Experimental study of the effect of high porewater salinity on the physical properties of a natural smectitic clay

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SKB

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Abstract

Natural smectitic clays for backfilling tunnels and shafts in deep repositories may be an alternative to mixtures of bentonite and ballast. Very salt groundwater is known to raise the hydraulic conductivity and reduce the expandability of clay materials in general and of bentonite/ballast mixtures in particular and the present study aimed at determining the impact of salt water on the major physical properties of natural smectitic clays, represented by the German Friedland Ton. The investigation showed that the compactability of the investigated clay is not significantly affected by the water content in contrast to bentonite/ballast fills, and that the conductivity and expandability are acceptable even at salt contents of up to 20 % if the bulk density at saturation is slightly higher than 2000 kg/m^3. For salt contents up to 3.5 % the corresponding density is around 1900 kg/m^3. In general, the investigated clay offers better physical properties than mixtures of bentonite/ballast mixtures with up to 30 % bentonite.

Summary

Earlier investigations have indicated that natural smectitic clay may represent an attractive alternative to mixtures of bentonite and ballast for backfilling of tunnels and shafts in deep repositories both from performance and economic points of view. Very salt groundwater is known to raise the hydraulic conductivity and reduce the expandability of clay materials in general and of bentonite/ballast mixtures in particular and the present study aimed at determining the impact of salt water on the major physical properties of natural smectitic clays, represented by the German Friedland Ton.

Laboratory compaction of the investigated clay to which different amounts of salt-free water had been added showed that the compactability gives a high density and is not significantly affected by the water content. This is in contrast to bentonite/ballast fills (MX-80/ballast) and speaks in favor of the use of natural clays since the preparation of bentonite mixtures must be very careful for reaching high densities. Determination of the hydraulic conductivity showed that it is lower than the average bulk conductivity of normal host rock (i.e. <E-10 m/s) for densities at saturation of about 2050 kg/m^3 (dry density 1670 kg/m^3) even at salt contents of up to 20 %, while 30/70 bentonite/ballast mixtures with a density of this order have a conductivity exceeding E-10 m/s even at the moderate salt content 1 %. For about 1900 kg/m^3 density (dry density 1430 kg/m^3) the conductivity of the natural clay is less than E-10 m/s and the swelling pressure somewhat more than 200 kPa when the salt content is 3.5 %. This density gives a swelling pressure of at least 100 kPa – i.e. the pressure required to provide effective rock support – irrespective of the salt content. For 30/70 bentonite/ballast fills with this density the swelling pressure almost vanishes when it is saturated with even weakly brackish water.

A general conclusion is that natural clays with the same properties as the investigated one offer better physical properties for serving as backfills than mixtures of bentonite/ballast mixtures with up to 30 % bentonite content.

Sammanfattning

Tidigare undersökningar har visat att naturliga smektitiska leror som återfyllning av tunnlar och schakt i djupförvar kan utgöra attraktiva alternativ till blandningar av bentonit och ballast både från funktionssynpunkt och när det gäller kostnader. Det har visat sig att grundvatten med hög salthalt kan höja den hydrauliska konduktiviteten och minska svällningsförmågan hos lera i allmänhet och hos bentonit/ballastblandningar i synnerhet och denna undersökning avsåg att bestämma inverkan av salt vatten på de viktigaste egenskaperna hos naturliga smektitiska leror representerade av den tyska Friedland Ton.

Laboratoriepackning av den undersökta leran med tillsats av olika mängd saltfritt vatten visade att packbarheten ger hög densitet och inte påverkas mycket av vattenhalten i motsats till blandningar av bentonit/ballast (MX-80/ballast). Det talar för användning av naturliga smektitiska leror som återfyllning eftersom en hög densitet hos blandningarna kräver noggrann proportionering för att få den önskvärda vattenkvoten. Bestämning av den hydrauliska konduktiviteten visade att den är lägre än medelkonduktiviteten hos berget på förvarsnivå (<E-10 m/s) vid en densitet av 2050 kg/m^3 (torrdensitet 1670 kg/m^3) även för salthalter upp till 20 %, medan blandningar av 30/70 bentonit/ballast med samma densitet har en konduktivitet som överskrider E-10 m/s till och med vid den måttliga salthalten 1 %. Vid en densitet av 1900 kg/m^3 (torrdensitet 1430 kg/m^3) är konduktiviteten hos den naturliga leran ca E-10 m/s och svälltrycket något mer än 200 kPa då salthalten är 3.5 %. Denna densitet ger ett svälltryck av minst 100 kPa – dvs erforderligt tryck för att stödja bergtaket i tunnlar – oberoende av saltinnehållet. För blandningar av 30/70 bentonit/sand med densiteten 1900 kg/m^3 i vattenmättat skick upphör i det närmaste svälltrycket redan då porvattnet är något bräckt.

En allmän slutsats är att naturliga leror med samma egenskaper som den undersökta erbjuder bättre fysikaliska egenskaper som återfyllningar i slutförvar än blandningar av bentonit och ballast med upp till 30 % bentonitinnehåll.

Contents

1 Scope

Earlier comparison of various fine-grained soils for backfilling of KBS3 tunnels and shafts has indicated that natural smectitic clays may serve equally well as mixtures of montmorillonite-rich Na bentonite and suitable ballast material which are presently prescribed for the concept [1]. A clay material of this sort, available in large quantities with very homogeneous constitution is the Tertiary Friedland clay in northeastern Germany. Preliminary laboratory experiments have indicated that Friedland Ton, which has a clay (minus 2 μm) content of about 90 % of which about 50 % are expandables (montmorillonite and mixed-layer smectite/mica), is relatively insensitive to salt, while bentonite/ballast mixtures are known to become very permeable when saturated with salt water. The objective of the present study was to investigate more systematically how Na and Ca solutions affect the hydraulic conductivity and swelling pressure of Friedland Ton with different bulk densities.

2 Test program

2.1 General

The investigation comprised the following tests:

1. Determination of the grain size distribution of Friedland clay prepared with a granule size that is estimated to be suitable for field compaction.

2. Laboratory tests for determination of the compaction properties and optimum water content.

3. Oedometer tests for determination of swelling pressure and hydraulic conductivity.

The clay material and tests are described in the report with focus on how the physical properties depend on the salt content of the porewater.

2.2 Description of material

2.2.1 Mineralogy

The clay contains 45 % expandable minerals (montmorillonite and mixed-layer mica/montmorillonite), 24 % quartz, 5 % feldspars, 13 % mica, 11 % chlorite, and 2 % carbonates [2]. The average chemical composition is as follows:

SiO_2 57 %, Al_2O_3 18 %, Fe_2O_3 5.5 %, MgO_2 2 %, CaO 0%, Na_2O 0.9 %, K_2O 3.1 %.

Na is the dominant adsorbed cation according to the manufacturing company DURTEC, Ihlenfelder Strasse 153, 17034 Neubrandenburg, Germany.

2.2.2 Grain size distribution

The manufacturer has given the grain (granule) size distribution data in Table 2.1, which are in good agreement with the results obtained in the present study. The water content of the material was 6.7 %.

Table 2.1. Grain size distribution of Friedland clay delivered for the present study. Data given by DURTEC GmbH.

Fractions, mm	Percentage of grain size representing each fraction
2–8	20.0
1–2	20.4
0.1–1	42.4
<0.1	17.2
Total	100

