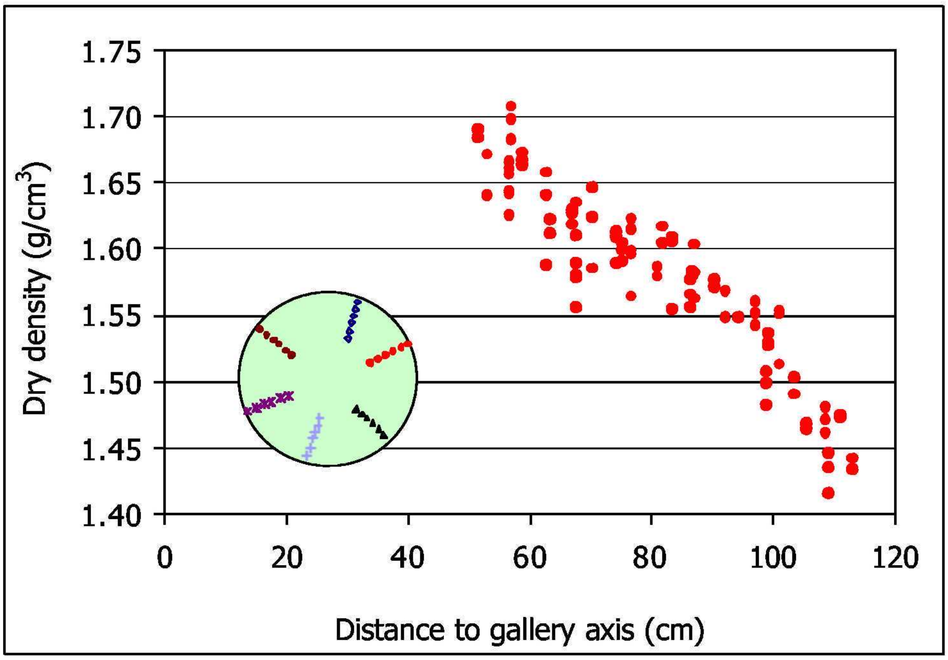

Figure 61: Dry density measurements along different radii in a vertical section of the bentonite with heater from the FEBEX gallery at GTS. The initial ρ_d of the blocks was 1.70 g/cm^3 (Villar *et al.* 2004)

The vapour generated spreads towards more external and colder regions of the system, where it condenses. This may give rise to local increases in the degree of saturation. The water that condenses in the colder areas may move once more towards the canister, due to a flow process caused by the suction gradients (Pusch *et al.* 1985), but it is now charged with dissolved mineral species (those originally possessed by the clay and those provided by the advective flow from outwards), which precipitate close to the canister when the water evaporates. In this way the process of evaporation/condensation is repeated and small "convection cells" are established, the most evident effect of which is an increase in salinity in the hotter areas (Villar *et al.* 1997). This vapour movement is, therefore, an important mechanism for the transfer of water content and heat.

Finally, once saturation is reached in the whole barrier, there could be a recovery and a certain homogenisation of the void ratio, although this aspect has not been possible to check yet. The controlled suction oedometric tests (see section 'Effect of the suction on volume change", Villar 2002, Lloret *et al.* 2003) have shown that during the hydration of the compacted bentonite irreversible strains may occur, these being larger as the overload is lower. In the barrier, and due to its confinement, the overload of the bentonite will be important in all cases, therefore irreversible strains are not expected to occur during hydration, except at the joints between the blocks and at the periphery, where the space between the bentonite and the rock may be sufficient to allow for free expansion of the clay. As a result, there would be an outer ring in which the bentonite would have a lower dry density –which would no longer be recovered– and a higher water content. This ring might constitute a source of water supply and would ensure its homogeneous distribution across the entire surface of the barrier (Villar 2002).

The dismantling of the first section of the FEBEX *in situ* test at the GTS has shown that the bentonite has experienced an overall drop in dry density, which implies a decrease of dry density from that of the compacted blocks, 1.70 g/cm^3, to an average density of the barrier of 1.58 g/cm^3. This is explained by the filling of all the construction gaps. This overall increase of volume is linked to a redistribution of pore sizes of the bentonite and an increase of the average pore size, which are not related to the position of the samples in the barrier. There is an increment of the percentage of large pores and a relative decrease of the percentage of smaller pores. However, the higher percentage of porosity –as in the case of the untreated blocks– corresponds to the size of less than 0.006 µm (Villar *et al.* 2004).

3.2 THM properties

As seen in the previous section, the thermal, hydraulic and mechanical aspects of the system interact significantly. Thus, variations in the degree of saturation affect both temperatures –as a result of the dependence of thermal conductivity on water content– and the distribution of stresses –due to the changes in suction linked to changes in water content–. Likewise, hydration leads to important changes in the volume and rigidity of the bentonite. The changes in porosity caused by stress variations affect the hydraulic conductivity and, therefore, the movement of water. In addition, the thermally induced strains lead to variations in stresses and to changes in hydraulic conductivity. The diffusion of vapour –favoured by the thermal gradients– and the dependence of water viscosity on temperature –with its consequences on hydraulic conductivity– also condition the water transport phenomena. The effects of the strains on the temperature may also have some importance, although the variation of thermal conductivity with porosity is not large.

The following sections review the knowledge about the modification of the THM properties of barrier materials after being subjected to repository conditions.

