Wet and Dry Effect on the Hydraulic Conductivity of Polymer Treated GCL Prototype

Michela De Camillis[1], Gemmina Di Emidio[2], Adam Bezuijen[3] & Daniel Verastegui Flores[4]

[1] Scientific Staff, Ghent University, 9052 Zwijnaarde, Belgium
[2] Assistant Professor, Ghent University, 9052 Zwijnaarde, Belgium
[3] Professor and Chief of laboratory of Geotechnics, Ghent University, 9052 Zwijnaarde, Belgium
[4] Professor, Institute of Mechanics, Materials and Civil Engineering (iMMC), Université catholique de Louvain, 1348 Louvain-la-Neuve, Belgium

ABSTRACT: Geosynthetic clay liners (GCLs) are widely used to isolate pollutants because of their low hydraulic conductivity to water. However, the performance of clay barriers may be impaired by prolonged exposure to electrolytic liquids which may lead to the compression of the diffuse double layer. The consequences are the increase of permeability and the loss of self-healing capacity. Moreover, the efficiency of the liners can further deteriorate by repeated wet and dry cycles, which may lead to desiccation of the bentonite and associated cracking. Modified bentonites have been introduced to improve the resistance of clay barriers to aggressive solutions. This study deals with a polymer-amended clay, HYPER clay. HYPER clay is treated with an anionic polymer and dehydrated and it shows enhanced performance in presence of electrolyte solutions. The effect of wet and dry cycles on the hydraulic conductivity to seawater of needle-punched GCLs prototypes of treated and untreated bentonite was investigated. The prototype samples containing HYPER clay 8% showed lower permeability compared to those containing untreated bentonite. However, the temperature suggested from the standard used in this study is extremely high and it does not represent the temperature in the field.

INTRODUCTION

Geosynthetic clay liners (GCLs) are widely used as barrier systems in waste disposal facilities. GCLs are factory-manufactured hydraulic barriers containing a thin layer of bentonite sandwiched between geotextiles or glued to a geomembrane. GCLs can preserve good efficiency in the long term if the bentonite ability to maintain low hydraulic conductivity is not decreased by interaction electrolyte solutions and desiccation (Rowe, 2013). In this regards, the European Union introduced in the Council Directive 1999/31/EC the requirements of clay liners of at least 1 m thickness and 1×10^{-9} m/sec permeability to both bottom and cover liners. Usually, bentonite performance is influenced by the fluids with which the GCLs come in contact. Ion concentrations and valences of the fluids play an important role for the cation exchange mechanism. Shackelford et al. (2000) reported the influence of the valence on the increase of hydraulic conductivity, which has also been associated with high concentration of monovalent cations. Moreover, the expansion capacity of the bentonite is restricted when multivalent cations are present (Katsumi et al., 2008).

Clay liners can be desiccated or wetted repeatedly due to climatic and environmental changes. As a consequence, desiccation can have a negative impact on the sealing capacity of the bentonite and it may lead to crack formations. Crack formations result in reduction of the liner thickness, allowing the development of potentially preferential flow paths which lead to an increase in permeability (Hewitt and Philip, 1999). The influence of wet and dry cycles on the deterioration of hydraulic performance has been documented by several field investigations (Koerner and Koerner, 2005).

The combination of high electrolyte solution with desiccation may lead to loss of efficiency of the clay liner. The bentonite self-healing capacity can be affected by crack formation during the dry phase (Lin and Benson, 2000; De Camillis et al., 2014) and the swelling capacity may decrease as the number of wet-dry cycles increases (Bouazza et al., 2007; De Camillis et al., 2014).

