EFFECT OF DRY/WET CYCLES AND CATION EXCHANGE ON THE PERMEABILITY OF A DENSE PREHYDRATED GCL

Francesco Mazzieri¹ & Erio Pasqualini²

¹ Technical University of Marche Ancona Italy. (e-mail: f.mazzieri@univpm.it)

² Technical University of Marche Ancona Italy. (e-mail: e.pasqualini @univpm.it)

Abstract: Cover systems for waste landfills often contain Geosynthetic Clay Liners (GCLs) as a hydraulic barrier. GCLs offers several advantages over more traditional compacted clay liners. However, recent experience has shown that in the long term, GCLs in caps and covers are prone to desiccation and cation exchange, which may lead to failure of the hydraulic barrier. Moreover, potential for desiccation of GCLs has been recognized also in base liners of MSW landfills due to the heat produced by waste decomposition. New products have been proposed to overcome the limitations of conventional GCLs. The paper concerns the evaluation of a dense, prehydrated (DPH) and polymer amended GCL subjected to dry/wet cycling. Distilled water (DW) and 0.0125 M CaCl₂ have been used as hydrating solutions. Severe desiccation was simulated by dehydrating the DPH GCL at 35 °C. Permeability tests with distilled water show a slight increase of hydraulic conductivity with dry/wet cycling (less than one order of magnitude). Results with 0.0125 M CaCl₂ show an increase in hydraulic conductivity of up to three orders of magnitude, probably as a consequence of cation exchange and reduction of self-healing properties of bentonite. The results indicate that, as conventional GCLs, DPH GCLs should be adequately protected (e.g. by thick surface layers) in caps and covers where severe desiccation and cation exchange are likely to occur.

Keywords:. Geosynthetic clay liner (GCL), cover, chemical compatibility, drying, cracking, permeability

INTRODUCTION

Geosynthetic Clay Liners, (GCLs), that consist of a thin layer of bentonite sandwiched between two geotextiles or glued to a geomembrane, are increasingly used as hydraulic barriers in landfills covers and liners as well as in remediation projects for contaminated sites. GCLs offer several advantages over traditional compacted clay liners, among others ease of installation, limited thickness and low permeability (Bouazza, 2001). The hydraulic performances of GCLs that do not comprise a geomembrane depend on the low hydraulic conductivity of sodium bentonite when permeated with dilute solutions (e.g. distilled water or tap water). Unfortunately, sodium bentonite is prone to cation exchange under typical field conditions, since multivalent cations (e.g. Ca²⁺,Mg²⁺) are dominant in pore water of most soils used as covering layers, with the rare exception of sodic soils. Cation exchange drastically alter the swelling properties and plasticity of bentonite (Rhul and Daniel,1997; Shackelford et al. 2000; Jo et al. 2005) and consequently the self-healing properties (Mazzieri and Pasqualini; 2000); however, as long as the barrier remains saturated, an increase in hydraulic conductivity of at most one order of magnitude is to be expected for a calcium exchanged bentonite (Egloffstein 2001).

Several laboratory studies and reported case histories have suggested that the hydraulic conductivity of GCLs can increase of several orders of magnitude if cation exchange is combined with desiccation and cracking of the bentonite (Melchior 1997; Lin and Benson, 2000). Typical applications where such circumstances may occur are landfill covers systems, as a result of seasonal fluctuations of temperature and moisture content (Meer and Benson, 2007). Potential for desiccation of the GCL has been identified also in bottom liner of MSW landfills due to the heat produced by waste decomposition (Southen and Rowe, 2005).