3.2.1 Hydraulic behaviour

The bentonite installed in the disposal facility as high density blocks (or pellets) manufactured from clay with its hygroscopic water content has a high value of suction, and consequently a high degree of water avidity. This will be the fundamental driving force for the saturation of the barrier with the water from the surrounding massif. The high suction generated by drying may, with increasing hydraulic gradients, speed up the arrival of the water to the internal areas of the barrier. However, as the clay saturates, its intrinsic permeability will decrease (see Figure 36), since the size of the flow channels becomes smaller. This process is accompanied by a decrease in the suction gradient responsible for the flow of water, as a result of which the saturation process will become steadily slower. This reduction in flow will be compensated only in part by the increase in relative permeability occurring with increasing saturation

(Villar 2002). Furthermore, the pressure of the water in the saturated pores of the host rock will be hydrostatic, and its magnitude will depend on the depth of the disposal facility, possibly being of the order of 5 MPa. The pressure gradient established in the disposal facility also forces water to enter the barrier, although this does not have a great influence until the suction of the bentonite becomes small, towards the end of the process. At this moment, if a hydraulic threshold exists (Figure 41), the full saturation of the barrier could be very much delayed, although this aspect has not been confirmed. On the other hand, the extent of the modifications in the void ratio will influence the hydraulic properties of the bentonite buffer, which are important for water, gas and radionuclide transport.

With respect to the bentonite water retention capacity, the samples from the Grimsel *in situ* test have shown a behaviour similar to that of the samples of untreated FEBEX clay compacted to the same dry density and subjected to similar suctions. This has been confirmed by measuring the suction of the blocks from Grimsel in the laboratory. These measurements have been related to the water contents determined in the same blocks and are plotted in Figure 62 as a retention curve. The curve determined for untreated FEBEX bentonite compacted at dry density 1.60 g/cm^3 is also plotted in the figure (Figure 51). This curve was obtained following a wetting path, which is the same process experienced by the samples from Grimsel. Overall, the points obtained in the blocks from Grimsel closely follow the curve for the untreated samples, what suggests that the retention capacity of the FEBEX clay has not appreciably changed after five years under repository conditions (Villar *et al.* 2004).

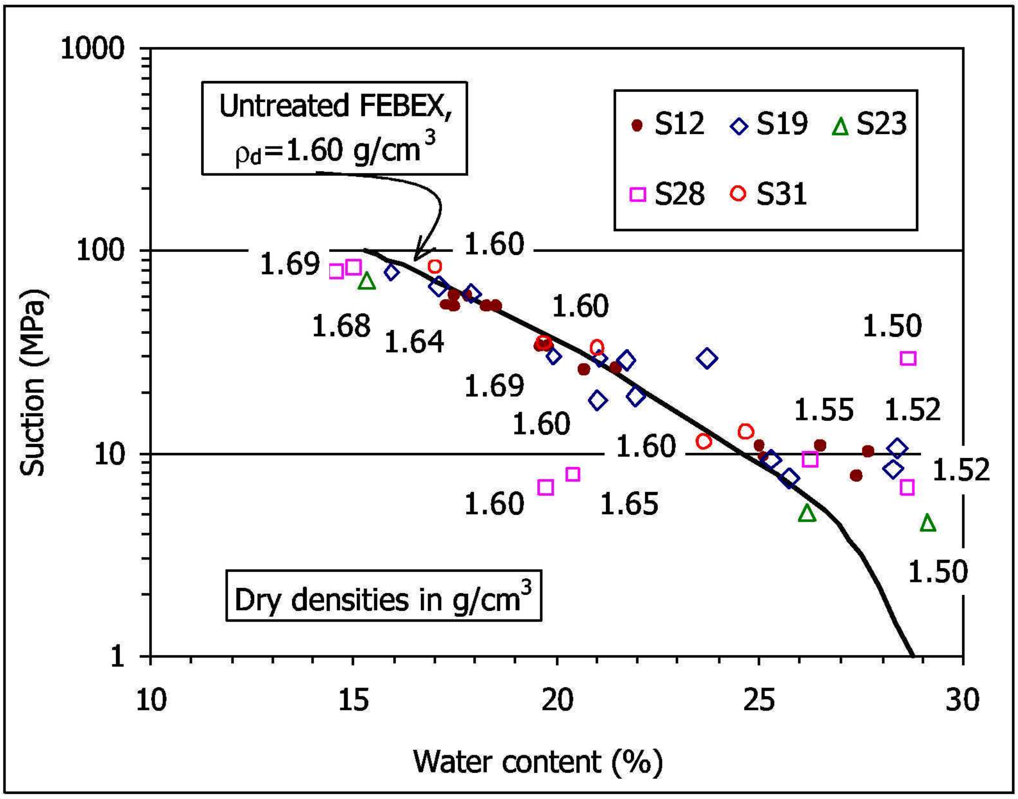

Figure 62: Suction values measured with capacitive sensors at the laboratory in blocks taken from different sections of the FEBEX *in situ* test at GTS (points) and retention curve determined in untreated FEBEX samples by control of relative humidity at constant volume (line) (Villar *et al.* 2004)

On the other hand, the air initially contained in the bentonite pores will become compressed as hydration advances, and the studies performed suggest that this air will be completely dissolved in the ingressing water, which will allow for the complete saturation of the barrier (Pusch *et al.* 1985).

Once the barrier is saturated, and assuming that the dry density of the assembly is around 1.60 g/cm^3, its permeability will be lower than 10^{-12} m/s, depending on the buffer considered and, to a lesser extent, the salinity of groundwater and other geochemical considerations (the effect of temperature will be probably negligible at this stage of the repository). The hydraulic conductivity measurements performed in samples retrieved from *in situ* tests have not shown any important variation of this property of the bentonite after being under repository conditions. Thus, Dixon *et al.* (2002) confirm that no discernible changes in the hydraulic conductivity properties of the buffer had occurred as a result of the 2.5 (BCE) or 6.5 (ITT, isothermal test) years of installation (Figure 63). In samples of Na bentonite coming from the LOT test, the hydraulic conductivity measured was lower compared to results obtained in untreated samples at high density, about the same at medium density, and higher at low density (Karnland *et al.* 2000). These authors attribute the slight modifications observed to the effect of salt on low-density material. Furthermore, the hydraulic conductivity of the samples of the FEBEX bentonite taken from Grimsel is clearly related to the dry density and the latter in turn is related to the position of the sample in the barrier (Figure 64). The values of the hydraulic conductivity measured for the samples of lower density (more hydrated) are in the order of the theoretical ones, but for samples of higher densities there is a large dispersion in the values obtained without any clear tendency (Villar *et al.* 2004). More analysis should be necessary to clarify this issue.