Chemically modified bentonites have been developed to overcome the problems of containment clay barriers, in order to provide more chemical resistance and sustainable performance. In recent years, several researchers have investigated new amended clays (Mazzieri and Pasqualini, 2008; Katsumi et al., 2008; Di Emidio, 2010, Malusis and McKeehan, 2013; Scalia et al., 2013; Bohnhoff and Shackelford, 2014). A product in this category is the Multiswellable Bentonite (MSB), which is a bentonite treated with Propylene Carbonate (PC) to activate the osmotic swelling capacity of the clay. Another manufactured patented GCL is the Dense PreHydrated GCL (DPH GCL). This material is densified by calendaring after prehydration with a polymeric solution containing Na-CMC, sodium polyacrilate and methanol. Katsumi et al. (2008) reported results of long-term permeability tests on MBS and DPH GCL. The results show noticeable resistance to electrolyte solution of NaCl and CaCl2 with concentration of ≤ 1 M. The main issue of these modified bentonite is that the polymer adsorption onto the clay might not last enduringly (Mazzieri and Pasqualini, 2008; Di Emidio, 2010). In a study conducted by Di Emidio (2010), HYPER clay has been developed. The innovative clay is treated with anionic polymer and it was demonstrated thought XRD analysis the intercalation of the polymer in the interlayer region of the clay.

The purpose of this research is to investigate how the permeability performance of a polymer modified bentonite, HYPER clay, and that of an untreated sodium-activated bentonite can be affected by wet-dry cycles in contact with highly concentrated electrolyte solution like seawater.

MATERIALS

Two GCLs were used in this study: GCL Na-act contains sodium-activated bentonite; GCL HC+8% contains an amended bentonite, HYPER clay, treated with 8% of an anionic polymer. The procedure is explained below. The bentonites were sandwiched between a woven and non-woven geotextiles. The geotextiles were held together by needle-punching. The initial thickness was 4.3 mm for GCL Na-act and 4.9 mm for GCL HC+8%. The mass per unit area of the bentonite in the prototypes before wet and dry cycles was 4.0 kg/m^2, for the untreated bentonite, and 5.1 kg/m^2, for HYPER clay.

The principle of HYPER clay (Di Emidio, 2010) is to combine sodium bentonite with an anionic polymer (Carboxymethyl Cellulose, Na-CMC). The treatment method consists of mixing the base clay with a polymeric solution in deionized water (DI) with a mechanical stirrer for 30 minutes to increase the surface area available for polymer adsorption. This slurry is then oven dried at 105° C for 16 hours. After drying, the bentonites were ground using first a mortar and pestle and then a Retsch Mortar Grinder RM 200. In this research 8% of polymer, by dry weight of clay, was used (HC+8%).

Sea water (SW) was selected as hydration solution. The sea water was collected in different periods of the year in the North Sea (Ostend, Belgium). The electrolyte solutions had altered chemical properties due to seasonal changing. Ostend_0 has EC = 49.9 mS/cm, salinity = 32.4 and pH = 7.78 while Ostend_1 has EC = 33.9 mS/cm, salinity = 21.3 and pH = 7.99. Some properties of the solutions are listed in Table 1.

Table 1. Chemical properties of the electrolyte solutions

	Ostend_0 (mg/L)	**Ostend_1 (mg/L)**
Na^+	11517,9	6691
K^+	469,2	564,4
Mg^{2+}	1281	798,21
Ca^{2+}	478,5	400
Cl^-	19897	11042,2
SO_4^{2-}	2352	7104
HCO_3^-	183,1	23,2
CO_3^{2-}	18	11,4
NO_3^-	43,4	11
EC (mS/cm)	49,9	33,9
Salinity	32,4	21,3
pH	7,78	7,99
Eh (mV)	213	183

METHODS

Swell Index tests

Swell index tests (SI) were performed for quality testing on the treated and untreated bentonites extracted from the GCL prototypes. The test method followed was in accordance with ASTM D5890. Swell index were performed to study the effect of polymer treatment on the swelling capacity of the clays analyzed. Specimens of 2 g of untreated bentonite and HC+8% were poured in 0.2 g increments into a 100 ml graduated cylinder containing 90 ml of DI. After the 2 g of clay were added, the cylinders were filled up to 100 ml with additional DI. The tests were allowed to equilibrate for 16 hours (ASTM D5890) and the final volumes of swollen bentonite were recorded.