2.3   Tests performed

2.3.1   General

The study comprised two separate test series representing different strategies. One was performed by Clay Technology AB (CT) and consisted of separate tests in which samples prepared by Proctor compaction in 100 mm diameter cylinders were saturated with the same type of solution that was subsequently percolated through them (Figure 2.1). In the other, which was made by DURTEC GmbH in cooperation with Geodevelopment AB (called D/G in the text), a powder sample was compressed into an oedometer with 100 mm diameter and 20 mm height to a density corresponding to 1900 kg/m^3 after water saturation (dry density 1430 kg/m^3) and then percolated from below in test series using a number of different solutions.
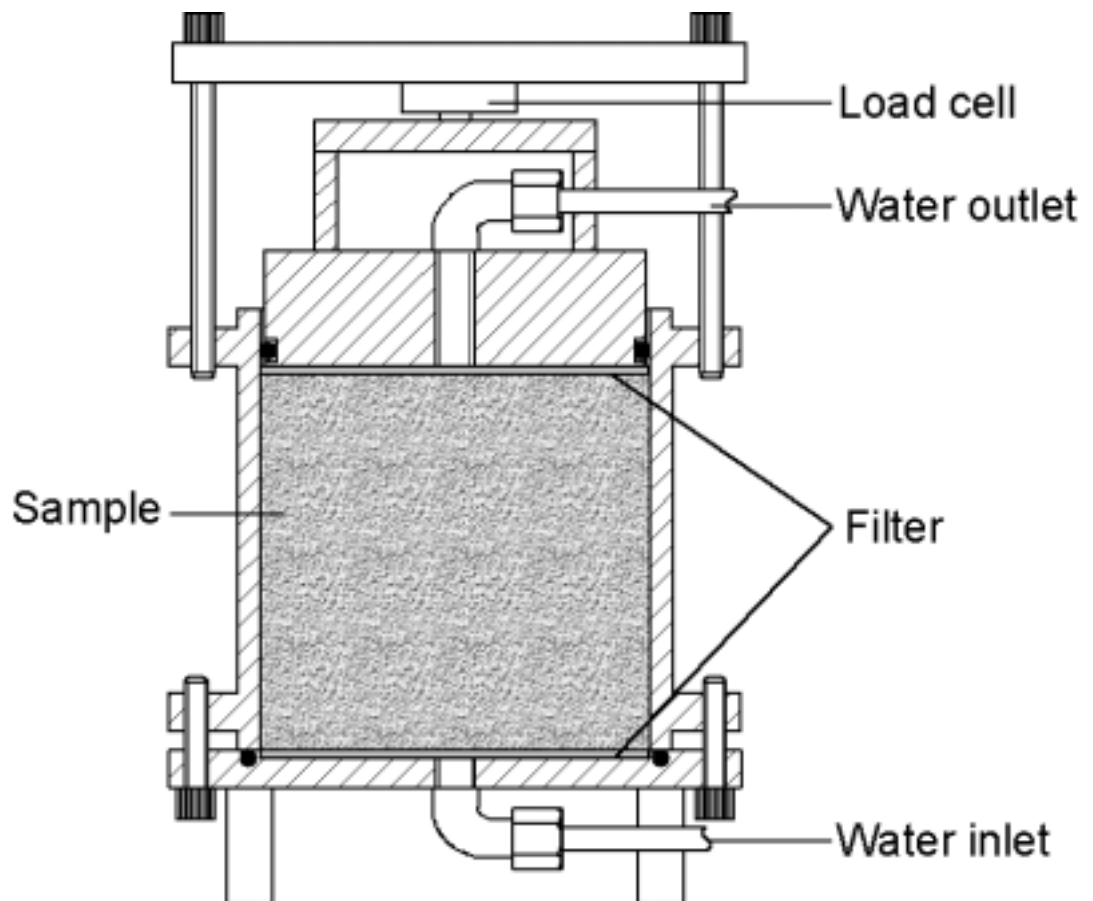
Figure 2.1. *Schematic picture of cell for compaction and subsequent percolation with different solutions with concurrent recording of the hydraulic conductivity and swelling pressure (CT). The height of the soil column was about 20 mm at the hydraulic testing, the diameter of the cell being 100 mm.*

The test pilosophies of the two series were different. Thus, while the experiments performed by D/G aimed at simulating the conditions on site with intial saturation and percolation with very salt water and then altering the composition of the percolated water successively from salt-free to very salt states in order to illustrate the impact of possible changes in groundwater composition, the CT tests were made on separate samples with no influence of preceding experiments.

2.3.2 Specification

Table 2.2. Is a compilation of the test program. K=Hydraulic conductivity, p_s=Swelling pressure.

No	Porewater	Density at sat., kg/m³	Property	Investi-gator	Comment
1	Distilled water	Different	Compactability	CT	Water content varied
2	20 % CaCl₂	1860	K, p_s	CT	Saturation and percolation
3	20 % CaCl₂	1890	K, p_s	CT	Saturation and percolation
4	20 % CaCl₂	1940	K, p_s	CT	Saturation and percolation
5	20 % CaCl₂	2020	K, p_s	CT	Saturation and percolation
6	10 % CaCl₂	1810	K, p_s	CT	Saturation and percolation
7	10 % CaCl₂	1880	K, p_s	CT	Saturation and percolation
8	10 % CaCl₂	1935	K, p_s	CT	Saturation and percolation
9	10 % CaCl₂	2025	K, p_s	CT	Saturation and percolation
10	3.5 % CaCl₂	1750	K, p_s	CT	Saturation and percolation
11	3.5 % CaCl₂	1790	K, p_s	CT	Saturation and percolation
12	3.5 % CaCl₂	1935	K, p_s	CT	Saturation and percolation
13	3.5 % CaCl₂	1995	K, p_s	CT	Saturation and percolation
14	20 % NaCl	1800	K, p_s	CT	Saturation and percolation
15	20 % NaCl	1910	K, p_s	CT	Saturation and percolation
16	20 % NaCl	1975	K, p_s	CT	Saturation and percolation
17	20 % NaCl	2055	K, p_s	CT	Saturation and percolation
18	10 % NaCl	1805	K, p_s	CT	Saturation and percolation
19	10 % NaCl	1875	K, p_s	CT	Saturation and percolation
20	10 % NaCl	1930	K, p_s	CT	Saturation and percolation
21	10 % NaCl	1990	K, p_s	CT	Saturation and percolation
22	Distilled water	1750	K, p_s	CT	Saturation and percolation
23	Distilled water	1865	K, p_s	CT	Saturation and percolation
24	Distilled water	1900	K, p_s	CT	Saturation and percolation
25	Distilled water	2030	K, p_s	CT	Saturation and percolation
I	20 % CaCl₂	1900	K, p_s	D/G	Saturation and percolation
II	Distilled water	1900	K, p_s	D/G	Directly after Test no I
III	10 % CaCl₂	1900	K, p_s	D/G	Directly after Test no II
IV	Distilled water	1900	K, p_s, flow type	D/G	Directly after Test no III
V	3.5 % CaCl₂	1900	K, p_s, flow type	D/G	Directly after Test no IV

3 Test report

3.1 General

The experiments were conducted in the period ranging from early March to mid July year 2000. The results of the two series agree reasonably well and they are also very similar to earlier test results with respect to the hydraulic conductivity and swelling pressure as shown in this report. The study demonstrates that the physical behavior of the investigated natural clay is not significantly affected by high concentrations of sodium and calcium solutions at densities that are expected to be reached at field compaction of air-dry, granulated clay powder.

The D/G test series gave indications of microstructural changes caused by cation exchange and coagulation effects, and the outcome contributes considerably to the understanding of the influence of the porewater chemistry on the performance of expandable clays.

3.2 Influence of porewater chemistry on clay performance

3.2.1 Swelling pressure components

Steric water

The swelling pressure is due to interlamellar hydration of expandable minerals and to osmosis related to interparticle forces. The firstmentioned depends on the type of adsorbed interlamellar cation, which determines the maximum number of hydrates and hence the maximum c-dimension of the stacks of lamellae. Thus, as shown by Table 3.1, the maximum interlamellar aperture is about 10 Å for fully hydrated Na montmorillonite and about 6 Å for Ca montmorillonite. These values correspond to a bulk density at saturation of around 1500–1600 kg/m^3 for Na montmorillonite clay assuming perfectly parallel and uniformly spaced stacks of lamellae and about 1700–1800 kg/m^3 for Ca montmorillonite, assuming that the stacks contain 3–5 lamellae in Na clay and about 10 lamellae in Ca clay. This means that interlamellar hydration does not contribute to the bulk swelling pressure at lower densities. Higher densities mean that such hydration is hindered, which is manifested by a swelling pressure that may be many tens of MPa when there is only one interlamellar hydrate layer, i.e. if the bulk density exceeds about 2100 kg/m^3 at saturation.

Table 3.1. Number and thickness of interlamellar hydrate layers in Å for montmorillonite clay [3].

M^1	1 hydrate	2 hydrates	3 hydrates
Mg	3.00	3.03	3.05
Ca	3.89	2.75	–
Na^2	3.03	3.23	3.48
K	2.42	3.73	–

[1] M adsorbed cation [2] Lithium resembles Na in this respect

Osmotic pressure

Swelling pressure investigations unanimously demonstrate that Na montmorillonite with porewater that is poor in electrolytes has a swelling pressure even for bulk densities at saturation below 1300 kg/m³ and that Ca montmorillonite shows no swelling pressure for less than about 1500 kg/m³ at saturation. Since steric interlamellar water does not contribute to the pressure at these low densities it is believed that the discrepancy results from errors in using concentrations rather than activities of the exchangeable cations and from neglecting formation of clay gels of tactoid type between dense aggregates of stacks [4], yielding a swelling pressure even after complete expansion of the stacks. The swelling pressure at this stage is largely due to double-layer interaction.

3.2.2 Thermodynamic theory

A thermodynamic model worked out by Yong and Warkentin [5] can be used for calculating pressures due to double-layer interaction. It is based on the assumption that the swelling pressure of clays consisting of charged tabular particles in parallel arrangement is determined by double-layer effects.

