

New perspectives for geosynthetic clay liners using calcium bentonite

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ABSTRACT: Recent investigations of excavated GCLs have shown that sodium (Na) bentonite undergoes an ion exchange by calcium (Ca) ions, converting it partially or fully into a calcium bentonite. As a result the original bentonite properties change, as well as the GCL permeability. It increases extremely when the converted bentonite layer dries getting irreversible cracks. Ca-ions causing ion exchange are usually present in most soils in situ. Initial employment of Ca-bentonite in a GCL eliminates the effect of ions exchange right from the outset. Since 1997 after extended research and testing the utilisation of a sewn GCL containing approximately 10000 g/m² Ca-bentonite has been started. In lab trials with Ca-enriched water the new GCL performed much better compared to the sodium GCL tested. Based on the lower water adsorption of a Ca-bentonite the moisture content in saturated state is much lower than that of a Na-bentonite and the corresponding plasticity index too. In conclusion, the Ca-GCL needs less moisture and is less sensitive to water reductions in site compared with a Na-GCL. The range of shrinking is limited by the low swelling capacity, too. The high bentonite content necessary to achieve a low permittivity makes the new GCL more resistant to mechanical and chemical effects.

1 INTRODUCTION

Geosynthetic clay liners (GCLs) have enjoyed widespread use as a sealing in landfill, roadway and hydraulic engineering over the past ten years. Until recently, exclusively sodium bentonite (natural or activated) has been used as a sealing component in GCLs. Commonly water permeability measured in the laboratory is very low, permittivity (ψ) ranges from 10^{-9} to 10^{-10} s⁻¹, even when GCLs are subjected to dry-wet or frost-thaw cycles.

Once being placed in site, GCLs are subjected to many different physical, chemical and biological influences effecting the original (virgin) permeability in a negative way. The most important factors are: the soil itself (grain size distribution, soil cover thickness respectively pressure, water retention capacity, dissolved ions); the particular climate conditions; the plants (roots) and the soil fauna. Evidently it is impossible to simulate in a lab all these circumstances together.

Investigations of excavated sodium GCLs in Germany in the last years and reports from the USA indicate an increase of the water permeability after a few years in situ. In the cases known, ion exchange in the Na-bentonite has been noticed. Ca-cations from the soil have replaced the Na-cations and changed the original properties of Na-bentonites, reducing e. g. swelling capacity etc. In this way an initial sodium-GCL can be completely converted into a calcium-GCL. The calcium content of most soils is sufficient to induce a remarkable ion exchange as Na-ions are lightly bound and the bentonite quantity in Na-GCLs is relatively small. If such effected GCLs desiccate becoming shrinkage cracks, the reduced swelling capacity makes the entirely sealing impossible and sealing effect of GCL is no more guaranteed.

Ion exchange problems cannot take place if using 'a priori' GCLs with Ca-bentonite. Note that Ca-bentonites are a little more permeable than Na-bentonites. Reasonably a Ca-GCL needs a thicker bentonite layer respectively more bentonite mass per m² to provide sufficient sealing effect.

The development of such a Ca-GCL was accompanied by extensive laboratory tests, investigations and studies. Experience to date would suggest that Ca-GCLs have important advantages.

2 BENTONITE PROPERTIES, INVESTIGATIONS OF VIRGIN SODIUM BENTONITES

The sealing properties of GCL are primarily determined by the bentonite employed. Its main component is the three-layer clay mineral montmorillonite. Bentonites for GCL have about 70 % to 90 % montmorillonite content. 'Montmorillonite is characterised by high specific surface area, high cation-exchange capacity, good plasticity properties, high swelling and water adsorption capacity, low permeability and low coefficients of diffusion' (Madsen, Nüesch 1994). The negative charge of the montmorillonite crystals is compensated by Na⁺, Ca⁺⁺ and Mg⁺⁺ cations. Dominating cations Na⁺ or Ca⁺⁺ determine whether montmorillonite is a sodium or a calcium one. Based on the different ion charge Na⁻ and Ca-bentonites provide distinctive activity to water (Eggloffstein 1997, Madsen, Nüesch 1994). For example (Madsen, Nüesch 1994) specify a water adsorption of 400 % to 700 % for an activated Na-bentonite and from 100 % to 300 % for a Ca-bentonite. Further, typical for sodium bentonites is a swelling volume (free swelling) of more than 25 ml/2g, but only about 10 ml/2g (say less than the half) for Ca-bentonite according to data available. Atterberg limits depend on the ion valency too, see examples in section 4.