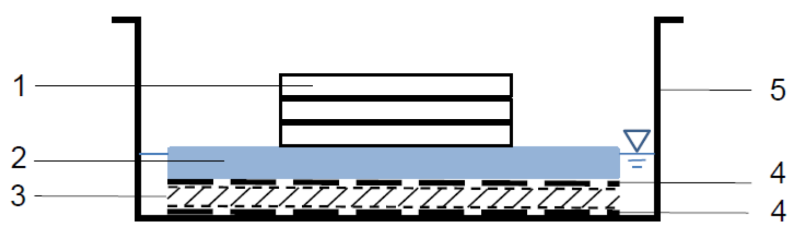
Wet and dry cycling

Wet and dry cycles (W-D) were performed following the draft Standard FprCEN/TS 14417. The standard defines the method for testing the influence ratio of wetting-drying cycles on the permeability through GCLs.

Specimens with an edge length of around 200 mm are saturated under a load corresponding to 4 ± 0.2 kPa for 48 h at constant room temperature. After saturation, the samples are dried under the same load in an oven at 110 ± 5 °C for 24 h. After drying the samples are allowed to cool to room temperature maintaining the same load for a minimum of 24 h. This wetting-drying cycle is performed four times before cutting a 100 mm diameter test specimen.

The hydration setup is shown in Fig. 1. The wetting cycles were performed using fresh seawater (Ostende_0).

These specimens were tested by means of hydraulic conductivity tests.



Key

1. Dead load
2. Pressure plate
3. GBR-C sample
4. Mechanically bonded nonwoven geotextile
5. Waterproof box

FIG. 1. Hydration setup
*GBR-C: clay geosynthetic barrier

Hydraulic conductivity tests

Hydraulic conductivity tests were conducted on the GCLs prototypes containing untreated sodium-activated bentonite and HYPER clay 8% exposed to W-D. The specimens were permeated with seawater Ostend_0 first and then with Ostend_1, when the previous solution was finished.

Tests were carried out in flexible wall permeameters with 100 mm of diameter. An effective stress of 27 kPa was applied according to ASTM D 6766. Termination criteria were followed according to ASTM D 6766. Specimens of the outlet were collected and analyzed to measure EC, pH, salinity and Eh.

RESULTS

Swell Index

The swelling ability of treated and untreated clays was quantified by means of standard swell index test as described above. Swell index results of sodium activated bentonite and HYPER clay 8% in deionized water are shown in Figure 2.

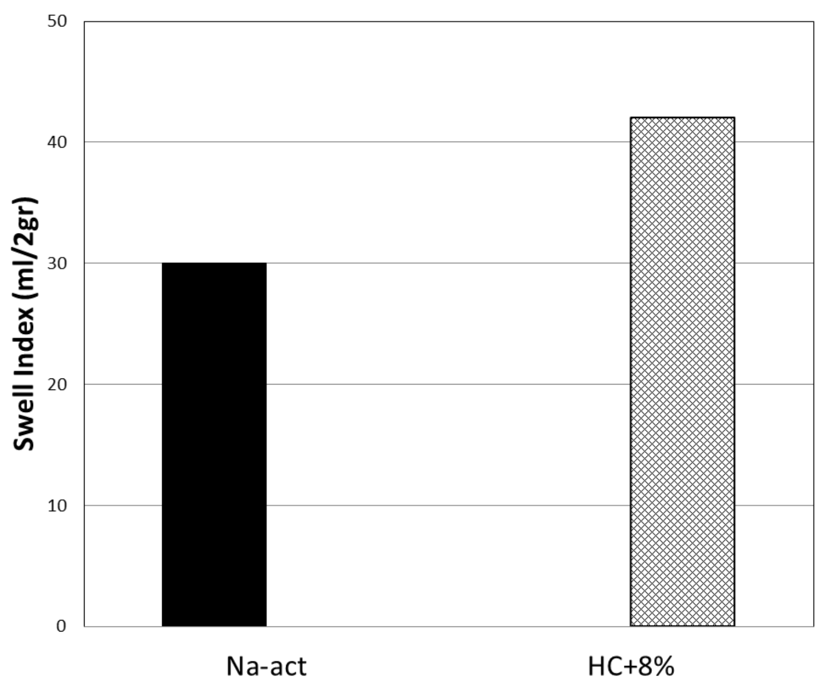


FIG 2. Swell index of Sodium-activated bentonite (Na-act) and HYPER clay 8% (HC+8%).

