Impact of wet-dry cycles on the swelling ability and hydraulic conductivity of a polymer modified bentonite

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ABSTRACT: Geosynthetic clay liners (GCLs) are widely used as hydraulic barrier to isolate waste disposal facilities thanks to their low permeability to water. In the field, the efficiency of GCLs can be altered by cation exchange and highly concentrated solutions during contact with aggressive liquids and by the desiccation caused by seasonal temperature changes. Wet and dry cycles lead to desiccation and cracking of the bentonite. As a consequence, the self-healing capacity, the swelling ability and the hydraulic conductivity are impaired, hence leading to cracking of the bentonite and contamination of the ground. HYPER clay is a polymer modified bentonite developed to provide enhanced performance in presence of electrolyte solutions. In this paper, the effect of different drying temperatures on the swelling ability (air, 40°C and 60°C) and hydraulic conductivity (40°C, 60°C and 105°C) of powder and GCLs prototypes containing HYPER clay and untreated clay subjected to wet-dry cycles with seawater were investigated and compared. Test results showed that the drying temperature plays an important role on the crack formation and self-healing time of the tested materials. The HYPER clay treatment improved the swelling ability and self-healing capacity of the bentonite subjected to wet and dry cycles. Moreover, the permeability of the GCLs containing HYPER clay was lower than that of the untreated bentonite after four wet-dry cycles.

Keywords: GCL, modified bentonite, hydraulic conductivity, wet and dry cycles

1 INTRODUCTION

Geosynthetic clay liners (GCLs) are bentonite-based liners that are gaining acceptance as hydraulic barriers in containment and sealing applications (Petrov and Rowe 1997). One important field of application is landfill-capping systems. The aim of clay liners is to limit the infiltration of moisture, due to rainfall or water migration, through the barrier into the waste and to limit the release of leachate and gasses from the waste.

GCLs are factory-manufactured clay liners containing a thin (~ 10 mm) uniform layer of bentonite sandwiched between two geotextiles or glued to a geomembrane. The major component of the bentonite in GCLs is sodium montmorillonite. Montmorillonite is part of the smectite family, which is characterized by a high specific surface area, weak interlayer bonds and high cation exchange capacity. Smectite is a class of hydrated 2:1 layer silicate minerals that have an expandable volume due to the retention of hydration cations. Sodium cations are able to bond with water molecules, increasing the interlayer space and forming tortuous flow paths. However, valence, concentration and dielectric constant of the hydrating solution influence the expansion of the diffuse double layer (DDL) of negatively charged clays (McBride 1994). Accordingly, hydraulic conductivity and swelling of bentonite are related to the thickness of the DDL. A decrease of the thickness leads to an increase of hydraulic conductivity resulting in particle attraction, shrinkage and cracking of clay (Shackelford et al. 2000). Therefore, bentonite is sensitive to chemical interactions with the hydrating liquid.

The service life of a GCL cover can also be impaired due to climatic forces. Heat waves, seasonal rainfall and groundwater migration may damage the hydraulic performance of the liners subjected to wet-dry ageing (Mazzieri and Pasqualini 2008; Rowe et al. 2011; De Camillis et al. 2016). As a result, the hydraulic conductivity increases and the self-healing capacity decreases due to the combination of compression of the DDL and desiccation (Egloffstein 2001; Meer and Benson 2007). As a consequence, cracks might not heal during rewetting due to the low swelling ability of the bentonite caused by the compression of the DDL thickness.

Several studies have been conducted to assess the effect of wet-dry cycles on the hydraulic conductivity and the swelling ability of clay barriers through different methods (Lin and Benson 2000; Bouazza et al. 2006; Thiel et al. 2006; Benson and Meer 2009; Rowe et al. 2011; Tang et al. 2011; Take et al. 2012; Hoor and Rowe 2013; Mukunoki et al. 2014; Zangl and Likos 2016). This phenomenon has usually been investigated using oven or air drying. The drying temperature has an influence on the cracking pattern. Take et al. (2012) examined the difference between air drying at 20°C and rapid drying in an oven at 60°C. The experimental findings indicated that the size of cracks formed in the bentonite core of a GCL is correlated to the drying temperature. In particular, air drying at 20°C produced larger cracks compared to oven drying at 60°C.