According to the theory overlapping of the diffuse layers of exchangeable cations results in a higher cation concentration between the particles than in bulk porewater, yielding an osmotic pressure that can be calculated by use of the van't Hoff equation. Following Yong and Warkentin one can express the concentration C_c of the cation concentration at the midpoint between two interacting plates as given by Eq. 3.1:

$$C_c = \pi^2/z^2 \, B(d + x_o)^2 \, 10^{-16} \qquad\qquad (3.1)$$

where:

z = valence of the exchangeable adsorbed cation
$B = 10^{15}$ cm/mmol
d = half distance between two clay plates in Å
x_o = correction factor of 1–4 Å

The interparticle spacing is given by Eq. 3.2 according to Yong and Warkentin. Expressing w in weight percent and S in m²/g, d is given in Å units:

$$d = 100 \, w/S \qquad\qquad (3.2)$$

where:

w = water content
S = the specific surface area.

Applying the van't Hoff equation, the swelling pressure p_s has the form given by Eq. 3.3:

$$p_s = RT(C_c - 2\,C_o) \hspace{4cm} (3.3)$$

where:

R = gas constant
T = absolute temperature
C_o = concentration of cations in the bulk solution in mol/l

The swelling pressure can be obtained from Eqs. 3.1 to 3.3 for any particle spacing, which can be estimated on the basis of the geometrical size and shape of the stacks of lamellae for a given bulk density. The basic theory gives much lower pressures than actually recorded as illustrated by the fact that MX-80 clay should have a bulk density at saturation of at least 1950 kg/m^3 to give a noticeable swelling pressure for 3.5 % NaCl porewater salinity while experiments show that a density of 1800 kg/m^3 at saturation yields an actual swelling pressure of about 200 kPa for this salinity [6]. However, modification of the theory [4] in much the same way as was recently made by Karnland [6] has given fair agreement beween calculated and experimental pressures. Hence, the altered form of the theory gives a theoretical swelling pressure of about 1 MPa for MX-80 clay with 1900 kg/m^3 density and 3 MPa for 2000 kg/m^3 when the clay is saturated with 3.5 % NaCl solution, while for 20 % NaCl, these densities yield a pressure of approximately 1 MPa irrespective of whether Na or Ca are in exchange positions. Using the modified theory the investigated natural clay with about half the content of expandables of MX-80, would give a swelling pressure of about 500 kPa for 1900 kg/m^3 density and 1.5 MPa for 2000 kg/m^3 density saturation with 3.5 % salt solution for both Na and Ca.

### 3.2.3 Impact of microstructure

Themodynamic models

Yong and Warkentin originally considered the role of the microstructure only in terms of particle orientation but later introduced tactoid-type elements to account for explaining the underestimation of the swelling pressure for salt porewaters [4]. For structurally isotropic clay with the particle orientation being uniformly distributed in space, the *S*-value to be introduced in Eq. 3.2 was taken as one third of the true value for montmorillonite, which is about 800 m^2/g. Similarly, the *S*-value should be 270 m^2/g also for clay buffers prepared by compacting clay granules, yielding a swelling pressure of about 1/3 of the value obtained for perfectly parallel particle orientation.

Stacks as primary particles

While Yong and Warkentin only considered monolamellar colloidal systems, both natural and artificially prepared expandable clays are built up of secondary particles, i.e. stacks of lamellae. The charge conditions within the stacks are different from those at the external surfaces of the stacks in the sense that only cations are present in the interlamellar space while complete electrical double-

layers are formed at the outer surfaces. At high bulk densities the charge conditions and the energy state of the hydrates in the interlamellar space control the expandability and swelling pressure, while at low densities, the interaction of stacks through their electrical double-layers determines these properties.

The impact of porewater salinity and bulk density on the swelling pressure caused by the stacking is illustrated by Figure 3.1. For Ca smectite the much fewer contacts between stacks, i.e. the lower frequency of interacting external double-layers, would mean that the influence of the salt content on the swelling pressure is appreciably smaller than for the Na case at low and intermediate bulk densities. Hence, in contrast to the statement in [6] there should be a difference between Na and Ca smectites as to their swelling behavior because of microstructural differences.
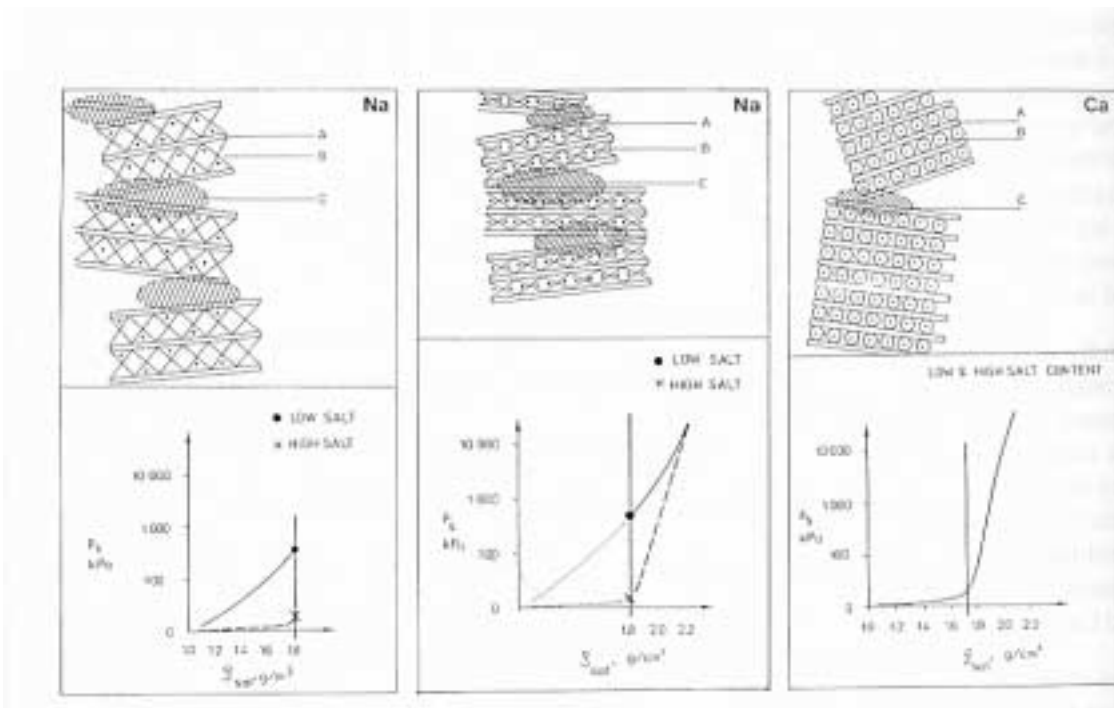
Figure 3.1. Schematic pictures of stack assemblages and influence of density (at water saturation, expressed in g/cm³) and salinity for Na and Ca montmorillonite clay. A) Lamella, B) Interlamellar space, C) Stack contact region with interacting electrical double-layers [6].

Density variation

Variation in density on the microstructural scale is well documented [7, 8] and has a much stronger impact on the swelling pressure than the particle orientation for clays prepared by compaction of clay granules. Thus, it has been demonstrated that the swelling pressure is a function of the volume fraction of dense aggregates and their density [7, 8]. The microstructural data required for predicting the swelling pressure can be obtained from application of the GMM model [8] and calculation of the swelling pressure has yielded values that are in very good agreement with experimental data for MX-80 in Na form for densities down to about 1500 kg/m³ at saturation.

3.3 Test description

3.3.1 Preparation

Test No 1 was made by compacting differently wetted clay powders in the Proctor cell (Figure 2.1) using the standard procedure for compaction tests, while the clay powder was compacted in two 2 cm layers tests in No 2–25 for determining the hydraulic conductivity and swelling pressure.

Tests No I–V were made on clay powder compacted in the oedometer by use of a hydraulic jack operating on a piston that fitted well in the oedometer ring. This procedure gave a more uniform structure and density of the sample, while Proctor compaction gave larger variations in homogeneity and a somewhat lower density at the periphery, which should give conservative values of the hydraulic conductivity and swelling pressure.

3.3.2 Test procedure

The clay materials in the cells were saturated with the respective solutions from below with concurrent recording of the swelling pressure. At pressure equilibrium recording of the rate of percolation was made by use of burettes. Backpressure was used in all tests except Tests No 2, 6, 10 14, 18 and 22, which all represented the lowest densities. The hydraulic gradient was in the range of 20 to 200. While Tests No 2–25 were made on different samples using only 1–2 days for recording the hydraulic conductivity, Tests No I–V were made on one and the same sample by exposing it to different solutions for much longer periods of time.