In deionized water HC+8% showed a larger swell index compared to untreated bentonite (42 ml/2g versus 30 ml/2g respectively). As a result, the treatment with the anionic polymer improved the swelling ability of the untreated clay.

The swell index is inversely related to the hydraulic conductivity (Jo et al., 2001), the hydraulic conductivity in fact increases with decreasing swell index (both normalized to their value in deionize water). Hydraulic conductivity test results will be described in the next section. They confirm such inverse relationship with swell index.

Hydraulic conductivity tests

Hydraulic conductivity tests were carried out on GCL prototypes containing untreated sodium bentonite and HYPER clay 8%. The specimens were permeated directly with seawater after being subjected to four wetting and drying cycles in the same electrolyte solution, e.g. seawater. Fig. 3 illustrates the results of hydraulic conductivity tests of Na-act and HC+8% permeated with SW after W-D.

Both specimens presented cracks after W-D cycles likely due to the strong electrolyte solution, the load applied and the drying temperature.

As it can be seen in Fig. 3, untreated bentonite reached a constant permeability ($k = 1.6 \times 10^{-8}$ m/s) after around 20 days. The permeation of Na-act was terminated when 139.5 pore volumes of flow passed through the specimen. HYPER clay 8% presented better behavior in contact with seawater. The test is still running because constant hydraulic conductivity has not yet been reached. The permeability is still decreasing and it reached values lower than the acceptable value of 1×10^{-9} m/s for a GCL.

The results of the untreated Na-act clay suggest that cracks may not be completely healed and the liquid can easily flow through the fissures before they are healed. Therefore, the electrolyte solution is more in contact with the clay particles near the cracks which are prone to cation exchange. As a consequence, the increase of permeability may be related to the combination of wet and dry cycles and the strong electrolyte solution. The behavior of bentonite is dominated by the thickness of the diffuse double layer (DDL). The DDL is in fact influenced by the concentration and the valence of the cations present in the electrolyte solution (Egloffstein, 2001). Increasing these parameters, the thickness of the DDL decreases. As a consequence, the clay structure becomes aggregated with short and fast flow paths, with a consequent higher permeability. On the other hand, HYPER clays showed a hydraulic conductivity lower than that of the untreated sodium bentonite. This is likely due to the thick diffuse double layer of the treated clay maintained open in the long-term due to the irreversible adsorption of the polymer into the clay (Di Emidio, 2010). The polymer intercalation in the interlayer helps to keep it thick adsorbing lots of immobile water molecules and ions. Therefore, the pore space is restricted and the flow pathways will be long and tortuous, decreasing the permeability of the soil.

Moreover, also the drying temperature might have an impact on the GCLs performance. Drying cycles resulted in a severe desiccation of bentonite. Untreated bentonite became severely affected by cracking after W-D in seawater. On the other hand, HYPER clay presented less volume of cracks.