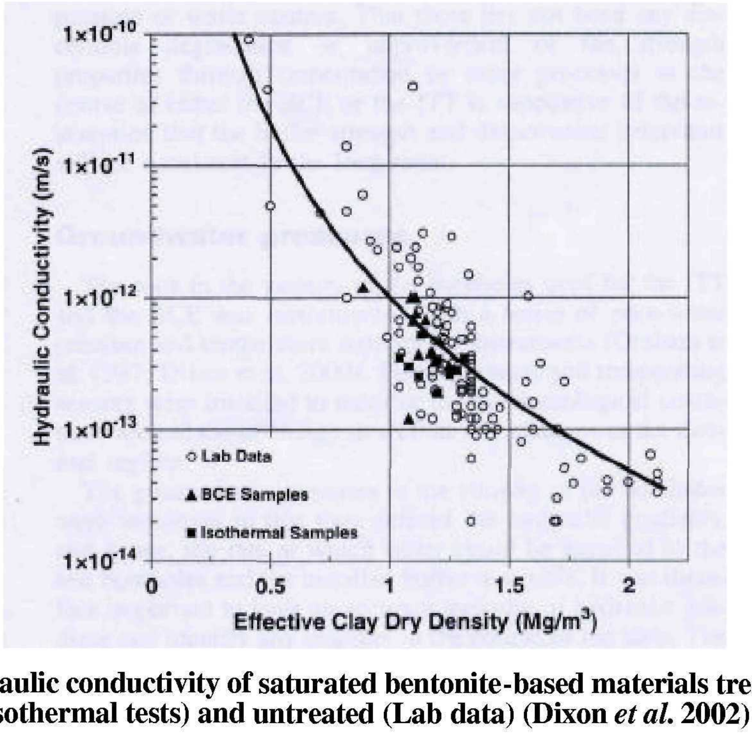

Figure 63: Hydraulic conductivity of saturated bentonite-based materials treated (BCE and Isothermal tests) and untreated (Lab data) (Dixon *et al.* 2002)

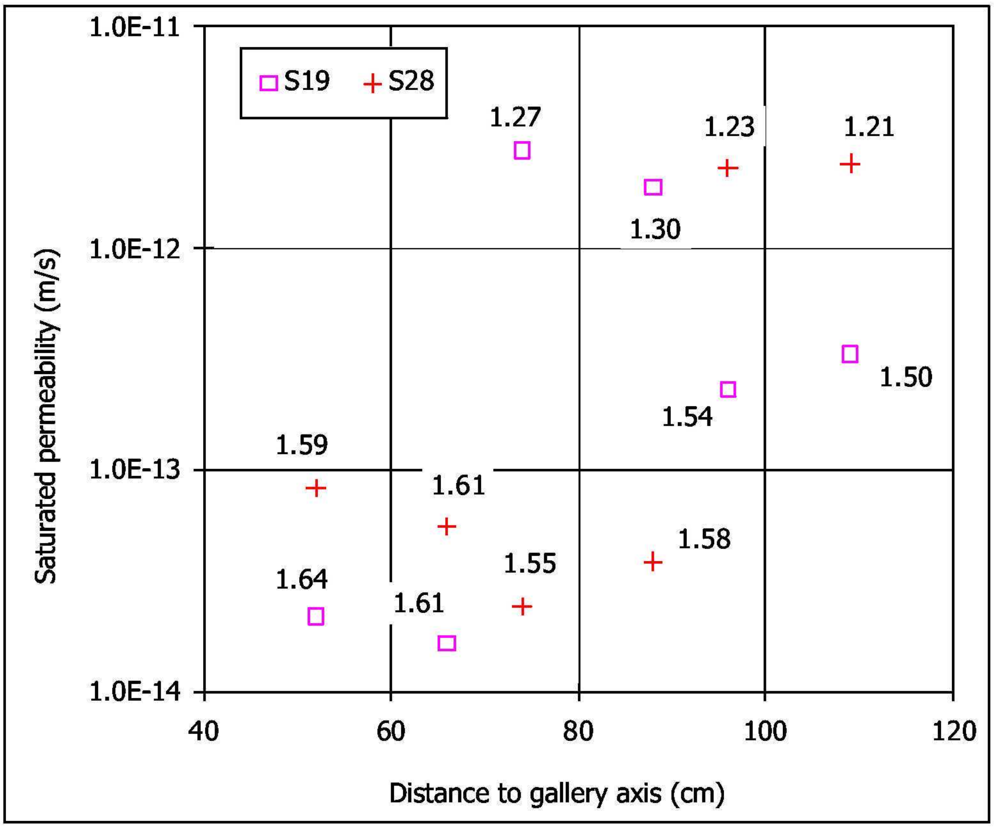

Figure 64: Hydraulic conductivity of samples of the FEBEX bentonite taken after the dismantling of heater 1 at the GTS (the dry density of the samples is indicated in g/cm^3) (Villar *et al.* 2004)

Long after the closure of the disposal facility, with the barrier now completely saturated and with the canister having degraded –these phenomena expected to occur after some thousand years– there might be an important generation of gases in the inner part of the barrier. The most important processes that would give rise to the generation of gas would be the corrosion of the steel canister, the bacterial degradation of organic matter and the radiolytic decomposition of water. Diffusion is the dominating mechanism of gas transport during the initial stages of saturation of the barrier, when the only available air is the one initially contained in the pores of the bentonite. In the event of very large volumes of gases, its evacuation through diffusion in the water might be difficult. In this case, the gas might escape via instantaneous "fractures" in the barrier, a process known as break-through that basically consists in transport via preferential paths that dilate and propagate (Volckaert *et al.* 1995, Rodwell *et al.* 1999).