One has to keep in mind that in a GCL bentonite is fixed between geotextiles and subjected to vertical stress in site, so free water absorption and free swelling are being changed (decreased).

Considering the noted sensitivity of Na-GCL to drying (shrinkage) with respect to parallel ion exchange (see section 1), more knowledge about the particular water content and consistency of different bentonites under different conditions is necessary. The investigations described below were undertaken with the aim to find out as a first step these properties of virgin bentonites, commonly used in three different GCLs in Germany.

Tests (LGA Bayern, unpublished) were conducted on two natural Na-bentonites, A (granular) and C (powder), and on one activated Na-bentonite B (trade name 'Seal 80-NB'). Additionally, one Ca-bentonite was tested also (see section 4). The investigation covered free water adsorption and Atterberg limits on bulk bentonite, as well as swell heaving under 20 kPa vertical stress and water conductivity under 25 kPa vertical pressure and 300 mm water head. The samples tested had a mass per unit area of 4500 g/m², corresponding to a bentonite content of 4000 to 5000 g/m² in common GCLs. Apart from swell heaving (see section 4) and permittivity, the results for the Na-bentonites A, B and C are summarised in Figure 1.

The 'free' water adsorption without any confinement (according to DIN 18132 A) of the bulk bentonite averaged to 800 %, while its (more realistic due to partial confinement) water content in permeability tests (P-tests) under 25 kPa was only 180 %. (The selected load is equivalent to that of 1.30 m cover soil in German landfill cappings.) The calculated indexes of consistency corresponding to these more realistic water contents in P-tests amount from 0.70 to 0.73 (scaled up 1000 x in Fig. 1). These values are close to the limit of stiff consistency (= 0.75), toward which cohesive soils including bentonite tend to develop cracks, i. e. the bentonite is tending to a state which could be critical. Note also, that water content and index of consistency refer to a bentonite state of fully saturation in P-test under unlimited water supply, which is the 'best case' for low permeability. Such optimal conditions for a bentonite (GCL) as sealing element exist in situ only in water ponds, in all other GCL applications the water/ moisture supply fluctuates with the time (seasons), and could be insufficient for saturation, resulting definitely not in the 'best case' mentioned above, and enforcing critical state trends.

In the (saturated) P-tests following permittivities ψ , s⁻¹ (average from two samples) were recorded: Na-bentonite A: 2.1×10^{-9} s⁻¹; Na-bentonite B: 1.8×10^{-9} s⁻¹; Na-bentonite C: 2.5×10^{-9} s⁻¹. Water permeability is indicated herein as permittivity ψ considering the particular specimen thickness (Note: $\psi = k/t$, with k for the coefficient of permeability, m/s, and t for the thickness, m); in this way the conductivity of different materials can be compared directly.

In summary, the three Na-bentonites tested provided values for all basic parameters within the same close range (Fig. 1). The activated sodium bentonite B differed insignificantly from the natural bentonites A and C. It should be noted that no mineralogical investigations were undertaken.

Additionally, the permittivity of the pure bentonite B was compared with the permittivity of the stitch-bonded GCL it is being used for. Practically no difference was registered. (Average GCL permittivity is $\psi = 1.8 \times 10^{-9} \text{ s}^{-1}$ from QC test series under 30 kPa and 300 mm water head). It may be concluded that for the defined conditions the composition manufacturing technique (stitching) does not effect the permeability of this GCL.