One possible solution to the above mentioned limitations of conventional GCL is to utilize engineered bentonites that are able to perform better under unfavourable conditions. Dense Prehydrated (DPH) GCL have been proposed recently as an alternative to conventional GCLs. A few studies in the literature have addressed the hydraulic performance of DPH (Shroeder, 2001; Kolstad et al., 2004; Katsumi et al., 2008). These studies have demonstrated that DPH GCLs preserve much lower permeability than conventional GCLs in presence of various chemical solutions as long as they remain saturated. Since DPH GCLs are commonly recommended in cover systems and landscaping applications, they can be exposed to dry/wet cycles as wells contact with salts leached from adjacent soils. Furthermore, due to their increased compatibility to aggressive permeants, applications in landfill liners are also particularly appealing. Hence, DPH GCLs may be potentially exposed to the same environmental conditions that have been shown to impair the efficiency of conventional GCLs. The primary purpose of this paper is to investigate in the laboratory the response of a DPH GCL to dry/wet cycling. Secondly, the effect of coupling desiccation with cation exchange is examined.

MATERIALS AND METHODS

Dense Prehydrated GCL

Dense Prehydrated GCL (DPH GCL) have been patentend and introduced on the market in the mid 90s. The principle of the material is to combine the well known benefits of prehydration and densification on the hydraulic conductivity of the Na-bentonite. During production of DPH GCL; sodium bentonite clay is mixed with a hydrating solution in a high speed, high shear mixer; this mass is then calendered under vacuum into a bentonite sheet with a low

void ratio (e=1.5). The bentonite sheet is then sandwiched between one high strength woven polypropylene geotextile and a perforated polyester scrim geotextile.

Densification reduces the volume of voids by replacing pore space with solids. Prehydration with a dilute solution (e.g. deionized water or tap water) renders bentonite more resistant to a given aggressive permeant than nonprehydrated bentonite (Shackelford et al., 2000). When sodium bentonite is hydrated with a dilute solution, swelling occurs as water molecules initially hydrate the clay surface ("crystalline" phase) and are subsequently attracted to the interlayer ("osmotic" phase). Most of the interlayer water is bound or "immobile" water, that does not take part to hydraulic flow (Mc Bride 1984). Thus, the hydraulic conductivity of bentonite is directly correlated to the fraction of bound water in the interlayer. Factors that affect the fraction of bound water directly affect the hydraulic conductivity of bentonite (Jo et al., 2004). If the hydrating liquid is a strong electrolyte solution or multivalent cations occupy the exchange sites (e.g. as a result of cation exchange), swell is confined to the "crystalline" phase, the fraction of bound water is lower and the hydraulic conductivity is higher.

Besides the benefit of prehydration, the hydrating solution of DPH GCLs contain organic molecules that are intended to improve the workability of the mixture during production as well as the resistance of bentonite to chemical interaction in the presence of aggressive permeants. The solution contains the polymer sodium carboxymethyl cellulose (CMC) and methanol. It is believed that large polymer molecules as CMC bind to the solid surfaces thus keeping the interlayer open (Kolstad et al:, 2004) even in presence of factors that would collapse the interlayer (e.g. increase of ionic concentrations and/or pore space invaded by multivalent ions).

Qiu and Yu (2007) showed that CMC intercalated in the interlayer of a sodium montmorillonite. The basal d-spacing increased from 1.25 nm to 1.47 nm upon addition of CMC to montmorillonite at a CMC to clay weight ratio of 2. The weight ratio of CMC to bentonite in DPH bentonite is of the order of 0.1 in the typical product (Flynn and Carter,1998). Shroeder et al (2001) performed random powder XRD analysis of dried DPH bentonite and found that the basal spacing of the was 1.47 nm compared to 1.25 nm that would have been expected for a sodium montomorillonite. They concluded that the additive had probably replaced Na⁺ cations in the interlayer. In addition to expanding the interlayer, CMC provides fungicidal and preserving action.