The hydraulic conductivity was evaluated by use of Darcy's law. At the termination of the tests the bulk density and water content of all samples were determined.

3.4 Results of Tests No 1–25 (CT)

3.4.1 Compactability (Test No 1)

The outcome of the Proctor compaction tests is shown in Figure 3.2, from which one concludes that the density of laboratory-compacted Friedland clay is not sensitive to variations in water content (distilled water). Thus, the dry density does not vary beyond the range of 1750 to 1790 kg/m^3 when the water content is changed from 7 to 21 % by weight. These densities correspond to 2100 to 2130 kg/m^3 at complete saturation. On site in a repository lower densities are expected and the range used in the laboratory experiments was therefore taken to be 1750 to 2050 kg/m^3.

It is of interest to notice that laboratory compaction of 30/70 bentonite/ballast mixtures yields dry densities on the same order of magnitude as the natural clay investigated in the present study but the water content must then be in a rather

narrow water content interval, i.e. 10–15 % [9]. From a practical point of view it is concluded that the natural clay is superior to 30/70 bentonite/ballast mixtures because it will have a high density without manipulating the moisture content. The wetting and careful control of the moisture distribution that is required for mixed backfills are hence superfluous.
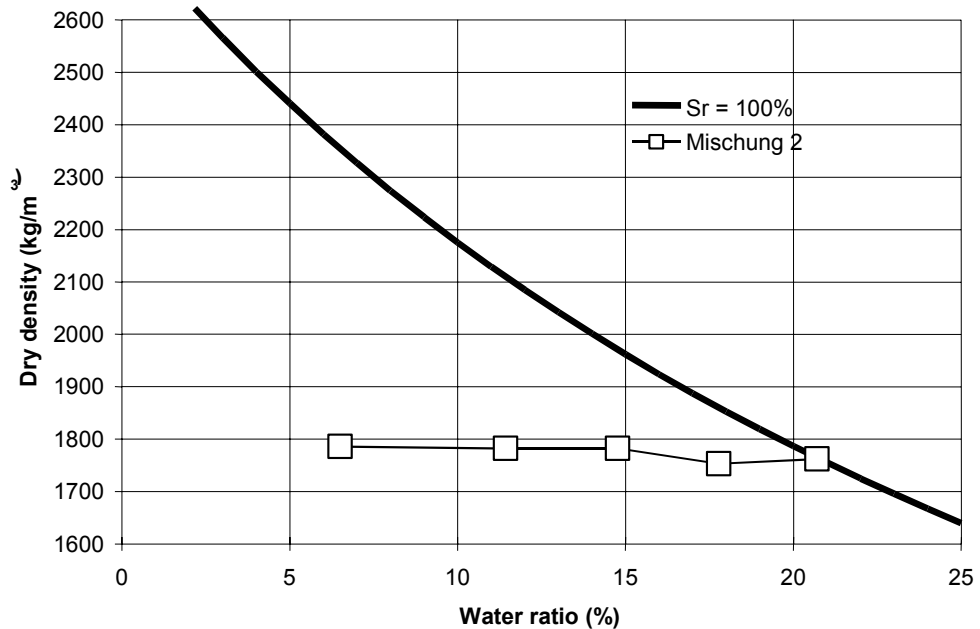

Figure 3.2. *Compaction tests (Test No 1, CT). (Mischung 2 represents the grain distribution in Table 2.1). The thick curve represents the theoretical maximum density (no voids).*

3.4.2 Hydraulic conductivity (Tests No 2–25, CT)

Figure 3.3 shows that there is no dramatic change in conductivity when the salinity is increased from that of ocean water to 20 %, hence providing an answer to the most important question that was raised at the planning of the test series. One can draw the following major conclusions from the study:

1. The conductivity does not exceed E-10 m/s for any salt content if the bulk density at saturation is 2050 kg/m^3 (dry density 1670 kg/ m^3) or higher. This value equals the average conductivity of the host rock at 500 m depth [9].

2. The conductivity does not exceed E-10 m/s for 3.5 % salt content of Ca-dominated water if the density at saturation is about 1950 kg/m^3 (dry density 1510 kg/ m^3)

3. Sodium gives a slightly higher conductivity than calcium, which is explained by the higher ion concentration, yielding stronger coagulation and larger voids. The values are about 2 times higher than for Ca.
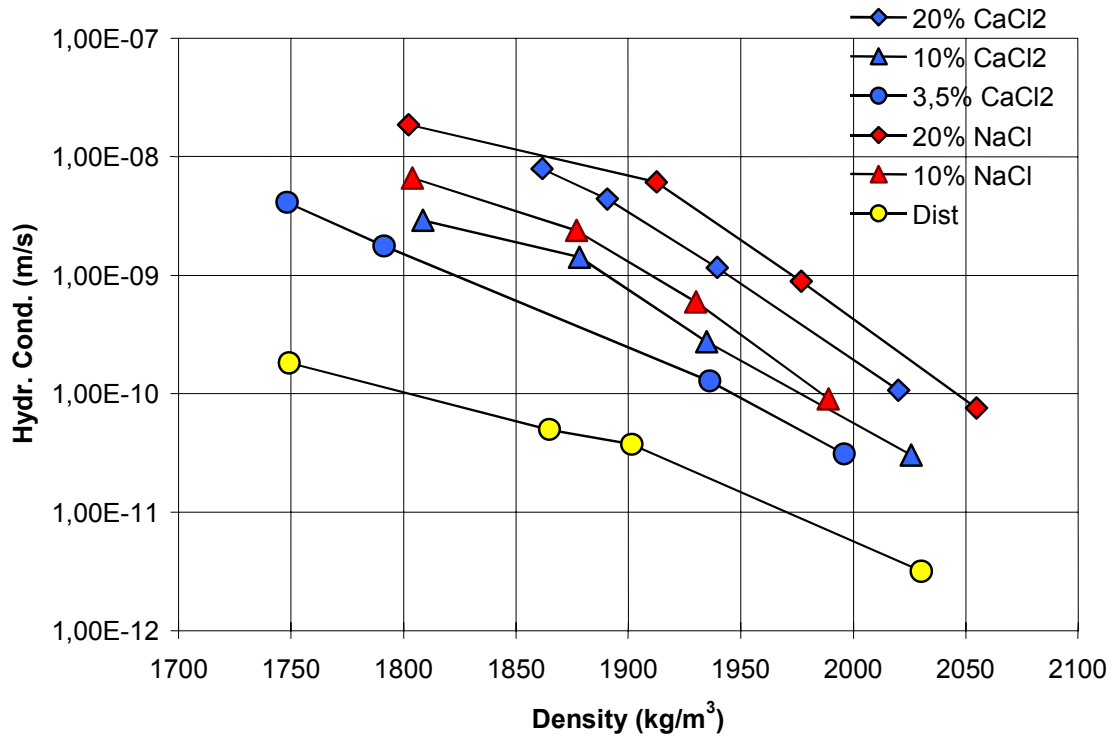
Figure 3.3. *Hydraulic conductivity tests on saturated samples (Tests No 2–25, CT).*

3.4.3 Swelling pressure (Tests No 2–25, CT)

Figure 3.4 shows that the swelling pressure exceeds the stipulated 100 kPa level for 1900 kg/m^3 and higher densities irrespective of the salt content of the porewater. The following general conclusions can be drawn from the graph:

1. For densities at saturation exceeding 1950 kg/m^3 (dry density 1510 kg/ m^3) the swelling pressure is higher than 200 kPa for all the investigated salt contents, and for 2050 kg/m^3 (dry density 1670 kg/ m^3) it is about 1 MPa for distilled water as well as for 20 % salt content (Na). For a density of 2100 kg/m^3 (dry density 1750 kg/m^3) earlier experiments have given the pressures 2.6 and 2.65 MPa when the clay is saturated with distilled water and 3.5 % CaCl$_2$ solution, respectively [2]. These data fit rather well with those obtained in the present study.

2. The linear rise in swelling pressure when the density at saturation is increased from 1900 to 2030 kg/m^3 and the clay saturated with distilled water is only apparent. In fact, the pressure for 1950 kg/m^3 is about 500 kPa.