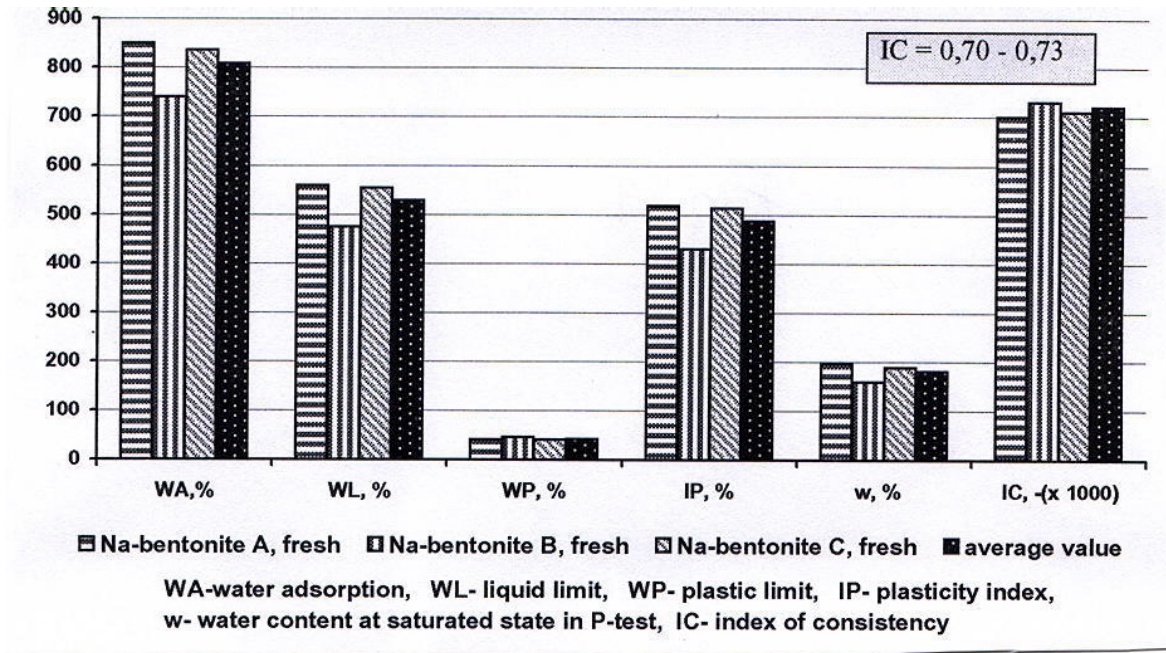


Figure 1. Parameters of 'virgin' Na-bentonites

3 EXPERIENCE FROM EXCAVATED MATERIALS

The changes in a sodium GCL after a certain period of use (in situ) become clear either when the function of the structure is visibly disturbed (e.g. empty water containment), or when GCL is deliberately dug up.

As early as 1993, Dobras, Elzea (1993) reported on the high permeability coefficient of $4 \times 10^{-7} \text{ m/s}$ of a GCL that had been used for containment sealing around fuel tanks in the USA. The indicated cause was ion transformation of the Na-bentonite by Ca- and Mg-ions, predominantly contained in the dolomitic limestone on the top. The cover soil was only 200 mm thick.

James et al. (1997) have described another case. Leaks were found in the GCL used to seal the vaulted ceilings of underground Victorian drinking-water reservoirs against atmospheric water. The ratio of Na+ to Ca++ in the excavated specimens was reversed; the bentonite layer exhibited cracks and had shrunk. The authors attribute the ion exchange to the calcium content of 2% in the fresh bentonite. At the same time they report that the quantity of soluble calcium in the cover soil exceeded by 7 to 8 times the quantity required for exchange of Na-ions in the bentonite. The authors also found that the deterioration in the function of the GCL was primarily a result of the shrinkage cracks, caused by transformation of the bentonite from Na- to Ca-form (cover soil 450 mm, of which 150 mm was Ca-free drainage gravel).

In 1996, the GCLs in the field trials at the Georgswerder landfill in Germany were excavated after increased flows through the GCLs were measured (Melchior 1996). Samples of two GCLs were recovered from instrumented fields and one was obtained from so called observation field.