Possible variations to the basic "recipe" of the prehydrating polymer solution include sodium polyacrylate (PAAS) and sodium hexametaphosphate (Flynn and Carter 1998). Adsorption of polyacrylate onto montomorillonite is expected to be small, at most 1.5 mg/g (Siffert and Espinasse, 1980). However, Qui and Yu (2008) showed that in CMC-modified montmorillonite expanded interlayer make acyrlate molecules enter more easily into interlayer "galleries" for *in situ* polymerisation. Coating of the solid surfaces with acrylate molecules strongly bound in their sodium form to the mineral surface is aimed at preventing conversion of treated bentonite into a calcium form that would reduce the swelling and self-healing performance of the DPH in case of dehydration (Flynn and Carter, 1998).

Table 1 summarises the main properties of the analysed material. Literature data refer to products described as similar to the DPH used in this study, that presumably belong to the same family of products. The actual composition of the hydrating solution of the used DHP GCL is not known to the writers. XRD analysis and FTIR spectroscopy were performed on-air dried bentonite retrieved from DPH GCL to gather information on the solid structure and composition. XRD diffraction showed a peak at $2\theta = 6.30^{\circ}$, that indicates a basal d-spacing of 1.40 nm, in accordance with the findings by Shroeder et al. (2001). The FTIR spectrum of air dried bentonite revealed weak but visible adsorption bands that can be ascribed to CMC. The swell index of dried bentonite in DW as per ASTM D 5890 was 16 ml/2g, that is much less than it is normally obtained for conventional natural or activated sodium bentonites in DW (\approx 30 ml/2g; Miles, 2002). However, a clear separation was not observed between the bentonite and the liquid, that remained turbid probably as a result of polymers' dispersing action. Finally, the specific gravity of solids Gs of bentonite as per ASTM D854 was 2.56, that is lower than the range commonly reported for conventional bentonites (2.60-2.70). A specific gravity lower than untreated bentonite is consistent with binding of organic molecules to the clay surface (Mc Rory and Ashmawy,2003; Kolstad et al., 2004).

Property	Value	Source
Dry mass of bentonite (ASTM	5.43 kg/m^2	-
D5993)		
Average core (bentonite) thickness	4.9 mm	-
Water absorption (ASTM D5890)	550%	Specification sheet
Liquid limit (JIS A 1205)	450%	Katsumi et al.
		2008
Specific gravity, Gs (ASTM D854)	2.56	-
W% (ASTM D2216)	44.8%	-
Montmorillonite content (XRD)	89%	Kolstad et al. 2004
pH (1:5 extract)	8.21	-
EC (1:5 extract)	3580 mS/cm	-
CEC	51.8	-
	meq/100g	

Table 1. Physical and chemical properties of DPH GCL used in this study

The Cation Exchange Capacity (CEC) was measured on the dried bentonite by the compulsive method (Rhoades et al. 1996). Major exchanged cations are Na⁺ (77. 71 meq/100 g), Ca²⁺ (12.43 meq/100), Mg ²⁺ (8.57 meq/100 g), K⁺ (2.39 meq/100 g), where deduction has been made for soluble cations, which were measured on a 1:5 soil to DW extract. Further details on the procedure are given in Di Emidio et al. (2008). The measured CEC is less than it would expected for common sodium bentonites (60-90 meq/100g; Egloffstein, 2001), which might partly reflect the protective action of polymer coating against cation exchange. Moreover, exchanged Na⁺ alone exceeds the CEC. This is probably a result of release of Na-bearing molecules (e.g. CMC) coating the clay particles, with only a fraction of Na-ions effectively bound to the surface and contributing to saturate the CEC:

Hydrating Liquids

Distilled water (DW) was used as a reference to investigate the swelling and permeability of the DPH bentonite subjected to dry/wet cycles. Soil pore water was represented by a 0.0125 M CaCl_2 solution (corresponding to 500 mg/L Ca²⁺), based on the study by Lin and Benson (2000). The Ca²⁺ concentrations in common soils' pore water is expected to be lower (40-160 mg/L Ca²⁺). Concentrations of up to 600 mg/L are possible in case of soils containing gypsum (Egloffstein, 2001). In the present study the 0.0125 M CaCl₂ solutions was selected mainly in order to compare the results with previous investigations. Moreover, a 0.010 M CaCl₂ solution is recommended in ASTM D5084 as standard water in zones with hard tap water. DW had EC=0.014 mS/cm and pH=6.8. The CaCl₂ solution was prepared by dissolving the appropriate amount of analytical grade hydrated salts in DW. The salt solution had Electrical Conductivity (EC)= 2.85 mS/cm and pH= 6.56.