3. The pressure span for bulk densities exceeding 1850 kg/m^3 (dry density 1350 kg/ m^3) is not more than about 200 kPa for the actual electrolyte spectrum.
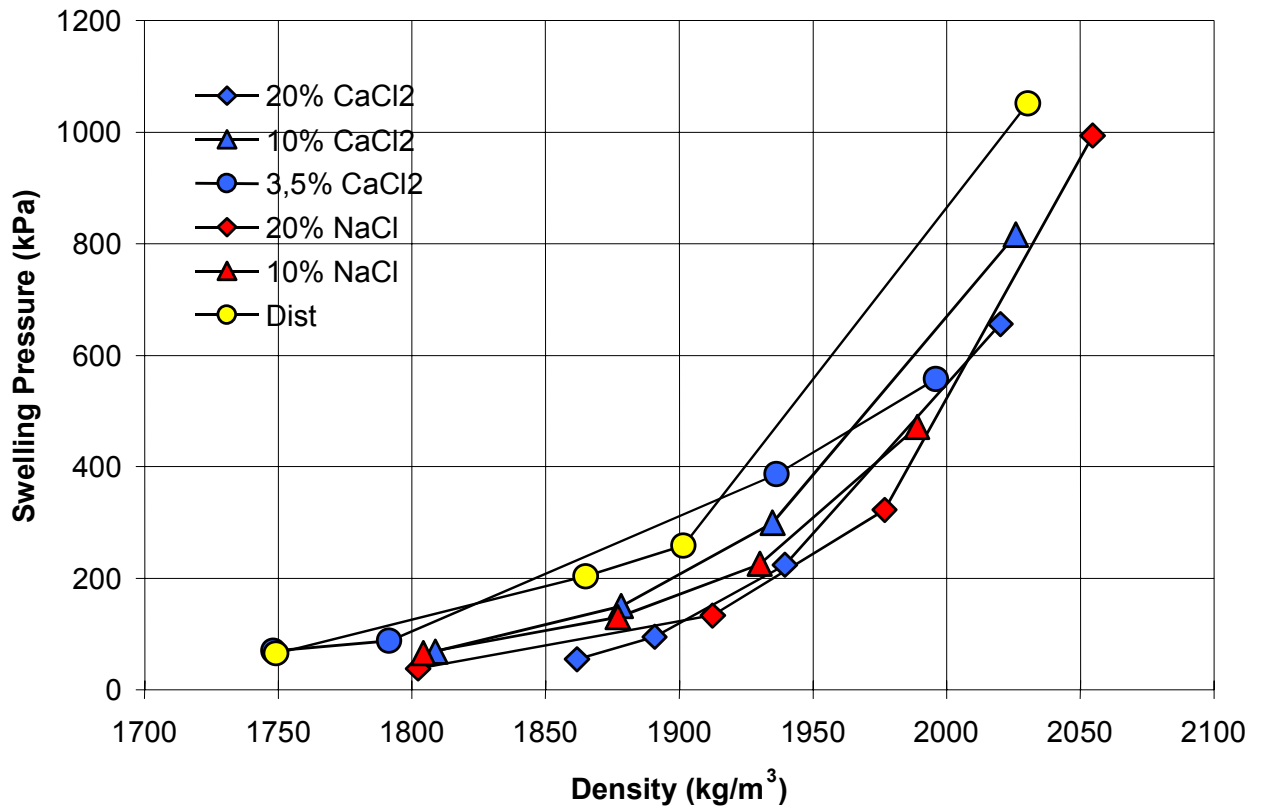
Figure 3.4. *Swelling pressure tests (Tests No 2–25, CT).*

3.5 Results of Tests No I–V (G/D)

3.5.1 General

The test equipment and performance deviated somewhat from those of Tests No 2–25, which has an impact on the results. This is commented on first.

Equipment

The test series was performed by use of an oedometer with 100 mm diameter and 20 mm height. The cell wall was threaded, the grooves being filled with stiff MX-80 paste for eliminating leakage along the periphery of the sample. Stainless steel filters with 1 mm thickness and 100 μm pore size confined the sample. The hydraulic gradient across the sample varied between 10 and 20 during the test series. The hydraulic conductivity values evaluated from the burette readings that gave the in- and outflow and the hydraulic gradient in the course of the percolation had an accuracy of better than 5E-13 m/s. The swelling pressure was recorded by use of strain gauges mounted on the oedometer bolts, the accuracy of the evaluated pressures being +/– 10 kPa.

Test procedure

Initial saturation was achieved by exposing the sample to 20 % $CaCl_2$ solution from the bottom filter until the hydraulic gradient was evened out and the sample was then left for reaching microstructural equilibrium for three days. It was then percolated with the various solutions.

The sample was maintained in the cell throughout the test series. The concentration of the percolate was kept largely constant by replacing the solutions in the burettes every third day. The salinity of the discharged water was measured in some of the tests.

The experimental procedure was as follows:

- Compaction of air-dry clay powder to a dry density of 1430 kg/m^3 for reaching a bulk density of 1900 kg/m^3 at complete water saturation. The actually obtained value was found to be slightly less than 1900 kg/m^3.

- The first step was to saturate the air-dry, compacted clay with 20 % $CaCl_2$ solution with concurrent recording of the swelling pressure. Starting with a very salt solution was decided for simulating the most critical condition that can be foreseen under field conditions.

- The second step was to percolate the sample with 20 % $CaCl_2$ solution with recording of swelling pressure and evaluation of the hydraulic conductivity. The test continued until the trend of dropping conductivity became obvious.

- The third step was to percolate the sample with distilled water. The test continued to yield almost steady state conditions with respect to swelling pressure and hydraulic conductivity.

- The fourth step was to percolate the sample with 10 % $CaCl_2$ solution to yield largely steady state conditions with respect to swelling pressure and hydraulic conductivity.

- The fifth step was to percolate the sample with distilled water. The test continued to yield almost steady state conditions with respect to swelling pressure and hydraulic conductivity.

- The sixth step was to percolate the sample with 3.5 % $CaCl_2$ solution to yield steady state conditions with respect to swelling pressure and hydraulic conductivity.

- The last step was to reverse the flow direction in the last test to see whether particle transport caused by flowing water was significant.

It was clear from the beginning that percolation with different solutions of the same sample would not give complete chemical equilibrium since ion diffusion and exchange would require several weeks. However, it was expected that relevant values would be obtained if the tests had sufficient duration and this goal is believed to have been reached. The fact that the drop in conductivity had not ceased completely at the termination of several of the tests makes the results from these experiments somewhat conservative.

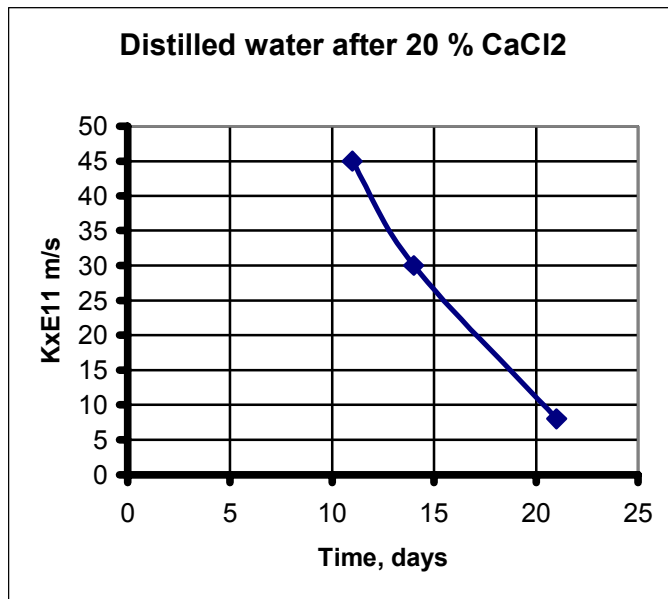### 3.5.2   Hydraulic conductivity (Tests I–V, G/D)

The results of the conductivity tests are shown in Figures 3.5 to 3.9. They show the time-dependent evolution of flow after the initial change that is due to adaption of samples and tubings to the imposed pressure state. This initial period lasts for less than a day after application of the constant pressure.
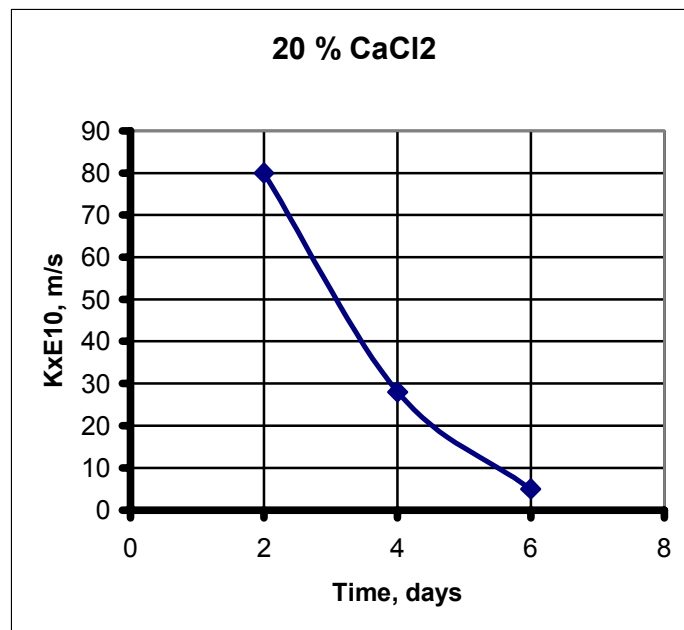
Figure 3.5. *Recorded hydraulic conductivity for percolation with 20 % CaCl$_2$ solution (Test No I, G/D).*

The flow through the clay sample was initially very high but dropped to about E-8 m/s after two days and further to about 5E-10 m/s after 6 days. The drop in conductivity is believed to be due to continued homogenization after the initial hydration and to clogging of voids by migration of particle aggregates set free and transported by flowing porewater.