The GCLs and the bentonites were subjected to very comprehensive examinations by four well-known German institutes. The main findings were that all GCLs were attacked more or less by Ca-ions and depending on the range of ion exchange shrinkage cracks have occurred, consequent to the extremely unfavourable site conditions. Detailed results and summarised assessment are compiled by Herold (1997). The research was used by the Deutsche Institut für Bautechnik (German Construction Engineering Institute) to approve in 1997-1998 GCLs for capping of German class I landfills. The approvals consider double-layer placed GCLs under strong regulations of the overall capping system to prevent any desiccation of the lower GCL sealing. This approach was based on the superior condition of the lower GCL layer at the overlaps at the Georgswerder site. Continued research by Melchior (1999) revises this state, discovering 1998 definite cracking in all of the samples from the bottom layer, too. The Georgswerder evidence gave rise to a thoroughly review and check on the existing GCL installation conditions.

Further excavations (Blümel, Brummermann 1998; Heerten et al. 1997; Maile et al. 1998) of GCLs at other sites in Germany, mainly landfills, were undertaken by manufacturers and officials to determine the state and performance of GCLs in different structures and under different conditions. The most recent evidence originates from the research on GCLs for groundwater protection at seven road sites (Ranis 1999).

The cases studied indicate:

- Presence of mostly Ca- and less Mg-ions in soil sufficient to remove the lightly bond sodium ions in Na-bentonite.
- Different degree of ion exchange takes place; in worst case original Na-bentonite-GCL is converted completely into a calcium one; both activated and natural sodium bentonite as well as chemically stabilised are affected.
- It takes only a few years till sodium bentonite undergoes ion-transformation, causing significant alteration of the initial bentonite and GCL properties.

The originally high water adsorption and swelling capacity diminish to levels that are typical of Ca-bentonites. Figure 2 indicates average values obtained on GCL excavated from four sites and compared with those fresh bentonites (scale for swelling volume 10 times enlarged). As a result of ion exchange the virgin bentonites have lost about two thirds of their initial free water adsorption and swelling capacity.

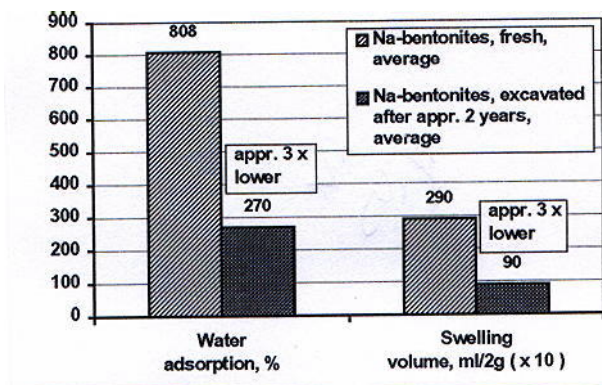


Figure 2. Average water adsorption and swelling volume of converted and virgin bentonites

In this connection, the determination of these parameter serves as a simple, indirect method of qualitatively ascertaining the ion exchange. The particular ion content (Na, Ca, K, Mg) of a bentonite can be determined quantitatively using other more complex methods.

The liquid limit is influenced by ion exchange, too. A reduction from originally approximately 500 % to about 170 % was recorded on the Georgswerder samples according to (Herold 1997). At the same time the plastic limit remains unchanged.

As the cation exchange capacity of the bentonite indicates the magnitude of negative charge of the montmorillonite crystals it keeps constant.

The ion exchange affects more or less the sealing properties of installed Na-GCL depending on whether, and if so to what extent the bentonite layer in the GCL has desiccated.

Permittivities obtained on GCLs excavated from five different sites are presented in Figure 3 and subdivided in group 2 and group 3 according to GCL installation depth of about 1.0 m (“deep”) for group 2 and from 0.20 to 0.40 m (“shallow”) for group 3. The recovered GCLs were examined after two years service use.