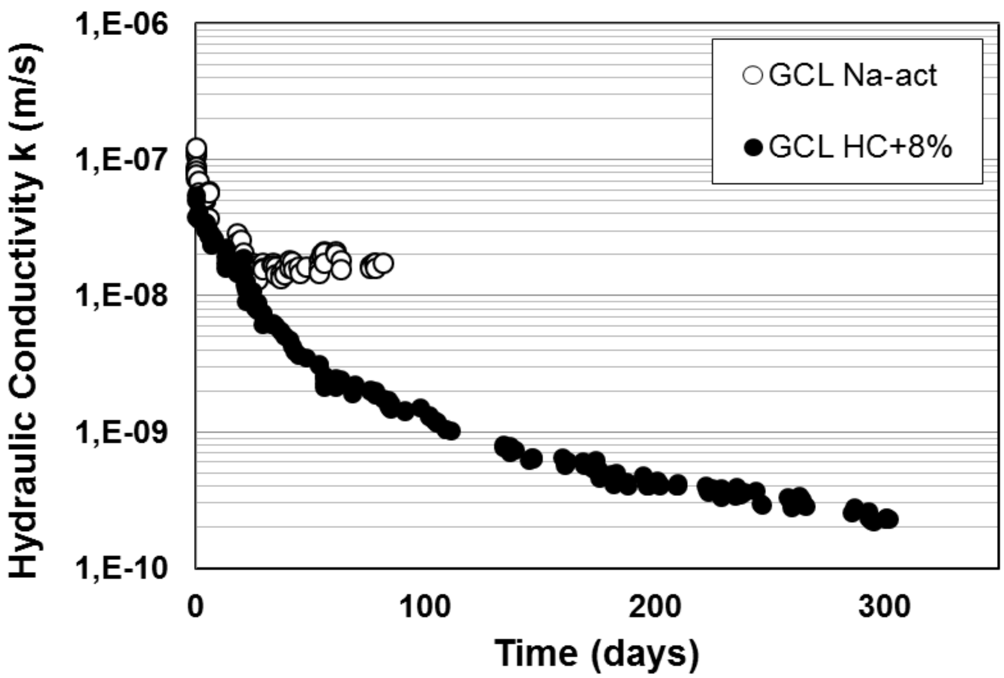


FIG 3. Hydraulic conductivity versus time of GCL containing untreated sodium-activated bentonite (GCL Na-act) and GCL containing HYPER clay 8% (GCL HC+8%).

The temperature used in this study differs from those used in prior studies (Lin and Benson, 2000; Mazzieri et al., 2009; De Camillis et al., 2015). Lin and Benson (2000) assessed the influence of wet and dry cycles with 0.0125 M calcium chloride at room temperature on GCLs containing Na-bentonite. Mazzieri et al. (2008) performed W-D on DHP GCL with 0.0125 M calcium chloride at 35°C. As reported by De Camillis et al. (2015), the permeability of untreated Na-bentonite subjected to W-D at 40°C increased up to 2.93×10^{-7} m/s at the fourth cycle (Fig. 4). The samples were permeated with deionized water during the first cycle and seawater in the consecutive cycles. On the other hand, the hydraulic conductivity of HYPER clay 8% remained low. Moreover, as indicated by De Camillis et al. (2015), the Na-bentonite specimen crumbled and HYPER clay 8% maintained a solid disk shape after being subjected to wet and dry cycles at 40°C in seawater. HC+8% presented no cracks in the core sample likely due to the gradual desiccation. The large swelling ability of HYPER clay helped to heal the cracks formed (on the edges of the sample) during the dry cycles and also to maintain low hydraulic conductivity.

This extreme desiccation (110 ± 5) °C is too severe and it will be unlikely observed in the field. On the contrary, a lower temperature is more representative of the reality and allows a gradual desiccation of the GCLs.