Immediately after termination of the test with 20 % salt solution, the burettes and all tubings were rinsed and filled with distilled water for the subsequent test (Figure 3.6).

Figure 3.6. *Recorded hydraulic conductivity for percolation with distilled water after the 20 % CaCl₂ solution treatment (Test No II, G/D).*

The flow through the clay sample stayed approximately constant from the termination of the preceding test (about 5E-10 m/s) for the first ten days but then dropped to about 7E-11 m/s after another 10 days. The delay is due to the rather slow ion diffusion of calcium ions out from the clay, these ions being replaced by protons. The exchange caused dispersion and reduction in size of open or gel-filled voids and hence the observed drop in conductivity.
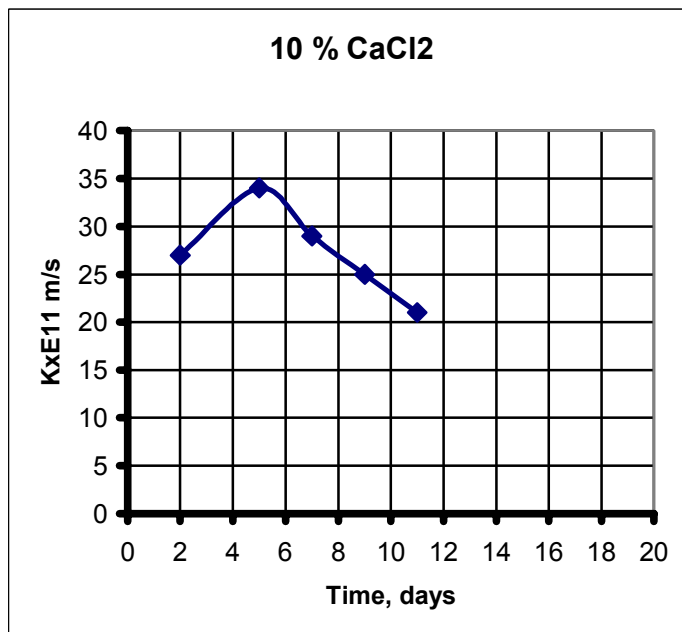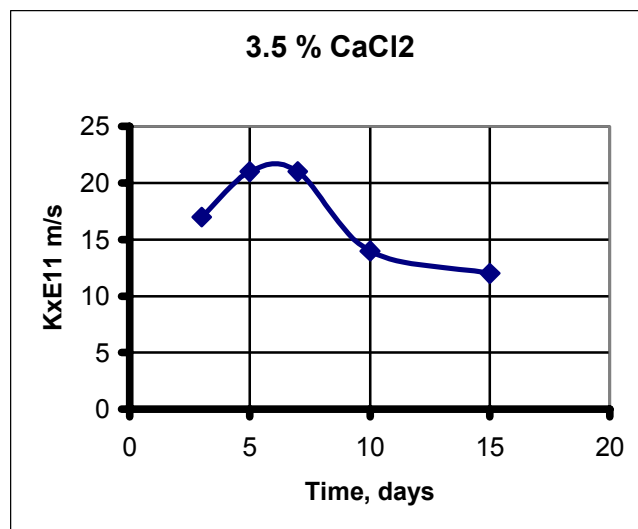

Figure 3.7. *Recorded hydraulic conductivity for percolation with 10 % CaCl₂ solution after the test with distilled water (Test No III, G/D).*

The flow through the clay sample rose slightly in the first five days and then dropped to yield a hydraulic conductivity of about 2E-10 m/s after about 11 days. This drop is believed to be due to migration of small particle aggregates causing clogging of larger voids.

Distilled water after 10 % CaCl2
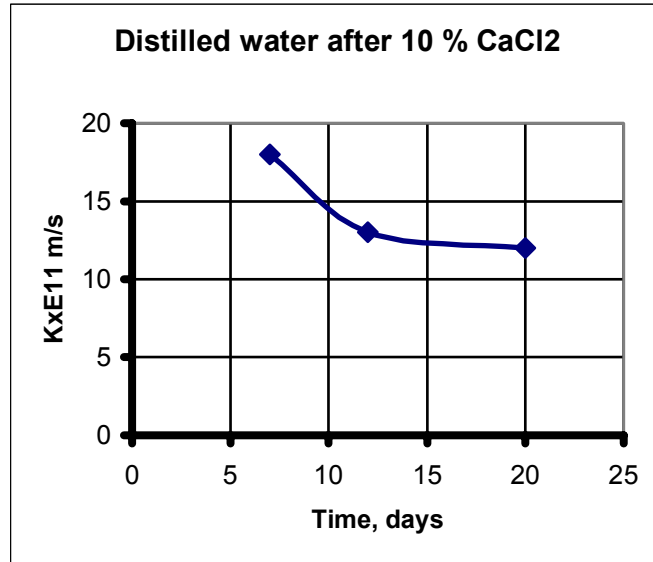
Figure 3.8. *Recorded hydraulic conductivity for percolation with distilled water following the test with 10 % CaCl$_2$ (Test No IV, G/D).*

The hydraulic conductivity remained constant at about 1.9E-10 m/s in the preceding test with 10 % CaCl$_2$ for about a week and then dropped to 1.2E-10 m/s in the following two weeks, indicating some dispersion and reduction in size of open or gel-filled voids.

3.5 % CaCl2

Figure 3.9. *Recorded hydraulic conductivity for percolation with 3.5 % CaCl$_2$ following the test with distilled water (Test No V, G/D).*

The hydraulic conductivity increased from the value 1.2E-10 m/s in the preceding test with distilled water to 2.2E-10 m/s in three days and then dropped back to about 1.2E-10 m/s in the following ten days. This indicates that clogging of voids by particle aggregates transported by flowing water compensated for the increase in conductivity caused by salt-induced coagulation of clay particles.

For testing the hypothesis that particle transport has a significant effect on the hydraulic conductivity the flow direction was reversed after termination of Test No V. The diagram in Figure 3.10 validates the hypothesis by demonstrating that the conductivity went up about three times in one day, after which it dropped to the initial value in the subsequent two days due to migration of released aggregates to new, clogging positions.
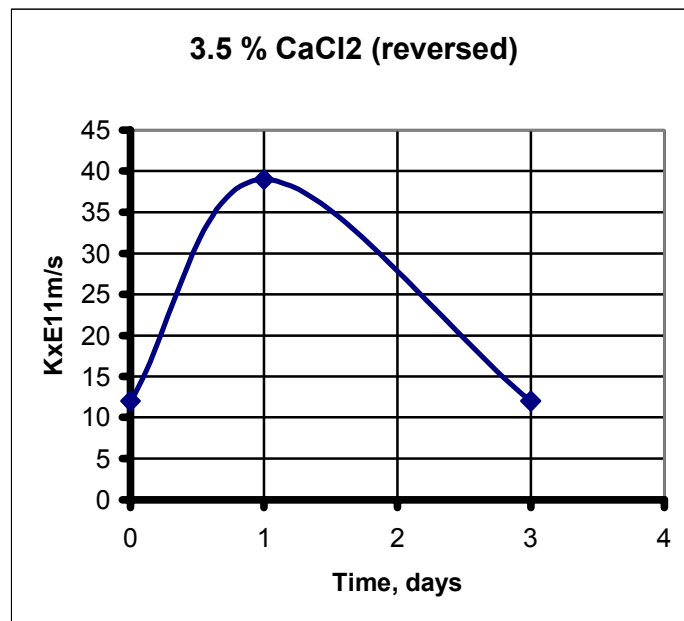
Figure 3.10. *Change in hydraulic conductivity by reversing the flow direction after completion of Test No V.*

3.5.3 Discussion of results of Tests No I–V, G/D

The evaluated hydraulic conductivity values of Tests No 1–V are summarized in Table 3.2.

Table 3.2. Evaluated hydraulic conductivity at the end of each test series.