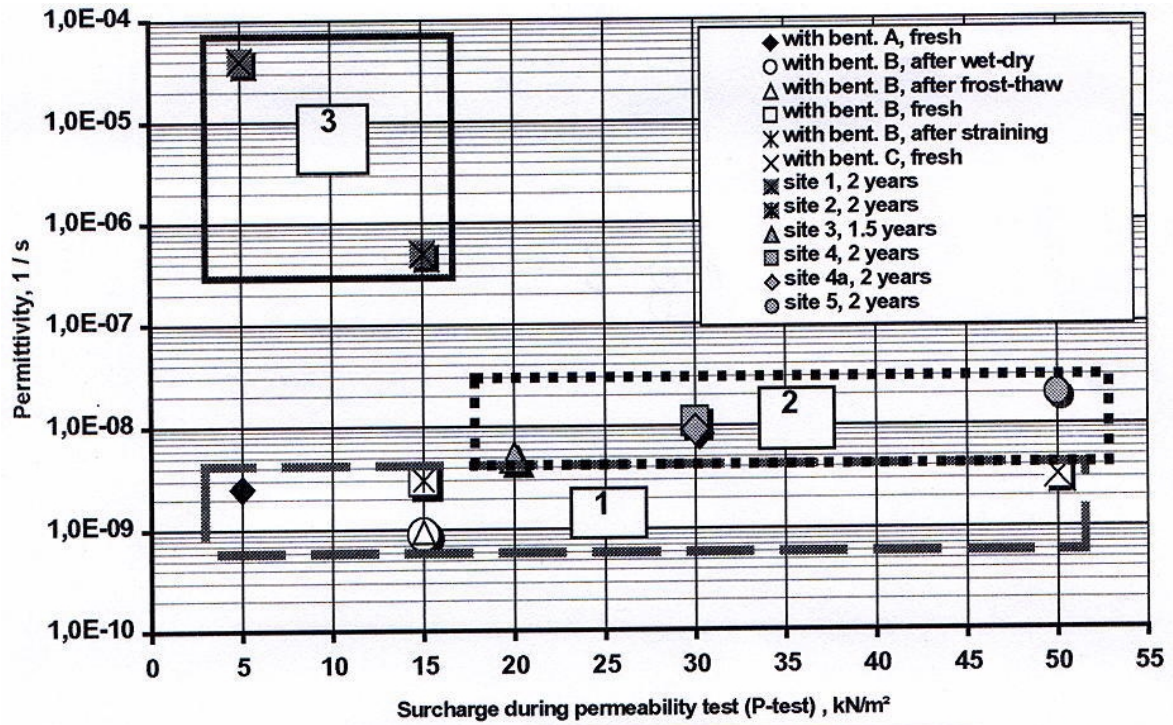


Figure 3. Permittivities of virgin and excavated sodium GCL

For comparison purposes permittivities of three factory-fresh Na-GCLs are indicated in group 1. This group compiles “simple” permittivities recorded under various normal stresses, as well as permittivities after submission to few dry-wet-, frost-thaw cycles or straining. Most important for group 1 is the fact, that permittivity of a virgin Na-GCL doesn’t increase after drying- or frost-impact, as the fresh bentonite possesses its full self-healing capability. Some negligible (3 to 4 times) permittivity variances herein result from the different test techniques and conditions.

The thinly covered GCLs from group 3 were subjected in site both to ion exchange and several dry-to-wet cycles at the same time. The thin soil covers have contributed the GCL-bentonite to desiccate getting shrinkage cracks, while ion exchange by Ca-ions has taken place making them irreversible. In group 3 the combination of drying and ion exchange results in an extreme increase in water permeability.

GCL-bentonites in group 2 have undergone some ion exchange but they’ve been protected from drying by the 1.0 m thick soil cover. X-ray photographs of these GCLs do not reveal cracks in the bentonite. At the same time, the thick cover provides a remarkable vertical pressure on the bentonite layer, which disables cracks to open. (This phenomenon is well known in geotechnics from

common clay layers.) The indicated permittivity deterioration is attributed only to the particular bentonite (ion) transformation. The pointed increase is about ten times (one order of magnitude) compared with the permittivity of fresh Na-GCLs in group 1. Comparison of group 2 (ion exchange) with group 3 (ion exchange plus desiccation cracks) leads to the conclusion, that drying of converted bentonites causes a permeability increase of approximately thousand times (three orders of magnitude).