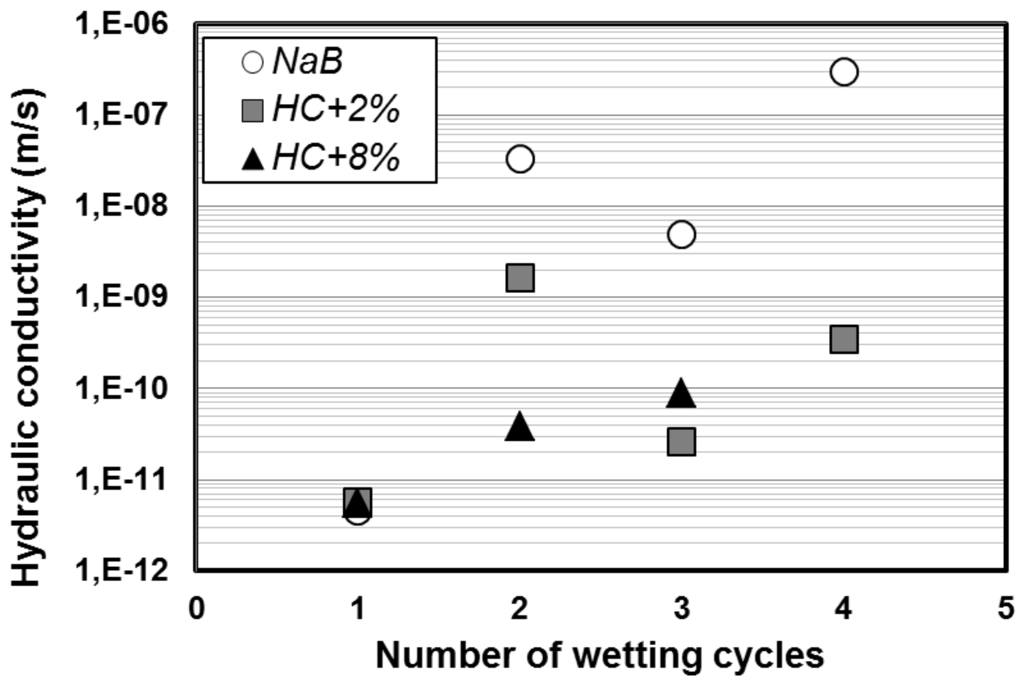


FIG 4. Hydraulic conductivities of sodium untreated bentonite (NaB), HYPER clay 2% (HC+2%) and HYPER clay 8% (HC+8%) at each wetting cycles (De Camillis et al., 2015).

CONCLUSIONS

The aim of this study was to simulate the condition of GCLs in the field in contact with strong electrolyte solutions. The effect of wet and dry cycles on the hydraulic conductivity was investigated for GCLs containing polymer treated and untreated bentonite. The efficiency of GCLs may be affected by wet-dry ageing, especially if electrolyte solutions are involved. The process that strongly affects the performance of GCLs is a combination of cation exchange, concentration of the permeant solution and crack formation. Cation exchange and ion concentration leads to reduction in swelling and self-healing capacity. On the other hand, crack formation induces an increase of the permeability of the bentonite because the liquid can easily flow through the fissures before they are healed. As a consequence, the electrolyte solution is more in contact with clay particles near the cracks which are prone to cation exchange. So, even if the cracks were healed, the permeability could increase due to the compression of the diffuse double layer thickness. These phenomena lead to a deterioration of the performance of GCLs and bentonite as barrier material. Consequently traditional GCLs are not able to contain leachate as sea-water and likely also not other strong electrolyte solutions but only fresh water.

The large increase in hydraulic conductivity of the GCL with untreated sodium-activated bentonite indicates that cracks did not seal properly. Most likely the compression of the diffuse double layer of the bentonite by high ionic concentration and presence of cracks led to an increase of the permeability above the limit value of 10^{-9} m/s.

HYPER clay treatment showed beneficial effects on the swelling and hydraulic behavior of the bentonite exposed to seawater. Crack formation was found after wet-dry cycles at (110 ± 5) °C. Such high temperature is not representative of the real condition in the field. However, the hydraulic conductivity test provides promising results. The large swelling ability of HYPER clay helped to heal the cracks formed during the cycles. The permeability reached values below the acceptable value of 10^{-9} m/s. It is likely that the presence of the polymer helped to keep the diffuse double layer open during hydration. Therefore, the structure of the modified bentonite remains dispersed and formation of preferential flow paths is limited. The drying temperature might have an impact on the performance of the GCL prototypes. The temperature suggested from the standard is extremely high, therefore it is not representative of the temperature in the field.

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