3.2.2 Mechanical behaviour

Simultaneously with hydration, expansion of the bentonite will occur in those areas in which there is space to allow for it. In those areas in which the bentonite is confined, the swelling pressure exercised by the clay will increase with saturation (Figure 27), what may cause compression of the drier areas and an increase in their density, enhanced by the shrinkage due to water evaporation in hot areas.

Drying to water contents of 4-6 percent does not reduce irreversibly the swelling capacity of the bentonite (Villar 2002), as a result of which the hydro-mechanical properties of the material should not be altered following the transient period, and with the arrival of the water the bentonite will expand and the canister will be confined. The models of microstructural behaviour show that the swelling experienced by a soil on decreasing suction under a given load is higher the higher the suction at the beginning of hydration, and that the relation between the percentage of strain and the decrease in suction increases on reaching lower values of suction, both aspects have been confirmed in tests performed with the FEBEX bentonite (Villar 2002). The results plotted in Figure 65 show that in the samples subjected to previous drying there is a very sharp increase of void ratio when suction decreases below the initial value (120 MPa). It must be pointed out that these results have been obtained by drying the bentonite through reduction of the relative humidity at a temperature of 20 °C, and not by drying at high temperature.

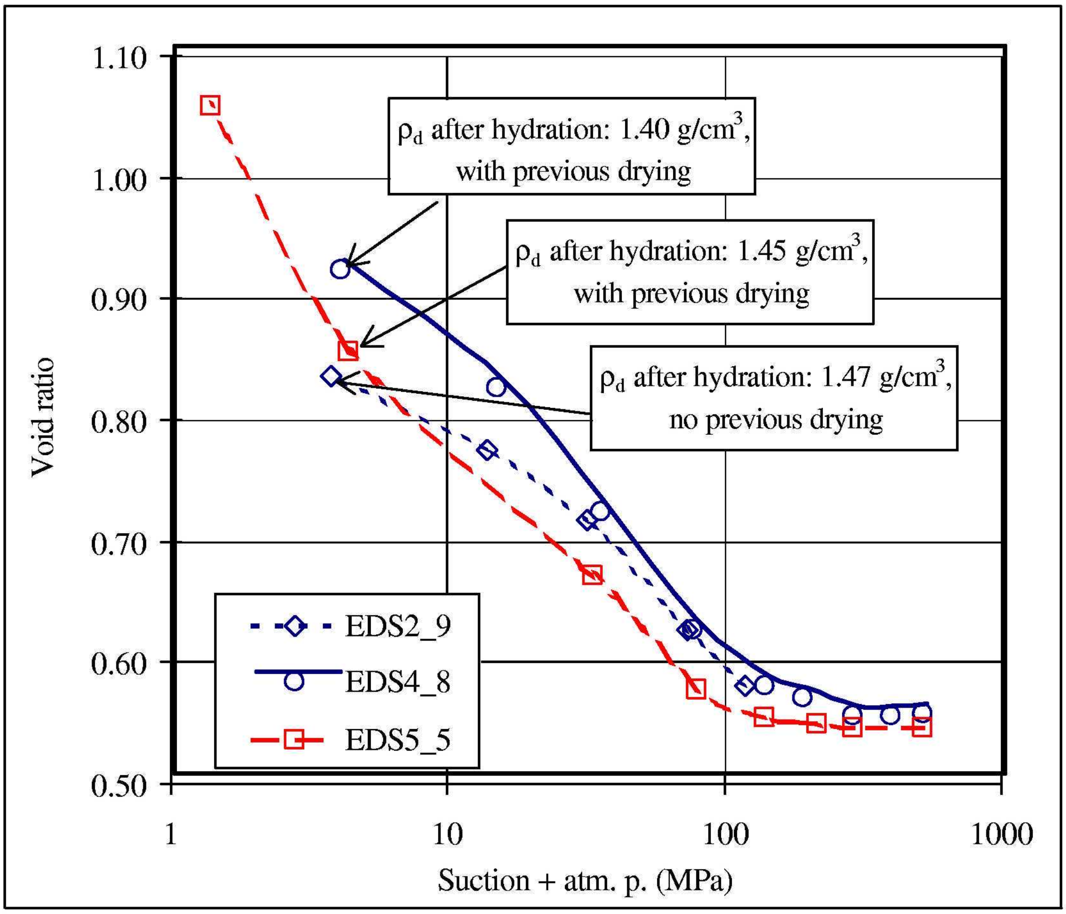

Figure 65: Comparison of the evolution of the void ratio of the FEBEX bentonite during hydration under a load of 0.1 MPa in three oedometric tests, two of which had previously included drying to a suction of 520 MPa (Villar 2002)

Furthermore, the swelling tests performed with bentonite retrieved from *in situ* tests have shown that the swelling capacity of the clay is not irreversibly affected. Pusch (1985) concluded that the swelling pressures recorded in samples taken very close to the heater in the BMT (Stripa, Sweden) were at least as high as those obtained for non-heated MX-80 bentonite saturated with salt water. The measurements of the swelling pressure presented by

Dixon *et al.* (2002) obtained in samples from the BCE and the ITT tests performed in Canada agree well with those obtained in untreated samples of the same dry density (Figure 66). The same has been found in MX-80 samples collected from the 1-year test of the LOT experiment, although for the lowest bentonite densities the swelling pressure was lower than the previously measured values (Karnland *et al.* 2000). This –like the hydraulic conductivity increase– is explained by the authors as a consequence of the effect of saline test solutions on low density material. The swelling capacity of the samples retrieved from the FEBEX *in situ* test at Grimsel –determined as deformation upon saturation– has not changed irreversibly after five years of being subjected to repository conditions, the values obtained for the treated samples being similar to those expected for untreated samples of the same initial dry density and water content (Figure 67).