Modified bentonites have been developed to improve bentonite performance in aggressive environments (Mazzieri and Pasqualini 2006; Katsumi et al. 2008; Di Emidio 2010; Malusis and McKeehan 2013; Scalia IV et al. 2014). To date, there are few studies on the effect of wet and dry cycles with aggressive electrolyte solutions on polymer modified bentonites (Mazzieri and Pasqualini 2008; Mazzieri et al. 2016). The authors suggested that the amendment was removed during the cycles, converting the modified bentonites into a conventional bentonite.

The aim of this study is to assess the effect of wet and dry cycles with seawater on the swelling ability, crack formation and hydraulic conductivity of a long-term performing polymer-modified bentonite, HYPER clay. HYPER clay technology consists of adsorbing irreversibly an anionic polymer, such as sodium carboxymethyl cellulose (Na-CMC) to clayey soils. Once the Na-CMC intercalates the clay platelets, the diffuse double layer is maintained open even in presence of factors that generally produce the collapse of the interlayer (Di Emidio 2010). Seawater was used to simulate highly concentrated solutions, inorganic part of a leachate or infiltration if the liner is used in a coastal area.

2 MATERIALS

Following the procedure developed by Di Emidio et al. (2015), a polymer solution was prepared by adding Na-CMC to deionized water. After mixing for 15 minutes with a high shear mixer, Na-activated bentonite (UC) was added and mixed for 30 minutes. The paste obtained was oven dried at 105°C for at least 16 hours to remove all excess water. After drying, the bentonite was first crushed manually with a mortar and a pestle and then with a mechanical grinder. In this research 8% of polymer, by dry weight of clay, was used to produce HYPER clay 8% (HC+8% or HC).

Table 1. Characteristics of seawater.

Na^+ [M]	0.5010
K^+ [M]	0.0122
Ca^{2+} [M]	0.0310
Mg^{2+} [M]	0.0585
Cl^- [M]	0.5610
SO_4^{2-} [M]	0.0229
HCO_3^- [M]	0.0030
CO_3^{2-} [M]	0.0002
NO_3^- [M]	0.0007
EC [mS/cm]	48.8
Salinity [-]	31.6
pH [-]	7.38
Ionic Strength [M]	0.764

Two needle-punched GCL prototypes were used in this study: GCL_UC containing sodium-activated bentonite and GCL_HC containing the amended bentonite. The bentonites were sandwiched between a woven and a non-woven geotextile with a dry bentonite mass of about 4.5 kg/m^2.

Fresh sea water (SW), collected in the North Sea (Ostend, Belgium), was selected as hydrating solution. Some properties of the solution are listed in Table 1.

3 METHODS

3.1 *Swelling ability*

The influence of different drying temperatures on the behaviour of HYPER clay subjected to wet and dry cycles was investigated and compared to the behaviour of untreated bentonite.

The temperature impact was studied using the procedure of the standard FprCEN/TS 14417 which defines the method for testing the influence ratio of wetting-drying cycles on the permeability through GCLs. To study temperature impact, although the standard suggests a drying temperature of 105°C, the cycles were performed at temperatures of about 20°C, 40°C and 60°C.

Powder dry bentonite with initial dry mass of 4 kg/m^2 and 0.718 porosity was evenly spread between two filter papers and two drainage layers in a 9 cm diameter oedometer cell. The samples were hydrated for 48 hours with deionized water (DW) in the first cycle and with seawater (SW) in the subsequent cycles, under an overburden pressure of 4 kPa, as recommended by the standard. During hydration the samples were wrapped in plastic foil to prevent evaporation. The samples were dried at the above indicated temperatures under a seating pressure of 4 kPa till the water content (w%) of both UC and HC+8% was lower than 25%.