Evidently the initial very low permittivity of fresh sodium-GCLs rises definitely with the times to different levels depending on the particular site conditions and influences.

4 GCL WITH CALCIUM BENTONITE

The need to develop and use a GCL with Ca-bentonite, as alternative to the conventional GCL with Na-bentonite, is based in short on the following fundamental evidence:

- Na-bentonites are converted in short terms to Ca-bentonites under ordinary conditions in situ;
- insufficient water supply increases the probability of especially unfavourable synergistic effects of ion exchange and desiccation on Na-bentonites;
- even a double-layer installation will not automatically prevent drying-out of the lower sealing

A characteristic of the newly developed GCL is the substantial bentonite content of 10,000 g/m² (two times more than in standard Na-GCL). The GCL bearing the trade name NaBento® is manufactured using sewing technique which makes the use of a heavy bentonite layer in this geocomposite possible.

The large content of Ca-bentonite is necessary in order to achieve a larger thickness of the sealing layer, required to compensate for the somewhat higher permeability of Ca-bentonites.

In order to determine the required parameters, a Ca-bentonite was investigated in advance similar to the Na-bentonites as described in section 2.

The properties established are summarised in Table 1 and compared with the average values of the Na-bentonites tested.

The free water adsorption of 307 % measured in the Ca-bentonite sample is in the upper limit of the range from 100 % to 300 % typically seen for Ca-bentonites. The bentonite water content during the P-test under vertical stress $\sigma = 25$ kPa was less than 100 %, which is one third of the free water adsorption of the Ca-bentonite. For comparison, in the P-test Na-bentonites had an average water content of 20 % of their free water absorption.

Swell heaving of 16 %, gauged in an oedometer testing device under $\sigma = 20$ kPa, is quite small compared with the average value of 64 % for Na-bentonites. It should be recognised, that the noticed volume expansion is related to the volume of shrinkage of a bentonite in a case of extreme desiccation.

Comparison of the consistency indices provides a very interesting result. Although both the Na-bentonites and the Ca-bentonite have a soft consistency in the P-test (in a saturated state), the Ca-bentonite with $I_C = 0.66$ is further away from the stiff consistency zone ($I_C \geq 0.75$) than the Na-bentonites at $I_C = 0.72$. This is in favour of the Ca-bentonites to the extent that in general, a typical 'soft' consistency is less likely to crack than a 'stiff' consistency.

The permittivity of the Ca-bentonite, tested on a layer with the defined mass per unit area of 4500 g/m², is almost three times higher than the average permittivity of the Na-bentonites.

Table 1. Comparison between Ca-bentonite and Na-bentonites in fresh state

	Standard	Ca-bentonite	Na-bentonites*
Mass per unit area (g/m ²)	DIN EN 965	4500	4500
Swell heaving (%)**	ASTM D 5890-95	16	64
Water adsorption (%)	DIN 18132 A	307	808
Liquid limit (%)	DIN 18122-1	190	530
Plastic limit (%)	DIN 18122-1	49	42
Plasticity index (%)	DIN 18122-1	141	488
Water content *** (%)	DIN 1812-1	96	180
Consistency index	DIN 18122-1	0.66	0.72
Permittivity *** (s ⁻¹)	DIN 18130-1	5.7 x 10 ⁻⁹	2.1x10 ⁻⁹

* Na-bentonite figures are average values for natural and activated materials.