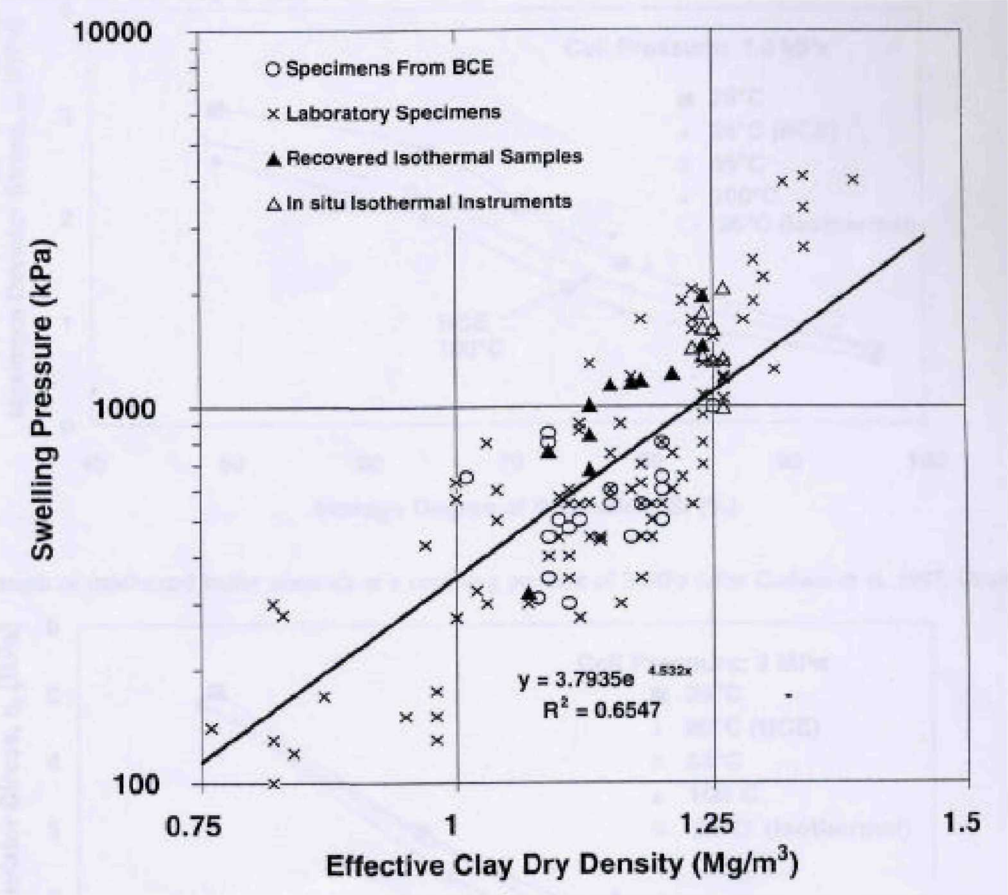

Figure 66: Swelling pressures developed by laboratory specimens (untreated) and samples recovered from the BCE and ITT (Dixon *et al.* 2002)

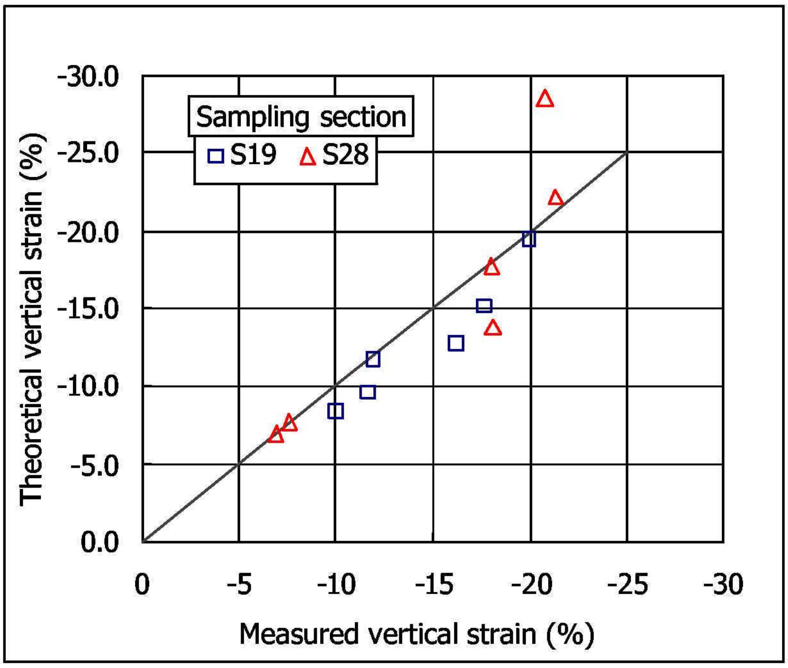

Figure 67: Final strain reached in soaking tests with deionised water performed in bentonite samples from Grimsel under a vertical pressure of 0.5 MPa compared to the theoretical value for untreated FEBEX bentonite (Villar *et al.* 2004)

Triaxial tests performed in samples recovered from the BCE and ITT tests (Dixon *et al.* 2002) indicate that there has been neither a discernible change in the stress-strain properties nor any degradation or improvement of the strength properties of the materials that have undergone thermally induced desiccation or isothermal water uptake. The shear strength and shear course of MX-80 samples recovered from the 1-year LOT test had not varied with respect to the untreated clay, according to the results of the triaxial tests presented by Karnland *et al.* 2000. The same authors present beam tests results that show that the treated clay has become slightly softer or more plastic, which is considered as an effect of heating. Additionally, no increase in tensile strength or decrease in strain at failure were observed.

A preconsolidation pressure lower than 10 MPa has been measured for all the samples from the FEBEX *in situ* test at Grimsel. This is an important decrease with respect to the initial preconsolidation pressure of the bentonite, since the compaction pressure applied to manufacture the blocks was around 40 MPa. It is considered that the microstructural changes associated with the volume increase experienced during hydration account for this decrease in the apparent preconsolidation stress. Furthermore, the preconsolidation stresses found are lower the higher the water content reached during the *in situ* experiment. These values are plotted as a function of suction in Figure 68, in which the apparent preconsolidation stresses found for untreated FEBEX bentonite subjected to different suctions are also shown (Villar 2002, Lloret *et al.* 2003).