During every wet and dry cycle, the swelling ability, self-healing capacity and crack formation of both treated and untreated bentonite were measured and compared.

3.2 *Hydraulic conductivity*

The hydraulic conductivity to seawater of a GCL sample containing untreated clay (GCL_UC) was compared to that of a GCL sample containing HYPER clay 8% (GCL_HC). Both samples were initially subjected to four wet and dry cycles with a drying temperature of 40°C, 60°C or 105°C. Wet and dry cycles were performed following the Standard FprCEN/TS 14417. Specimens with an area of 200 by 200 mm were cut out of the GCL prototypes. Tape was added to the edges to prevent bentonite loss. Each sample was placed between two drainage layers in a large glass plate. The samples were hydrated for 48 hours under an overburden pressure of 4 kPa. The samples were then dried in an oven until the water content of both GCLs was lower than 25%.

At the end of the fourth drying cycle, a sample with a diameter of 10 cm was cut from the panels and placed between two porous stones and filter papers into a flexible wall permeameter cell. The hydraulic conductivity to seawater was then measured using the falling head test method. An effective stress of 27 kPa was applied according to ASTM D6766.

4 RESULTS

4.1 *Swelling ability*

The vertical swell of all the samples at the end of each wetting cycle is represented in Fig. 1. The initial height of UC and HC+8% was equal to 8.8 mm and 7.9 mm respectively.

As expected, the largest swell was obtained during the first cycle with deionized water. Rewetting the samples with seawater, during the second cycle, did not induce any vertical swell in the UC samples, which is consistent with the Gouy-Chapman theory.

For UC dried at room temperature at about 20°C, the swell decreased gradually during the consecutive cycles. Increasing the drying temperature from 20°C to 40°C resulted in a nearly constant swell during the third and fourth cycle. However, the swell further decreased during the fifth wetting with seawater. The sample dried at 60°C showed similar swelling during the third wetting compared to the second wetting. However, the swell dropped during the fourth wetting and then slightly increased during the fifth wetting.

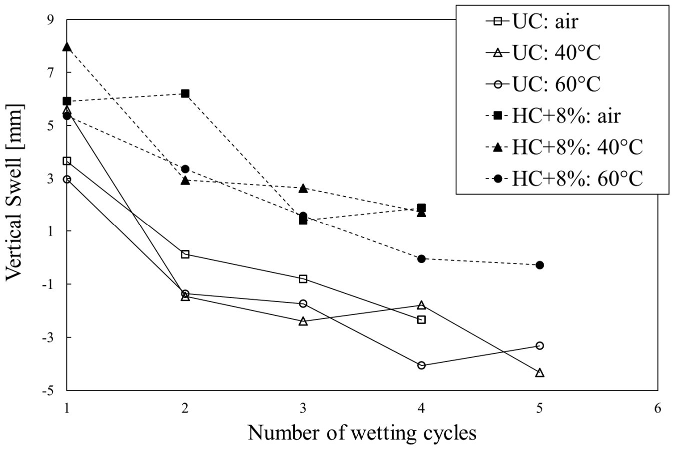

Figure 1. Vertical swell at the end of each wetting cycles of untreated bentonite (UC) and HYPER clay 8% (HC+8%).

At the end of the first drying cycle, no cracks were visible in the samples that were slowly air-dried at room temperature (pictures not available). Drying the samples at 40°C on the other hand, resulted in few cracks both in UC and HC+8% (Fig. 2). Further increasing the drying temperature from 40°C to 60°C resulted in more cracks both for UC and HC+8%. Therefore, increasing the drying rate led to the formation of more cracks both for untreated and polymer treated bentonite.