** under $\sigma = 20$ kPa

*** P-test, under $\sigma = 25$ kPa

The average properties of the Ca-bentonite, currently applied in the Ca-GCL are:

Swelling volume	11 ml / 2g
Water adsorption	220 %
Liquid limit, W_L	173 %
Plasticity index, I_p	133 %
Water content (in the P- test)	92 %
Consistency index, I_c	0.61

The Ca-GCL was subjected to the usual permeability evaluations, simulating various conditions in the laboratory. The following permittivities (ψ) were determined in rigid-wall permeability cells under a vertical pressure $\sigma = 15$ kPa and a water head of $\Delta h = 300$ mm:

$\psi = 6.10^{-9} \text{ s}^{-1}$ after 5 dry-wet cycles (drying to constant weight)

$\psi = 3.10^{-9} \text{ s}^{-1}$ after 3 frost-thaw cycles

$\psi = 4.5.10^{-9} \text{ s}^{-1}$ under 10% straining in hydrated state, $\sigma = 30$ kPa

$\psi = 8.3.10^{-9} \text{ s}^{-1}$ under $\sigma = 5$ kPa, $\Delta h = 300$ mm

In order to test whether the theoretical stability of the Ca-bentonite opposite Ca-ions is in fact true, trials (LGA Bayern, unpublished) with CaCl₂-solutions were carried out on specimen of the Ca-GCL and examined for permeability. For comparison purposes a Na-GCL was tested too.

The Ca-GCL specimen was placed directly in a tank containing an aqueous solution of 200 mg CaCl₂ / 1 H₂O and was surcharged to $\sigma = 15$ kPa for 18 days. In the consequent conductivity test, carried out in a rigid-wall-permeameter under $\sigma = 15$ kPa and $\Delta h = 300$ mm, the sample was permeated with the same CaCl₂ solution for 25 days. Within the first few days the permittivity settled at $\psi = 4.4x10^{-9} \text{ s}^{-1}$, reducing continuously to $\psi = 3.3x10^{-9} \text{ s}^{-1}$ during the next 15 days, after which the value remained constant. At this test-stage the permittivity of the Ca-GCL was not influenced by the ionic solution employed. Then the test was continued applying a much stronger solution of 80 g CaCl₂ / 1 H₂O for a further 14 days. The permittivity increased rapidly up to ten times, and then showed no further change. According to the bentonite supplier, the increased permittivity of the Ca-GCL specimen is attributed to highly concentrated electrolyte solution, which reduces the expansion of the diffuse ionic layers and reduces the swelling potential. The electric conductivity of the strong solution was 100,000 $\mu\text{S} / \text{cm}$, while that of the lower concentrated solution was 440 $\mu\text{S} / \text{cm}$. The negative effect of concentrated electrolyte solutions on bentonites is explained theoretically in (Gleason, M. et al. 1997); tests on the bentonite component of a Na-GCL are outlined in (Quaranta, J. et al.).

A similar approach was adopted for the Na-GCL specimen, except that it was pre-hydrated with de-ionised water for 3 days under load $\sigma = 15$ kPa. After that it was stored in the strong solution of 80 g $\text{CaCl}_2 / 1 \text{ H}_2\text{O}$ for 30 days under the same load. In the following permeability test, it was permeated for 11 days with pure water instead of ionic solution. A permittivity $\psi = 2.10^{-7} \text{ s}^{-1}$ was initially recorded, which remained constant until the end of the test. Although the Ca-GCL was examined under more unfavourable conditions, as it wasn't hydrated prior to the contact with the ionic solution, it remained less permeable (about 10 times) than the Na-GCL tested. (Gleason et al. 1997) have carried out similar investigations, on equally thick 10 mm-layers of two Na-bentonites and one Ca-bentonite, indicating similar results.

In order to examine the behaviour of the Ca-GCL under conditions of dehydration, which probably may occur in situ, a simple index test was carried out. A sample of Ca-GCL, previously saturated under vertical pressure $\sigma = 2.5$ kPa was exposed to drying in an oven with a circulating air flow of 30° C . The same low pressure was applied during drying as well. The bentonite water content was recorded by weight in regular time intervals during the drying process. The same procedure was followed simultaneously for a sample of a Na-GCL. The change of the bentonite water content of both Ca- and Na-GCL samples during dehydration are shown in Figure 4. In the dangerous crack-forming-zone of about 100 % water content the Ca-GCL released moisture much more slowly than the Na-GCL.