Permeability tests with dry/wet cycles

Test specimens 10.16 cm (4'') in diameter were obtained from the DPH roll by means of a circular template and of a sharp cutter. The putty-like nature of the clay prevented loss of clay in preparation of test specimens.

Two specimens were permeated directly with the test solutions without any previous treatment, with the purpose to establish the baseline hydraulic conductivity. Permeability test were performed in flexible wall permeameters. The specimen were allowed to equilibrate inside the permeameter without gradient for about two weeks. It is to be noted that although bentonite in DPH is prehydrated, further swell and water absorption do occur upon contact with the test solutions particularly at low effective stress. The average applied effective stress was 15 kPa during hydration and 12.5 kPa during permeation. Tests were of the variable head type with the applied head ranging from 100 to 130 cm.

Previous experience with long-term permeability tests of DPH GCL with DW (Di Emidio et al., 2008) revealed that gas bubbles develop in drainage lines, probably as a result of biological activity. In the attempt to minimize gas production, DW was spiked with 500 mg/L of the antimicrobial Dowicil [®]Qk-20.Addition of the biocide to DW changed only slightly the chemical parameters of DW (EC= 0.078 mS/cm and pH =6.92); moreover, the biocide was not expected to interact chemically with the clay (Jo et al., 2005). Gas bubbles did not develop significantly in the test with CaCl₂ solution. Termination criteria stated in ASTM D6766 were generally followed.

Two specimens were subjected to dry/wet cycles alternated with permeability test after each wetting. The sequence started with the first wetting, performed in an oedometer cell at vertical pressure 12.5 kPa. The cell was inundated with the desired liquid (DW or 0.0125 M CaCl₂) and the displacements were monitored until equilibrium. The cell liquid was monitored for Electrical Conductivity and pH. Changes occurred over time as a result of evaporation form the upper surface of the oedometer cell and/or release of soluble and exchanged cations from the soil matrix into the pore solution. In the attempt to minimize changes in the cell fluid chemistry and particularly in the Ca²⁺ concentration, the cell fluid was drained off and completely replaced with fresh solutions every six or seven days. The hydraulic conductivity at the end of the first wetting was not measured but assumed to be equal to the reference hydraulic conductivity. After the 1st wetting, a dehydration phase (drying) was carried out.

Dehydration was performed in a thermostatic chamber at constant temperature of 35 °C and relative humidity ranging between 20 % and 40 %. A vertical pressure of 6 kPa was applied during the drying phase by a dead load with the purpose to simulate overburden pressure. It is to be noted that the vertical pressure acting in typical applications (e.g. covers) is expected to be at least 15-20 kPa. In order to monitor mass loss, the specimen was momentarily unloaded and weighed. The drying phase was considered completed when mass loss ceased. On average, constant mass was achieved within 20-30 days.

Rehydration was then performed in the oedometer cell. The vertical pressure was maintained at 6 kPa. At the end of the rehydration phase, the specimen was transferred in the permeameter for hydraulic conductivity testing. An average effective stress of 12.5 kPa was applied during permeation. In total, four wetting cycles were carried out.

RESULTS AND DISCUSSION

Hydraulic conductivity of as-received DPH GCL

The hydraulic conductivity of DPH GCL to DW and 0.0125 M CaCl₂ are illustrated in Figure 1a. Results refer to about 400 days and 580 days of continuous permeation with the DW and CaCl₂ solutions respectively. Test durations corresponds to 2.98 pore volumes of flow and 4.89 pore volumes of flow respectively. The termination criteria stated in ASTM 6676 were generally met with the exception of the EC effluent-to-influent ratio, that remained out of the acceptable zone (1.0 ± 0.1) for both tests (Fig. 1b).