For HC+8% on the other hand, the vertical swell in DW was always higher compared to UC. The swell of the sample HC + 8% dried at room temperature slightly increased during the second wetting. The swell then dropped during the third wetting but was still higher compared to UC.

Further increasing the drying temperature from 40°C to 60°C, resulted in a gradual decreasing swell during the three consecutive cycles. The vertical swell then remained constant during the fourth and fifth wetting. Therefore, the swell of HC+8% was still maintained until the fourth cycle for all three drying temperatures while UC already collapsed during the second wetting with SW.

Each sample was subjected to a visual inspection to investigate the desiccation pattern at the end of the drying cycle and the self-healing capacity after rewetting. Fig. 2 shows the samples of UC and HC+8% at the end of each wet and dry cycle for the different drying temperatures.

Visual inspection showed that the cracks present in the samples dried at 40°C likely healed upon rewetting with SW. Drying the samples at 60°C did not only result in the formation of more cracks but also apparently lower self-healing capacity upon rehydration with SW after 48 hours. Two days of hydration might not be enough and more days may be needed to heal the cracks in HC+8% dried at 60°C. This could be due to the formation of an impermeable shell around the cracks of the HYPER clay sample. On the contrary, the cracks present in untreated clay (60°C) seemed to be healed upon rewetting with SW. However, the water adsorption and vertical swell of UC significantly decreased during the second wetting with SW that indicates the collapse of the DDL. HYPER clay on the contrary showed higher water adsorption and swelling compared to UC, as well as two orders of magnitude lower permeability (see section 4.2).

As it can be seen in Fig. 2, the area of intact bentonite between the cracks decreased with increasing drying temperature. Slowly air-drying at about at 20°C the bentonite resulted in a desiccation network where the width of the cracks was larger than at 40°C or 60°C, in agreement with the findings of Take et al. (2012).

Comparing the samples dried at 40°C and 60°C at the end of the fifth wetting cycle with SW, some surface cracks were visible for HC + 8% while the surface of UC seemed smooth. However, the vertical swell of the polymer treated bentonite was higher compared to UC as it can be seen in Fig. 1. As reported in De Camillis et al. (2016) and shown in Fig. 3, μCT Scanner and analysis demonstrated that the cracks developed on the polymer modified bentonite, HYPER clay 8%, were only superficial, with lower volume compared to those of untreated bentonite and they did not propagate downwards. This indicates that the diffuse double layer of the polymer treated bentonite was still sufficiently thick while for untreated clay the diffuse double layer collapsed due to wet and dry ageing.

This indicates that some quality tests, such as determination of the self-healing capacity of the bentonite based on visual inspection of the surface, is not sufficient for polymer treated bentonite and actual hydraulic conductivity tests are recommended.