Porewater	Time, days	Hydraulic conductivity, m/s	Max. value in test, m/s
20 % CaCl$_2$	6	4.7E-10	8.0E-9 (initial)
Distilled water	21	7.5E-11	5.8E-10 (initial)
10 % CaCl$_2$	13	2.5E-10	2.5E-10 (final)
Distilled water	19	1.2E-10	1.8E-10 (initial)
3.5 % CaCl$_2$	18	1.2E-10	2.1E-10 (after 5 days)
3.5 % CaCl$_2$, re-versed flow direct.	3	1.2E-10	3.8E-10 (after 1 day)

Like the experiments comprising Tests No 2–25, Tests No I–V showed that there is no dramatic change in conductivity when the salinity is increased from very low values to 20 % if the density is 1900 kg/m³ or higher. One can draw the following major conclusions from the latter series:

1. The hydraulic conductivity for the very high salt content 20 % $CaCl_2$ is on the order of E-9 m/s, or about 3 times lower than indicated by Test No 3.

2. The hydraulic conductivity for the high salt content 10 % $CaCl_2$ is about 2E-10 m/s, or about 4 times higher than indicated by Test No 8.

3. The hydraulic conductivity does not exceed about E-10 m/s for 3.5 % salt content of Ca-dominated water, which is about 3 times lower than indicated by Test No 12.

4. The hydraulic conductivity for percolation with distilled water is about E-10 m/s, which is about 2 times higher than indicated by Test No 24. The difference is probably that the homogenization of the clay in the latter test was very good while some salt-induced microstructural organization remained when percolation of the 1900 kg/m³ sample with distilled water took place in Tests No II and IV.

5. The hydraulic conductivity is relatively independent of the salt content; it ranges between about E-9 and E-10 m/s with a tendency to stabilize at slightly more than E-10 m/s after a number of changes in the chemical composition of the percolate.

The lower conductivity values evaluated from Tests I–V can be explained by the long testing time that allowed transported particles to clog voids, and by the better homogeneity of this sample than of the Proctor-compacted clay in Tests No 2–25.

3.5.4 General conclusions from all hydraulic conductivity measurements

Table 3.3 is a compilation of the hydraulic conductivity measurements.

Table 3.3. Generalized hydraulic conductivities in m/s for different clay densities and salinity of the percolated solution.

Bulk density	20 % salt content	10 % salt content	3.5 % salt content	0 % salt content
2000	5E-10	7E-11	3E-11	7E-12
1950	2E-9	4E-10	E-10	2E-11
1900	5E-10 to 8E-9	2E-10 to E-9	E-10 to 3E-10	4E-11 to E-10
1850	E-8	5E-9	7E-10	7E-11
1800	2E-8	8E-9	2E-9	E-10

Applying the criterion that the hydraulic conductivity of tunnel and shaft backfills should not exceed the average bulk conductivity of the host rock the required conductivity is about E-10 at about 500 m depth [9].

The data in Table 3.3 show that for groundwater salinities of the commonly recorded order at a few hundred meters depth, i.e. 1 to 3 % with calcium as dominant cation, the hydraulic conductivity of the investigated natural clay is about E-10 m/s when the density is 1900 to 1950 kg/m^3 at saturation. In order to obtain this conductivity for a groundwater salinity of 10 % the density should be increased to about 2000 kg/m^3 (dry density 1590 kg/m^3) while the extreme case of 20 % salinity would require a density of 2050 kg/m^3 (dry density 1670 kg/m^3) to yield this conductivity. The compactability of the clay makes it possible to reach the latter density (cf. Figure 3.2) but field tests are required for finding out whether densities of this order can be achieved.

It is of interest to see that mixtures of 30/70 bentonite/ballast with a density at saturation of 1950 to 2090 kg/m^3 at saturation (dry densities 1510 to 1730 kg/m^3) have a conductivity of E-11 when percolated with distilled water, i.e. on the same order of magnitude as the natural clay. The mixtures are, however, very sensitive to the electrolyte content of the percolate as illustrated by the fact that the mixture with 1950 kg/m^3 density had a hydraulic conductivity of 1.1E-10 when tested with 1 % $CaCl_2$ solution [2]. The difference can be explained by the fact that while practically all grains in the natural clay contain expandable minerals that contribute to form a rather homogeneous structure, only about 20 % of the minerals in 30/70 bentonite/ballast mixtures play this role.

3.5.5 Swelling pressure (Tests No I–V, G/D)

The recording of the swelling pressure gave the results compiled in Table 3.4.

Table 3.4. Evaluated swelling pressure in Tests No I–V.

Porewater	Time, days	Swelling pressure, kPa	Max. value in test, kPa
20 % CaCl$_2$	6	190	220
Distilled water	21	220	230
10 % CaCl$_2$	13	190	220
Distilled water	19	195	220
3.5 % CaCl$_2$	15	–	–

Three major conclusions can be drawn from the measurements:

1. The swelling pressure is largely independent of the porewater salinity for the investigated density 1900 kg/m^3 at saturation.

2. The pressures are on the same order of magnitude as in preceding pilot tests, i.e. 300–400 kPa for Friedland clay with a density of 1900 kg/m^3 saturated with distilled water or 3.5 % CaCl$_2$ solution [2].

3. Replacement of the porewater with largely different salinities does not yield significant changes in swelling pressure of the investigated clay with 1900 kg/m^3 density.

3.5.6 General conclusions from all swelling pressure measurements

The results of all the swelling pressure measurements are compiled in Table 3.5.

Table 3.5. Generalized swelling pressures in kPa for different clay densities and salinity of the percolated solution.

Bulk density	20 % salt content	10 % salt content	3.5 % salt content	0 % salt content
2000	550	650	550	800
1950	200	300	400	550
1900	100–200	200	300*	200–250
1850	50	100	200	200
1800	25	50	100	100

* Overestimated

The following major conclusions can be drawn from the measurements:

1. The swelling pressure is at least 100 kPa irrespective of the porewater salinity if the bulk density is 1900 kg/m^3 or higher.

2. The swelling pressure is largely independent of the porewater salinity for the density 1900 kg/m^3.

Comparison with saturated bentonite/ballast mixtures shows that the mixtures perform less well at densities of 1900 to 2100 kg/m^3. Thus, a mixture of 30 % MX-80 and 70 % ballast with 2100 kg/m^3 density yields a swelling pressure of 500 kPa at saturation with distilled water, i.e. a value that is considerably lower than that of the natural clay, and it drops to 200 kPa at saturation with Ca-rich brackish water [2]. This is in contrast with the natural clay of the same density since it shows practically no decrease in swelling pressure on percolation with any of the salt solutions used in the study. For a density of about 1900 kg/m^3 the swelling pressure is estimated to be much less than 100 kPa of 30/70 mixtures and it will drop to almost zero when saturated with Ca-dominant solutions with more than 3.5 % salinity. The reason for the difference in swelling pressure of such mixtures and of the investigated natural clay is that the density of the clay phase in the mixtures will not exceed about 1600 kg/m^3 when the bulk density is in the range of 1900 to 2000 kg/m^3 while the density of large parts of the clay matrix of the natural clay will be the same as the bulk density.

4 Discussion and conclusions

4.1 Assessment of tests and test procedures

The tests are believed to have yielded largely conservative data on the influence of salt solutions on the hydraulic conductivity and swelling pressure because the most comprehensive test series was made on relatively heterogeneous material with some parts having somewhat lower density than average, and the testing time being very short.

The test series with one and the same sample being percolated by different solutions involved flow with concurrent exchange of adsorbed cations and replacement of ions distributed in the voids. The fact that almost steady state flow conditions were obtained in the hydraulic testing indicated that the major part of cation exchange process had taken place when the tests were terminated but the degree of cation exchange was not recorded. However, some information on this issue could be obtained by analyzing water discharged from the oedometer in the course of the test with distilled water (Test No IV) after Test No III with 10 % $CaCl_2$ solution. The measurements were made by recording the electric resistivity of the 10 ml water sample contained in the outflow burette. Table 4.1 gives the data.

Table 4.1. Approximate content of $CaCl_2$ in water discharged from percolated samples.

| Test	Time, days	Hydraulic conductivity, m/s	Concentration, %
No IV, (Distilled water after test with 10 % $CaCl_2$)	8	1.8E-10	5.2
	14	1.3E-10	0.7
	19	1.2E-10	0.3

One finds that the Ca concentration in the discharged water was almost 50 % of that of the infiltrated solution (10 %) in the preceding experiment about one week after start of the percolation with distilled water and remained at a level corresponding to brackish water (300 ppm) even after 3 weeks. This is explained by delayed release of earlier adsorbed Ca. This shows that the large majority of the very salt solution with which the sample had been treated in the preceding experiment had migrated from the sample in somewhat less than three weeks and that the hydraulic conductivity approached a value representative of porewater with a very low salt content. While this conductivity is expected to have been reduced further if the percolation with distilled water had continued, the opposite phenomenon at percolation with salt solutions is also probable. Thus, it would take several weeks to reach an equilibrium stage with all the porewater in the voids filled with the salt water.