The average hydraulic conductivity (k) to DW was 8.8×10^{-10} cm/s. The hydraulic conductivity to 0.0125 M CaCl₂ displayed a somewhat larger scatter; the average value of the last 60 days of permeation was 8.1×10^{-10} cm/s. Therefore, the permeability of DPH to either liquid was very close. The k value for DW is slightly higher than the value k= 4.1×10^{-10} cm/s obtained by Kolstad et al. (2004) on a similar DPH product with deionized water, probably as a result of the lower average effective stress applied in the present study during permeation (12.5 kPs vs. 20 kPa).



Figure 1. a) Hydraulic conductivity of DPH GCL to DW and CaCl₂ solution, b) Effluent-to influent EC ratio

The effluent-to-influent EC ratio was very high (up to 80) during permeation with DW (see Figure 1b). This result is not unusual when GCLs are permeated with deionized water or distilled water (Jo et al., 2005): it reflects both the low EC of the influent solution (the denominator of the ratio) and the washing-out of soluble salts from the bentonite. In the case of DPH GCL, part of the unbound polymeric additive is probably also flushed into the pore water (CMC is a water-soluble polymer). No specific analyses of the effluent were performed to confirm the presence of organic molecules. However, the samples of the effluent liquid often presented a sort of filamentous deposit that could be ascribed to polymer molecules. In any case, long term interactions are not expected for DW, therefore the hydraulic conductivity was not expected to change despite the fact that the effluent-to-influent EC ratio remained high (8.5).

The effluent-to-influent EC ratio with the 0.0125 CaCl₂ solution was lower than with DW. However, in absolute terms the peak EC of the effluent (14.6 mS/cm) was much higher than the peak EC of the effluent (6 mS/cm) with DW. This result can be explained by the higher EC of the influent solution (2.85 mS/cm), by the washing-out of soluble salts but also by the release of exchanged Na⁺ ions. The effluent-to-influent ratio after 560 days of permeation was 1.63. However, although chemical equilibrium was not yet achieved, further changes in hydraulic conductivity were not to be expected. Kolstad et al. (2004) permeated a DPH GCL with 1 M CaCl₂ solution and found hydraulic conductivity of 4.2 x10⁻¹⁰ cm/s. Katsumi et al. (2008) permeated a DPH GCL with 0.25 M. 0.5 M and 1 M CaCl₂ solutions for test duration up to 4 years. The hydraulic conductivity values were of the order of $1x10^{-10}$ cm/s regardless of the CaCl₂ concentration used. In essence, these studies demonstrated that, as long as they remain saturated, DPH GCL exhibit extremely low hydraulic conductivity even to strong electrolyte solutions of multivalent cations. The increased performance can be ascribed to the combination of prehydration, densification and chemical amendment, although according to Kolstad et al.(2004), prehydration is by far the predominant factor. It is worth mentioning that a

nonprehydrated conventional GCL permeated with 0.0125 M CaCl₂ had hydraulic conductivity of 1×10^{-8} cm/sec after about 600 days of permeation (Benson et al., 2007).

Dry/wet cycles

The impact of dry/wet cycles on the swelling behaviour of DPH GCL can be examined in Figure 2. The vertical swell vs. time curves during the various wetting cycles are reported. During the first wetting, a slightly larger swell was observed for the CaCl₂ solution than for DW. However, for the CaCl₂ solution, the swell started to decline after about 21 days whereas it remained relatively constant for DW. The swell stabilized in about 65 days. The difference in the swell behaviour can be ascribed to the presence of Ca^{2+} cations, that invaded gradually the bentonite pore water via diffusion and advection, in response to moisture and osmotic gradients.