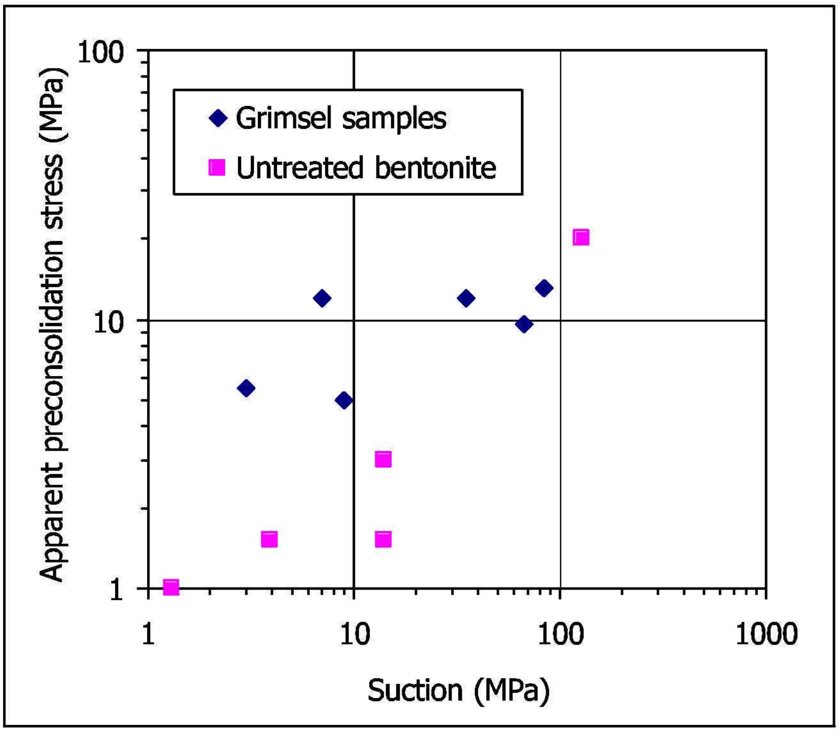

Figure 68: Apparent preconsolidation stress of samples from Grimsel and of untreated bentonite (Villar *et al.* 2004)

3.2.3 Temperature effects

The thermal conductivity increases with the water content of the clay and, consequently, during the transient stage it should be higher for the external part of the buffer and lower in the desiccated area near the canister. This has been verified in samples retrieved from the FEBEX *in situ* test (Figure 69). Nevertheless, the thermal conductivity of the unsaturated buffer is still sufficient to favour the dissipation of the heat, as has been demonstrated in several *in situ* tests.

On the other hand, it is expected that full saturation of the buffer be reached before the dissipation of the thermal gradient. However, it still remains unclear whether the high temperatures around the canister would hinder the full saturation of the inner part of the barrier or just delay it, but this seems to closely depend on the actual temperatures reached in the barrier and on its thickness. In most repository concepts it is expected that the temperature in the buffer be less than 100 °C. In the 1-year test performed at Äspö for the LOT Project, the bentonite barrier –10-cm thick– had reached almost full saturation, the temperatures during the test being below 90 °C. On the contrary, at the BCE test performed at Lac du Bonnet (Canada), the areas of the buffer (whose thickness was 25 cm) adjacent to the heater had a water content below the initial one after 2.5 years of heating. In the FEBEX *in situ* test, the bentonite closer to the heater had water contents below the initial ones after five years of heating (the surface temperature of the heater was of 100 °C), although they were recovering after the intense initial drying (Figure 70). The thickness of the bentonite barrier in this case was of 65 cm.

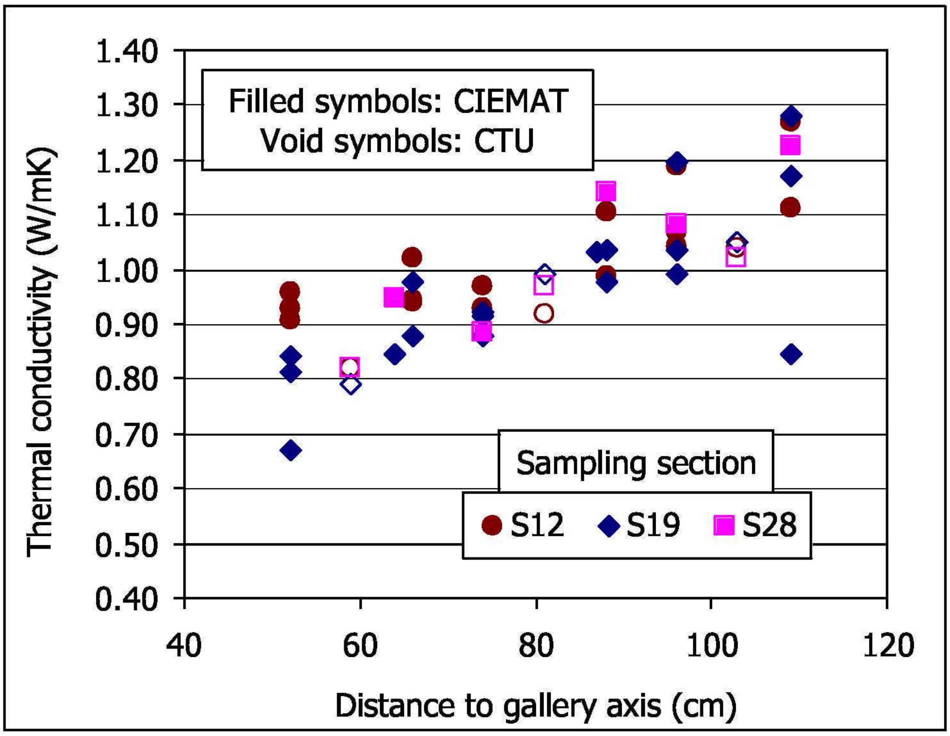

Figure 69: Thermal conductivity measured in samples from the FEBEX test at Grimsel by two laboratories (Villar *et al.* 2004)