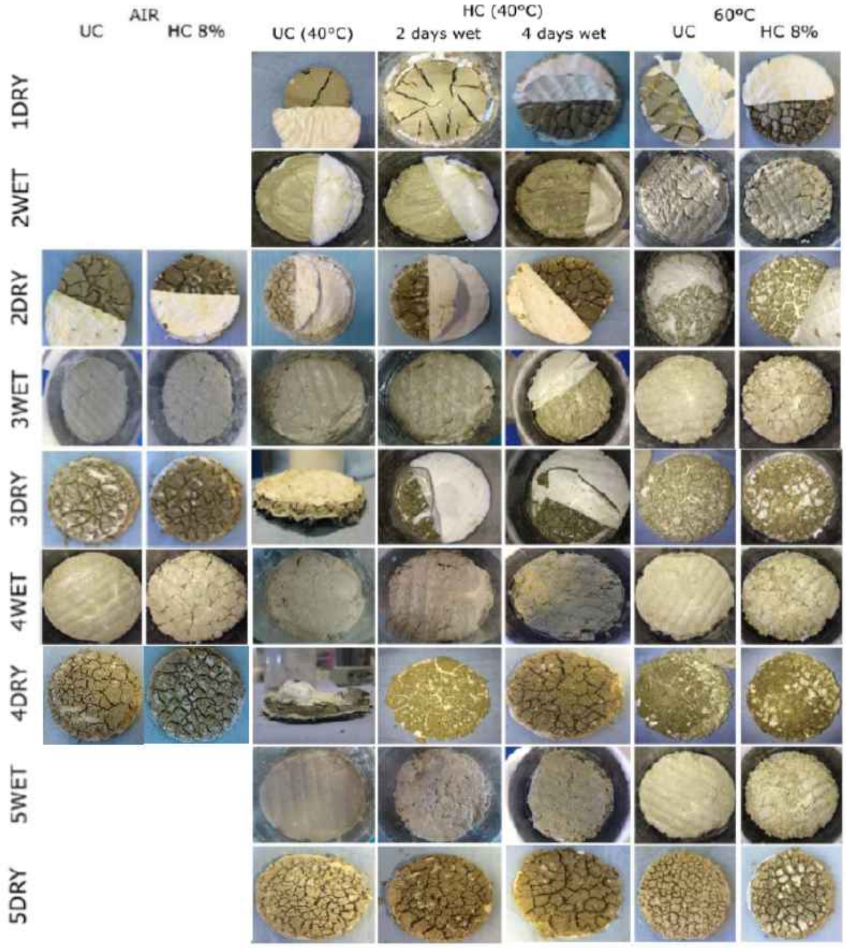

Figure 2. Untreated clay (UC) and HYPER clay 8% (HC) at the end of each wet and dry cycle for different drying temperatures (room temperature of about 20°C, 40°C and 60°C).

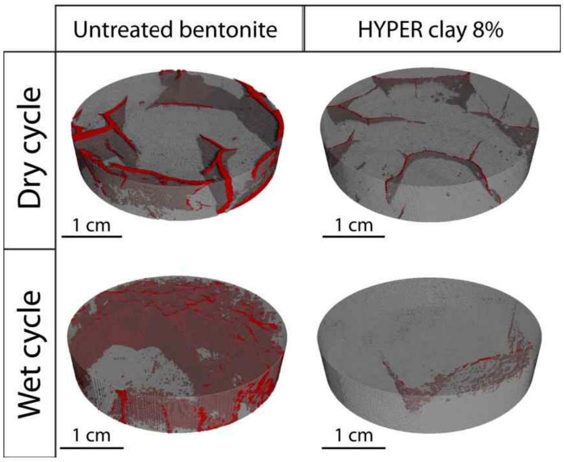

Figure 3. 3D rendered results of the μCT scan of untreated bentonite and HYPER clay 8%, after the 3rd dry cycle and during the 4th wet cycle. Open syneresis cracks are displayed in red. (De Camillis et al., 2016)

4.2 *Hydraulic conductivity*

The impact of different temperatures on the hydraulic conductivity of GCL prototypes is shown in Fig. 4. The graph represents the hydraulic conductivity of the GCLs containing untreated bentonite (GCL_UC) and HYPER clay 8% (GCL_HC) after four wet and dry cycles versus drying temperature.

The hydraulic conductivity of GCLs was tested at 40°C, 60°C and 105°C. In general, 40°C drying temperature is more representative of the reality. Drying at 60°C simulates, in fact, high temperatures that could occur in extreme cases, such as for a GCL placed under a black geomembrane (without soil protection) that absorbs UV radiation (Rowe, 2014).

GCL_HC presented always a permeability lower of about 1-2 orders of magnitude than that of GCL_UC at every given condition. In addition, the better hydraulic performance at 40°C of the GCLs might be related to the effect of prehydration. As demonstrated by Lin and Benson (2000) and Mazzieri and Pasqualini (2008), prehydration may delay but not prevent the increase of hydraulic conductivity.