Figure 2. Swelling of DPH GCL with DW and 0.0125 M CaCl₂ during dry/wet cycling

Bouazza et at. (2007) pointed out that in studies where the effect of cation exhange is to be investigated, care should been exercised in evaluating the degree of exchange that is actually achieved at each stage of testing. To do so, masses of solids of the chemical solutions should be measured so that the solid-to-liquid ratio can be determined precisely. Moreover, the concentration of relevant cations (Na⁺ and Ca²⁺) should be measured at each stage, since the exchange equilibrium depends on the concentrations and the ionic strength of the solution. This type of measurement were not carried out during the test. In any case, the evaluation of exchange equilibrium would require knowledge of Ca/Na selectivity coefficients (Appelo and Postma; 1999) for polymer amended bentonite, that may not necessarily be the same as untreated bentonite.

The fraction of CEC of DPH bentonite not occupied by calcium is from Table 1 equal to (51.3 - 12.5) meq/100 g= 38.8 meq/100 g; from the initial dry mass of the DPH specimen (44.42 g), one can calculate that at least 17.23 meq of Ca $^{2+}$ must be supplied to replace the exchangeable cations. The equivalent concentration of the 0.0125 M CaCl₂ solution is 25 meq/L. Therefore, at least 0.680 L of solution must have been added to the bentonite. Although precise account of the amount of solution added was not taken, it is noted that the cell liquid was replaced several times during the test, each time approximately 0.35 L were drained from the cell and replaced with fresh solution, corresponding to the addition of 8.75 meq Ca^{2+} . Although accurate evaluation of the total Ca^{2+} taken up by the bentonite would required knowledge of the concentration just before replacing the cell liquid with fresh solution, it is believed that enough Ca^{2+} was supplied to potentially saturate the CEC of DPH with Ca^{2+} . Furthermore, replacement with fresh solution removed the exchanged Na^+ released into the pore solution, so that the system was essentially open with respect to Na^+ (Bouazza et al. 2007). In any case, the contraction of the specimen at the later stage of the test (>21 days) is consistent with a cation exchange in the saturated clay. Replacement of monovalent Na⁺ by divalent ions reduces the fraction of bound water, that is partly expelled from the interlayer. However, when cation exchange occurs in a saturated clay, the clay retains more bound water that it would retain with the same cation population on the exchange site, but starting from a dry state. To extract more water molecules from the interlayer, osmotic gradients must overcome the adsorptive forces that bind water to the clay. As long as the clay remain saturated, adsorptive forces appear predominant, unless additional energy (e.g. heat) is supplied (Jo et a., 2004).

Drying resulted in severe desiccation of bentonite. The residual water content at the end of the drying phase was estimated assuming that the initial dry mass of solids and geotextiles had remained constant. Gravimetric water contents of the order of $5 \div 10$ % were estimated. The authors recognize that such extreme desiccation are unlikely to be observed in the field. For instance, Meer and Benson (2007) tested GCL samples exhumed from four landfill covers and found a minimum water content of 30.9 %. Southen and Rowe (2005) simulated desiccation of GCLs beneath a geomembrane as a result of thermal gradients. They found minimum gravimetric water contents of the order of 20%. Therefore, test results described herein are to be intended as the response to a given set of applied conditions more than representative of actual site conditions.

Despite the heavy desiccation, test specimens did not initially crack, but rather shrank volumetrically. This depends mainly from the absence of lateral clamping during dehydration. Dried bentonite formed a monolithic and relatively hard disk. Thorough mixing during factory prehydration and vacuum extrusion of DPH GCL probably confer high density and homogeneity to the bentonite core.