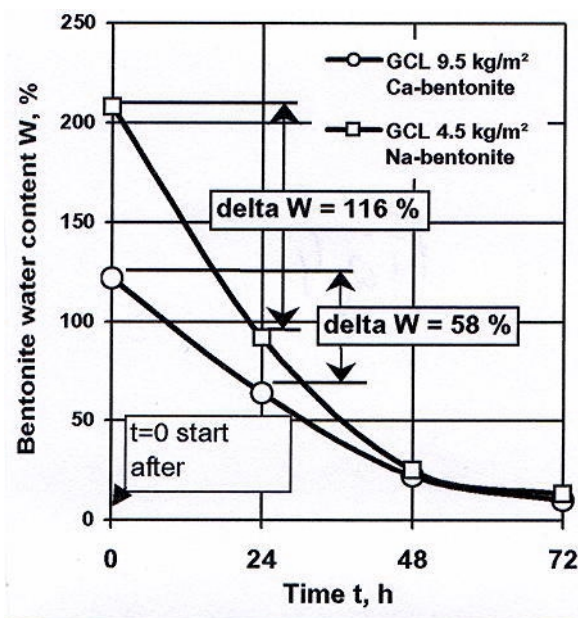


Figure 4. Change in water content of bentonite in Ca-GCL and Na-GCL during laboratory drying at $T = 30^\circ \text{ C}$, $\sigma = 2.5$ kPa

The performance of the Ca-GCL in the tests conducted to date is regarded as positive. Measurements and observations in situ on three locations have also been in progress for a few years. The results of these field-trials up to date confirm this favourable assessment. Further monitoring will continue over the next years.

5 SUMMARY

Experience with Na-GCLs to date has shown increases of initially very low permeabilities in various applications.

It has been found that mainly due to the presence of calcium ions in most soils, the Na-bentonites are partially or fully transformed into Ca-bentonites. The process can take place quite quickly (in two to three years). As a result, the water adsorption capacity, the swelling capacity and the liquid limit of Na-bentonites reduce to values typical for Ca-bentonites. Water permeability typically increases by one order of magnitude (approximately up to 10 times).

Additionally, synergistic effects of ion exchange, drying out and the development of cracks may take place. Drying is caused by too thin soil cover layers and/or layers of soils with poor water retention. In such “synergistic” cases, the water permeability of the GCLs may increase by several orders of magnitude (up to 1000 times in some critical cases registered). The general problem is that transformed bentonite (from Na- as installed to Ca- with reduced swelling capacity) is not able to close the cracks any more. The cracking process, which is reversible in Na-GCLs, becomes irreversible after the ion exchange has taken place.

Consequently, appropriate measures are very important to prevent Na-GCLs from desiccation: deep installation of the Na-GCL (over 1 m deep) ; covering by soil layers with high moisture retention capability, suitable for the actual location, climatic conditions etc. Drying out accompanied by crack development is not likely if the Na-GCL is permanently in contact with saturated soil.

Note, that in any case the increase in permeability as a result of ion exchange will take place.

The use of Ca-bentonite is regarded as providing the prospect of a more reliable application for GCLs.

Ca-bentonite neutralizes the risk and the negative effects of ion exchange. When installing Ca-GCLs in normal soils, the bentonite properties do not change while being exposed to Ca-ions. The particularly unfavourable synergistic effect of ion exchange and drying out cannot occur in Ca-GCLs.

Due to lower water adsorption capacity and lower water content, while the Ca-bentonite still has a favourable, soft consistency, the Ca-GCL needs less water for swelling and is less sensitive to a reduction in water content than a Na-GCL. The lower swelling capacity results in a reduced tendency to shrinkage and therefore less development of cracks when drying out when compared with Na-bentonites.

However, the bentonite mass per m² in a Ca-GCL must be increased in order to gain the same low permittivity as in the case of Na-GCLs. In the sewn Ca-GCL (NaBento®) presented in this paper, this is realized by providing a mass per unit area of 10,000 g/m² Ca-bentonite. The large bentonite mass also provides a greater robustness than with conventional GCLs.

In summary the Ca-GCL will not show any significant changes in permittivity while in use, which can not be stated for Na-GCLs. Three different field tests set up about three years ago have confirmed these statements.

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