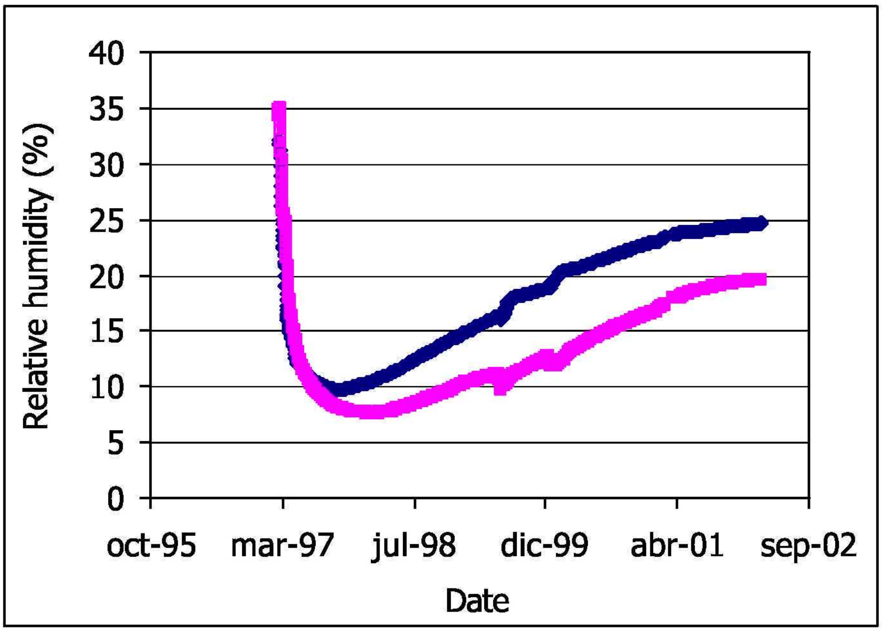

Figure 70: Evolution of the relative humidity of the bentonite registered by two capacitive sensors located near the heater in the instrumented section E1 of the FEBEX *in situ* test (AITEMIN data base)

The effect of a thermal gradient on the kinetics of hydration is being studied in laboratory tests in which a 40-cm long column of compacted FEBEX bentonite is hydrated under thermal gradient (test GT40) and at isothermal conditions (test I40). The evolution of the temperature and the relative humidity is shown in Figure 71 for the test performed under thermal gradient and in Figure 72 for the test performed at isothermal conditions. By comparing both figures, a faster initial hydration in the test performed under thermal gradient seems to occur, since the increase in the humidity registered by the first sensor (RH1) is higher in test GT40 and the sensor placed further (RH2) starts to perceive the humidity increase much earlier in the case of infiltration under thermal gradient than in the case of infiltration at laboratory temperature. In the first case, the humidity recorded by sensor RH2 could come in the form of water vapour from the lower part of the column. Otherwise, the increase of hydraulic conductivity with temperature would account for this initial quicker hydration of the test under thermal gradient. However this behaviour is reversed as saturation proceeds and at the present moment, after 19000 hours (more than 2 years) of hydration, the average relative humidity recorded by the three sensors in test GT40 is 68 percent and in test I40 is 82 percent.

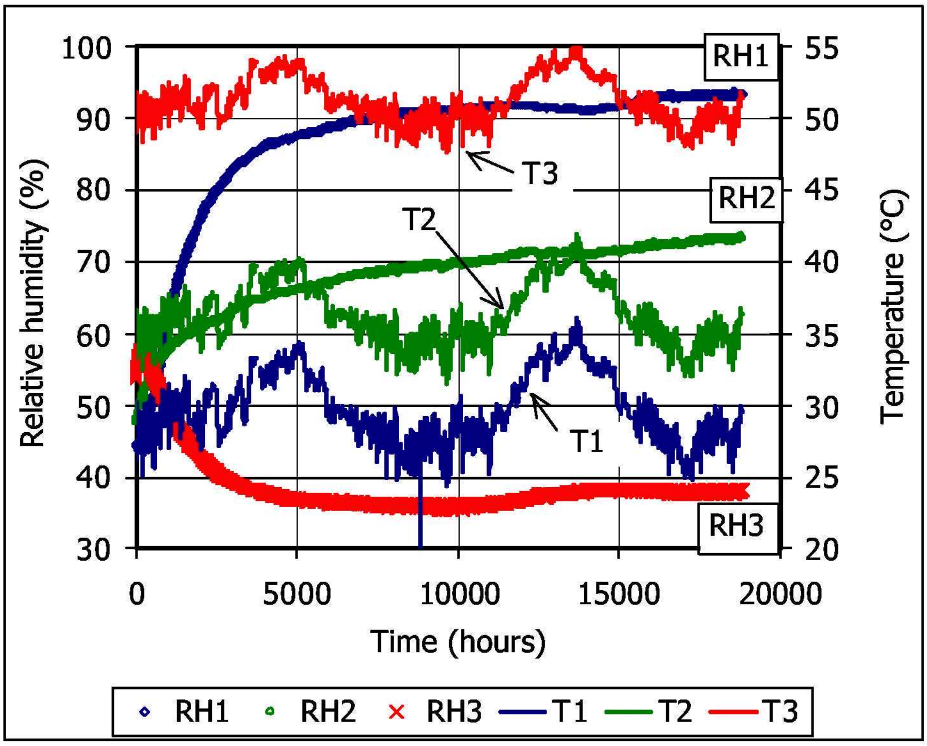

Figure 71: Evolution of temperature and relative humidity in the test performed under thermal gradient (GT40) during infiltration (sensor 1 placed at 30 cm from the bottom, 2 at 20 cm and 3 at 10 cm) (Lloret *et al.* 2004)