After the first dehydration, test specimens were installed in the oedometer and rewetted (Figure 2). The final swell with DW was much less than in the 1st wetting (2.5 mm vs. 5.9 mm). This was not expected, and even more so since the vertical stress during the second wetting was 6 kPa while it had been 12.5 kPa during the 1st. However, in the following cycle, the final swell was 6.5 mm. The authors suspect that an unidentified technical problem occurred during the second wetting phase with DW, that prevented the specimen from swelling. In any case, test results suggest a reduction of swelling power with dry/wet cycles since the final swell at the 4th wetting was only of 2.2 mm. A possible explanation is that drying caused volumetric shrinkage, so that the diameter of the dried specimens was usually $8\div10\%$. less than the initial diameter. Therefore, the specimen had to swell both vertically and radially upon rewetting. Secondly, permeability tests were performed at the end of the 1st, 2nd and 3rd wetting cycles. Permeability testing required consolidation of the specimen to a higher effective stress (12.5 kPa). The various manipulations of the test specimen might have caused disturbance as well as loss of bentonite, which is particularly sticky when wet. In this respect, it is noted that at the end of the 3rd drying, the mass of the specimen was 37.41 g, that is significantly less than the initial calculated dry mass of solids (45.93 g). It is evident that some loss of solids (and polymers) must have occurred.

At the end of the 3rd drying phase, a visible crack formed on the specimen (marked by the dotted line in Figure 3a). Incidentally, the whitish stains on the bentonite surface visible in the picture are fragments of the desiccated filter paper remained attached to the clay. The perforated polyester scrim geotextile had deteriorated and had been previously removed. The crack was apparently shut upon rewetting (Figure 3b).

The final swell in the 2^{nd} wetting for the CaCl₂ solution was less (3.39 mm) than in the 1^{st} (5.0 mm), despite the lower vertical stress applied. This was somewhat expected and confirmed that cation exchange had probably occurred during the first wetting. Moreover, the final vertical swell decreased monotonically during the following cycles. Besides the impact of cation exchange, similar considerations as a to the reasons for the reduced swelling power of bentonite can be made. A significant mass reduction was observed also with this specimen: the estimated initial dry mass of solids was 44.42 g while the mass at the end of the 3^{rd} drying was of 40.32 g. Thin fractures appeared on the DPH at the end of the 2nd drying. A large crack formed at the end of the 3rd drying (marked by a dotted line in Figure 3c), that appeared to shut close upon rehydration (Figure 3d).



Figure 3. Aspect of DPH GLC specimens in: **a**) DW-end of 3rd drying; **b**) DW-end of 4th wetting; **c**) 0.0125 M CaCl₂-end of 3rd drying; **d**) 0.0125 M CaCl₂-end of 4th wetting

Hydraulic conductivity

The hydraulic conductivity of the DPH test specimens at the end of each wetting cycle is illustrated in Figure 4. The values plotted for the 1st wetting are hydraulic conductivity values of the as-received DPH GCL. The hydraulic conductivity to DW water increased slightly from 8.8×10^{-10} cm/s to 2.8×10^{-9} cm/s. The modest increase is to be ascribed more to the disturbance induced by the testing method than to the impact of dry/wet cycling itself. Careful inspection of the test specimen was performed after testing to check for cracks or fissures but no visible defects were found. Furthermore, unhealed defects usually lead to hydraulic conductivities orders of magnitude higher. The clay mass appeared intact and homogeneously hydrated.



Figure 4. Hydraulic conductivity at each wetting cycle

A small portion of the specimen was used for a free swell test as per ASTM D 5890. Interestingly, the result was 28 ml/2g against 16 ml/2g of as-received bentonite. The bentonite settling pattern was very similar to that observed for standard Na-bentonite, with significantly less turbidity than with the as-received bentonite. In essence, dry/wet cycling and repeated permeability testing seem to have removed the polymers, converting the DPH bentonite back into a standard Na-bentonite. This interpretation is also consistent with the mass loss of the specimen.

The hydraulic conductivity with the calcium chloride solution remained relatively low $(6.8 \times 10^{-9} \text{ cm/s})$ only after the 2nd wetting. Thereafter, the hydraulic conductivity increased dramatically to 5.5 $\times 10^{-5} \text{ cm/s}$. This values are typically encountered with bentonite that contain cracks or defects that are not healed upon rehydration (Lin and Benson, 2000). The hydraulic conductivity remained essentially unaltered after the 3rd wetting.