The low hydraulic conductivity of GCL_HC indicates that the swelling of the polymer treated bentonite closed the cracks formed during desiccation. This demonstrated that the adsorption of the polymer helped to maintain the diffuse double layer open during hydration with SW after four wet and dry cycles.

As it can be seen in Fig. 4, the hydraulic conductivity of the samples tested at different temperatures is almost constant. Milder seawater (EC=33,9 mS/cm) was used for the tests carried out at 105°C, still this represents a strong electrolyte solution.

The impact of different drying temperature might be related more to the self-healing timing rather than to the hydraulic conductivity. As reported by De Camillis (2017), increasing the drying temperature, the time required for swelling bentonites to heal their cracks increased.

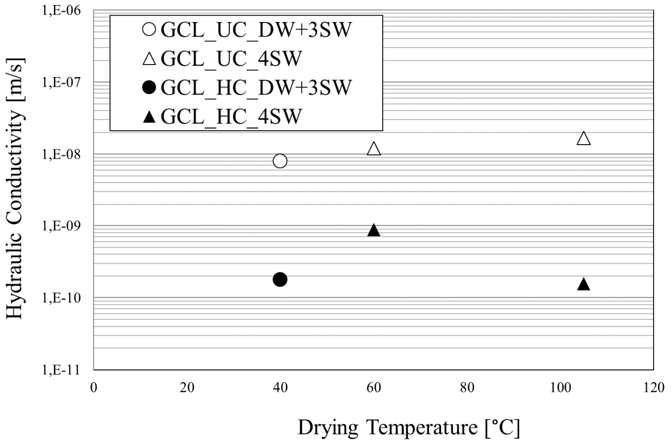

Figure 4. Hydraulic conductivity of GCL prototypes containing untreated bentonite (GCL_UC) and HYPER clay 8% (GCL_HC) after four wet and dry cycles with prehydration (DW+3SW) and direct exposure to seawater (4SW) at 40°C, 60°C and 105°C.

5 CONCLUSIONS

Geosynthetic clay liners are widely used in landfill applications to isolate waste liquids from the environment. GCLs must ensure low hydraulic conductivity in the long term to avoid migration of contaminants in the surrounding soil and groundwater.

In this research, the performance of a GCL containing untreated sodium activated bentonite was compared to a GCL with HYPER clay 8%, under wet and dry cycles in seawater.

Untreated and polymer treated powder bentonites were subjected to four wet and dry cycles (DW+3SW) with three different drying temperatures (room temperature at about 20°C, 40°C or 60°C) in oedometer cells. At the end of each cycle, the swelling ability and self-healing capacity were investigated. In general, the vertical swell of HC+8% was higher compared to UC during each wetting cycle independently of the drying temperature. This is likely due to the adsorption of the polymer, which enhances the behaviour of the bentonite when subjected to wet and dry cycles using seawater. The swell of HC+8% was still consistent until the fourth cycle for all three drying temperatures.

Increasing the drying temperature, the amount of cracks present in UC and HC+8% increased. However, the crack width was higher for the samples that were slowly air-dried compared to the samples rapidly dried in an oven set at 40°C or 60°C.

The hydraulic conductivity to SW of two GCL prototypes (GCL_UC and GCL_HC) initially subjected to four wet and dry cycles was measured. During each drying cycle, the samples were oven dried at 40°C, 60°C or 105°C. In general, the hydraulic conductivity of the polymer treated bentonite was always about 1-2 orders of magnitude lower compared to the untreated base bentonite. The drying temperature did not influence the hydraulic conductivity. On the contrary, it had an impact on the self-healing time. Indeed, increasing the drying temperature the specimens healed slower.

It can be concluded that the polymer treated bentonite showed enhanced behaviour even after wet and dry cycles with a strong electrolyte solution, e.g. seawater. Therefore, HYPER clay might be a valuable substitute for untreated bentonite in GCLs under aggressive conditions.

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