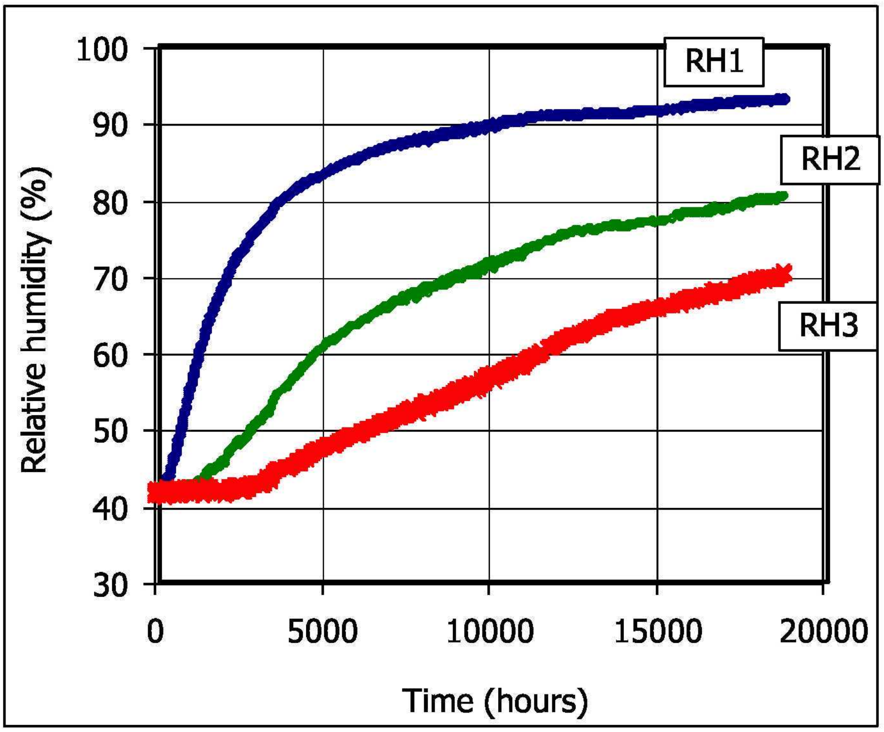

Figure 72: : Evolution of relative humidity in the test performed under isothermal conditions (I40) during infiltration (sensor 1 placed at 30 cm from the bottom, 2 at 20 cm and 3 at 10 cm) (Lloret *et al.* 2004)

The effect of the temperature on the behaviour of bentonite would be noticeable only in the first stages of the repository, specially during the transient phase. In any case, the impact of the temperature on the properties analysed does not appear to be drastic, as we have seen in the previous sections.

3.3 Open questions

With respect to the barrier saturation there are two key questions to understand and model the time to reach full saturation and the evolution in the final stages:

- The repercussion of the thermal gradient on saturation, specially for high temperatures. Experimental results confirm that saturation is delayed when a thermal gradient exists, specially if the temperatures reached in the barrier are higher than 100 °C. It remains unclear if full saturation would be hindered up to dissipation of the thermal gradient.

- The possible existence of a hydraulic gradient threshold, which would prevent the movement of water in the final stages of the repository, when suction is very low and the only driving force is the hydraulic pressure.

On the other hand, it has not been possible to experimentally check if the density gradients observed in the bentonite barrier are recovered when the final water content of the barrier is homogeneous and if the volume changes induced during saturation are irreversible.

There is not enough experimental evidence regarding the modification of hydro-mechanical properties of bentonite by the effect of temperature. In turn, it is unknown if the possible modifications induced by temperature on the bentonite are reversible or not.

There is uncertainty on the volume of gas and corrosion products generated. Some available data suggest that the volume of the corrosion products could be huge, the repercussion on the stress state of the barrier being of major importance. The same could be said if the gas generation rate were large.

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Thermo-Hydro-Mechanical Characteristics and Processes in the Clay Barrier of a High Level Radioactive Waste Repository. State of the Art Report

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Thermo-Hydro-Mechanical Characteristics and Processes in the Clay Barrier of a High Level Radioactive Waste Repository

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79 pp. 72 figs. 102 refs.

Abstract:

This document is a summary of the available information on the thermo-hydro-mechanical properties of the bentonite barrier of a high-level radioactive waste repository and of the processses taking place in it during the successive repository operation phases. Mainly the thermal properties, the volume change processes (swelling and consolidation), the permeability and the water retention capacity are analysed. A review is made of the existing experimental knowledge on the modification of these properties by the effect of temperature, water salinity, humidity and density of the bentonite, and their foreseen evolution as a consequence of the processes expected in the repository. The compiled evolution refers mostly to the FEBEX (Spain), the MX-80 (USA) and the FoCa (France) bentonites, considered as reference barrier materials in several European disposal concepts.

Características y Procesos Termo-Hidro-Mecánicos de la Barrera de Arcilla de un Almacenamiento de Residuos Radiactivos de Alta Actividad

Villar, M.V.

79 pp. 72 figs. 102 refs.

Resumen:

Este documento es un resumen de la información existente en la actualidad sobre propiedades termo-hidro-mecánicas de la barrera de bentonita de un almacenamiento de residuos radiactivos de alta actividad y de los procesos que tienen lugar en ella durante las direrentes fases de operación del almacenamiento. Se analizan especialmente las propiedades térmicas, los procesos de cambio de volumen (hinchamiento y consolidación), la permeabilidad y la capacidad de retención. Se revisa el conocimiento experimental existente sobre la modificación de estas propiedades por efecto de la temperatura, salinidad del agua, humedad y densidad de la bentonita, y su evolución previsible como consecuencia de los procesos que se esperan en el almacenamiento. La información recopilada se refiere fundamentalmente a las bentonitas FEBEX (España), MX-80 (Estados Unidos) y FoCa (Francia), consideradas materiales de barrera de referencia en varios conceptos europeos de almacenamiento.

CLASIFICACIÓN DOE Y DESCRIPTORES

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HIGH-LEVEL RADIOACTIVE WASTES; THERMODYNAMIC PROPERTIES; MECHANICAL PROPERTIES; CLAYS; GEOCHEMISTRY; RADIOACTIVE WASTE STORAGE; MATHEMATICAL MODELS; COMPUTER CODES