Concern was that mass loss had interested mainly the edge of the specimen, so that sidewall leakage might have occurred during testing. In order to detect preferential flow paths, the permeant was spiked with the anionic tracer methyl orange, but no conclusive evidence of preferential leakage along the edge was found. The specimen coloured rather uniformly. Microcracks invisible to the naked eye had probably formed throughout, that were responsible for the high hydraulic conductivity.

A small portion of the bentonite was dried and used for a free swell test (ASTM D 5890). The result was 9 ml/2g. The settling pattern and the value of free swell were typical of calcium bentonites (Miles, 2001). It is possible to conclude that bentonite has been converted into a calcium exchanged form as a result of displacement of Na^+ ions initially occupying the exchange sites, free or linked to the coating polymers.

The obtained result are very similar to those described by Lin and Benson (2000) with regard to a conventional GCL. For the sake of comparison, their results are reported in Figure 4. The final hydraulic conductivity values of DPH and conventional GCL are very close. The apparently slower evolution of the hydraulic conductivity is probably a result of the different test procedure adopted by in their study. Firstly, the specimen were permeated directly with the salt solution whereas a preliminary wetting phase in the odeometer was carried out before permeation in the present study. Hence, both the contact time and the amount of available Ca²⁺ ions supplied at each wetting may have been less than in the present study, so that the cation exchange of sodium for calcium proceeded more slowly. Secondly, the specimens were air-dried to constant mass at room temperature, whereas a thermostatic chamber at 35 °C was used in this study. Lower residual water contents were probably achieved in this study. Therefore, the seemingly more rapid deterioration of DPH relative to the conventional GCL must not necessarily be interpreted as an inferior performance in response to dry/wet cycling.

CONCLUSIONS

The impact of dry/wet cycles on the hydraulic conductivity of a DPH (dense prehydrated GCL) was investigated. Distilled water (DW) and a 0.0125 M calcium chloride solution were used as wetting liquids. Drying was carried out in a thermostatic chamber at 35 °C. The impact of dry/wet cycles on the hydraulic conductivity was found to be scarcely significant if the hydrating fluid is DW, in analogy with conventional GCLs. Although a certain reduction of swelling capacity was observed, the major effect of dry/wet cycling appears the removal of polymers added to the bentonite and re-conversion into a standard sodium bentonite.

The hydraulic conductivity increased dramatically after few wetting cycles with the calcium chloride solution as hydrating liquid. The bentonite was eventually converted into a calcium-exchanged form, with a drastic reduction of swelling capacity. Formation of desiccation cracks that did not close upon rehydration are probably responsible for the increase in permeability, in analogy with conventional GCLs.

Conversely, continuous permeation with the calcium chloride solution had no significant effect on the hydraulic conductivity, in accordance with previous studies on DPH GCLs. The authors recognize that dehydration states achieved in this study were particularly severe. Further research work is necessary to estimate the actual degree of dehydration that can occur in typical applications. In this respect, in-depth investigation of the dehydration behaviour

of DPH in response to suction gradients as a function of retention properties of adjacent materials is also necessary. Test results suggest that DPH GCLs should be protected by adequately thick surface layers if common Ca and Mg-rich soils (i.e. not sodic) are used as covering layers and severe desiccation may occur. The "adequate" thickness is another subject that requires further investigation, since it is still a subject of debate also for conventional GCLs. Finally, test data on samples exhumed from real scale projects are still lacking for DPH GCLS and are therefore particularly desirable.

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Corresponding author: Dr Francesco Mazzieri, Technical University of Marche, Via Brecce Bianche, Ancona, Italy. Tel: 00390712204410. Email: <u>f.mazzieri@univpm.it</u>.

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