One concludes from this checking that the hydraulic conductivity values and swelling pressures from Tests No I–V for a density of 1900 kg/m^3 at saturation should deviate somewhat from those in Tests 2–25. Considering also the influence of the shorter testing time in the latter series it is believed, however, that the true conductivity and swelling pressure for this density represent the mean values of the two series.

4.2 Theoretical modelling

4.2.1 Hydraulic conductivity

The hydraulic conductivity is a function of the presence and interconnectivity of open paths of capillary dimensions. They are extremely small and with limited continuity in very smectite-rich clay like MX-80 of high density and their geometrical features can be evaluated from ultrathin sections of suitably prepared clay samples [10]. In principle, one can distinguish quantitatively between denser and softer parts of the clay matrix by digitalizing micrographs and ascribing to them characteristic average hydraulic conductivities the net average conductivity of a clay element can be calculated (Figure 4.1). Such analyses have yielded good agreement between microstructural parameters and the conductivity of bulk clay.
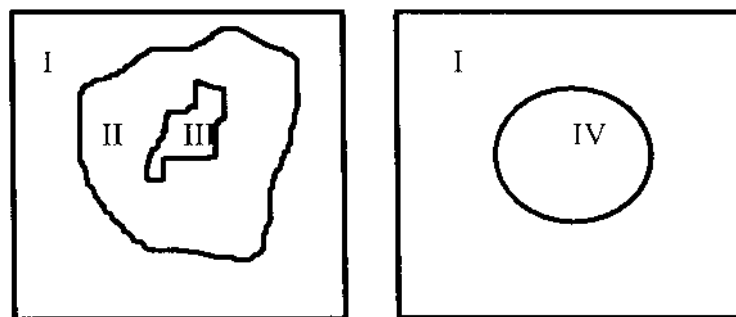
Figure 4.1. *Schematic micrographs. Left: Typical picture of ultrathin section with varying density in soft matrix region (I to III). Right: normalized picture with defined channel cross section and density of the clay gel in the channel (IV) and of the clay matrix (I).*

Typical relationships between the bulk density and gel density are shown in Table 4.2 for MX-80.

Table 4.2. Bulk and gel densities for saturated MX-80 [8].

Bulk density, kg/m^3	Gel density, kg/m^3
2130	2000
1850	1650
1570	1150

There is close similarity of the bulk and gel densities when the former exceeds 1850 kg/m^3, which is explained by a high degree of microstructural homogeneity, while for 30/70 mixtures of bentonite/ballast, the difference is obvious even for higher densities. This means that pervious channels filled with soft clay gels are abundant in the mixtures and since these gels collapse on percolation with salt solutions, forming separate coagulated aggregates and leaving larger voids unfilled, the hydraulic conductivity will be significantly increased.

The microstructure of the investigated natural clay with 1900 kg/m^3 density at saturation is much more homogeneous than that of 30/70 mixtures with the same bulk density but not as unifrom as the clay matrix of MX-80. Thus, Figure 4.2 shows no large variations in density but the presence of open voids with diameters ranging from a few μm down to nanometers, in which minute particles can migrate. The open voids are assumed to be responsible for the lower self-sealing ability of the natural clay than of MX-80 simply because of the different contents of expandable minerals.
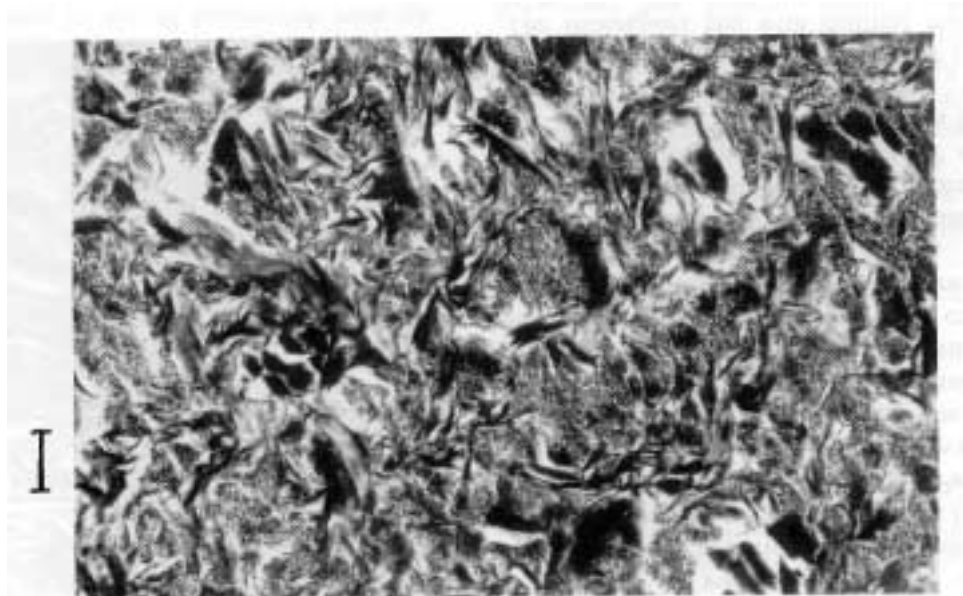
Figure 4.2. *Microstructure of 500 Å ultrathin section of Friedland Ton with a density of 1900 kg/m^3 as observed in a transmission electron microscope. The darkest parts are dense particles of rock-forming minerals, mainly chlorite, medium-gray parts are mainly mixed-layer and montmorillonite clay minerals, while white parts are open voids. The bar is i μm long.*

4.2.2   Swelling pressure

Application of the upgraded version of the Yong/Warkentin theory given in [6] yields pressures that are of the right order of magnitude. Hence, for the investigated natural clay with about half the content of expandables of MX-80 clay and assuming isotropic particle orientation, one would expect a swelling pressure of around 200 kPa for 1900 kg/m^3 density and 500 kPa for 2000 kg/m^3

density irrespective of the adsorbed cation type at saturation with 3.5 % salt solution. This agrees rather well with the actually recorded values that are given in Table 3.5. For 10 to 20 % salt solutions the corresponding calculated pressures should be about 300 kPa for 1900 to 2000 kg/m^3 density, which is about 30 to 50 % higher than the recorded values. The relatively good agreement between theoretical and actual pressures demonstrates that thermodynamically based models that make use of double-layer theories and simple microstructural rules give reasonable results. However, further development is required to take the actual difference between Na- and Ca-adsorption into consideration.

The fact that the large majority of all structural elements contain expandables and have a density comparable to the bulk density, explains that the microstructure of the water saturated clay is much more homogeneous than mixtures of 30/70 bentonite/ballast. The density of the clay component of such mixtures is lower than the bulk density, which makes them vulnerable to high salt contents.

4.3 Implications of the study

The compactability of the investigated natural clay suggests that it can be applied in KBS3-tunnels and shafts with a density corresponding to 1900 to 2100 kg/m^3 after saturation. For about 1900 kg/m^3 density the conductivity is less than E-10 m/s, which is the average bulk conductivity of normal host rock at repository depths, and the swelling pressure somewhat more than 200 kPa when the salt content is 3.5 %, which represents the most saline groundwaters that are ussually found in granitic rock at these depths. This exceeds the pressure required to provide effective rock support irrespective of the salt content. If backfills consisting of the investigated clay can be applied with a density of 2050 kg/m^3 at complete water saturation the hydraulic conductivity is lower than E-10 m/s even at salt contents of up to 20 %, and the swelling pressure will be higher than 600 kPa.

Comparison with 30/70 bentonite/ballast mixtures show that they are less suitable than the investigated clay. Thus, for a density of 1900 to 2100 kg/m^3 at saturation the hydraulic conductivity will exceed E-10 m/s even at moderate salt contents. They can be equally well compacted as the investigated natural clay but it is required that the mixtures are wetted to an optimum water content and that the mixing of the components is very careful for getting homogeneus products. These processes require current quality control and proper storage of the components, which all represents rather high costs. A general conclusion is that natural clays with the same properties as the investigated one offer better physical properties for serving as backfills than mixtures of bentonite/ballast mixtures with up to 30 % bentonite content.

A final note is that a definitive decision on possible replacement of the present number one candidate 30/70 bentonite/ballast backfill by a natural clay of the investigated type requires determination of the density that can be obtained at in-situ compaction or by preparing blocks.

5 References

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