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Research paper

Effect of wet-dry cycles on polymer treated bentonite in seawater: swelling ability, hydraulic conductivity and crack analysis

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ABSTRACT

Waste disposal facilities are often isolated by clay liners in order to prevent pollutant migration into the ground. Bentonite is used as barrier material thanks to the low conductivity to water. However, the hydraulic performance may be impaired by contact with aggressive liquids due to cation exchange and highly concentrated solutions. Moreover, the combination of cation exchange and wet-dry cycles can further affect the efficiency of the bentonite as barrier material. This study was carried out to evaluate the effect of wet-dry cycles with seawater on untreated sodium bentonite and HYPER clay. HYPER clay is a polymer modified bentonite with enhanced performance in presence of electrolyte solutions. Bentonite and bentonite treated with 2% and 8% polymer by dry weight of the clay were evaluated for their swelling ability, self-healing capacity, crack formation and hydraulic conductivity by means of one-dimensional swell test, μCT scanning and hydraulic conductivity tests. The specimens were subjected to 6 wet-dry cycles for the swell tests and to 4 cycles for the hydraulic conductivity tests. One-dimensional swell tests results showed that HYPER clay 8% had swollen the most and that its thickness after the 6th wet-dry cycle was comparable to the original thickness of untreated bentonite during its maximum swelling in deionised water. μCT analysis demonstrated the better self-healing capacity and the smaller volume of cracks of HYPER clay compared to untreated bentonite. Unlike the untreated clay, HYPER clays maintained low permeability to seawater throughout the wet-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 sodium or calcium bentonite sandwiched between two geotextiles or glued to a geomembrane. Nowadays, GCLs are considered an efficient alternative to compacted clay liners (CCLs) thanks to their low permeability to water. The main advantages of GCLs, compared to CCLs, are the limited thickness, easy installation and good compliance with differential settlements of underlying soil or waste (Bouazza, 2002).

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 hydrated 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. Ion exchange that occurs in the bentonite can alter its physical properties (Meer and Benson, 2007). Several laboratory studies have investigated barrier performance deterioration of GCLs in contact with electrolyte solutions (Bouazza,

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2002; Jo et al., 2005; Kolstad et al., 2004; Petrov and Rowe, 1997). These studies have elucidated the influence of valence and concentration of different electrolyte solutions. For instance, Jo et al. (2005) conducted hydraulic conductivity tests using different salt solutions and concentrations, such as NaCl, KCl and $CaCl_2$. They found that the permeability increases up to 3 orders of magnitude in the presence of strong electrolyte solutions ($CaCl_2 \geq 50$ mM) compared to deionised water.

Nevertheless, 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., 2014, 2016. Temperature as high as 70 °C may occur due to daily thermal cycles when a geomembrane overlies a GCLs (Take et al., 2014). As a result, the hydraulic conductivity increase and the self-healing capacity decreases due to the combination of ion exchange and desiccation (Egloffstein, 2001; Meer and Benson, 2007). In particular, desiccation and contact with high electrolyte solutions are responsible for the collapse of the diffuse double layer and crack formation. Therefore, crack formation 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 hydraulic conductivity and the swelling ability of clay barriers through different methods (Lin and Benson, 2000; Bouazza et al., 2006, 2007; Thiel et al., 2006; Benson and Meer, 2009; Komine et al., 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. However, 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.

Cover liners might undergo differential settlement due to loading inducing cracking and loss of fluids. In this regards, Mukunoki et al. (2014) investigated the deformation field of compacted clay liners specimens by means of a bending apparatus combined with X-ray CT. High resolution X-ray Computed Tomography (μCT) can be used in order to internally observe crack formation of geomaterials (Cnudde and Boone, 2013), as well as the self-healing capacity of the material.

To date, the impact of wet and dry cycles on the swelling ability and hydraulic conductivity is understudied, particularly for the influence of the first hydrating liquid and the electrolyte solution composition. Several studies confirmed the effectiveness of prehydration prior to contact with the electrolyte solution during wet and dry cycles. For instance, Lin and Benson (2000) and Bouazza et al. (2007) reported that the initial exposure to low concentrated liquids, e.g. deionised water or tap water, temporarily delayed the reduction in swelling and the increase in permeability. Therefore, prehydration may improve bentonite resistance to aggressive solutions compared to non-prehydrated bentonite. On the other hand, Lin and Benson (2000) and Bouazza et al. (2006) directly permeated needle-punched GCLs with 0.0125 M $CaCl_2$ and this caused an irreversible damage to the efficiency of the GCLs as barrier liner due to the combination of cation exchange and crack formation. However, GCLs exposed to low concentrated solutions during wet-dry cycles might retain their ability to self-heal during rehydration, as reported previously by Lin and Benson (2000), Bouazza et al. (2007) and Benson and Meer (2009). Nevertheless, Benson and Meer (2009) highlighted that the increase in hydraulic conductivity in GCLs specimens is linked to the RMD (defined as the ratio of the total molarity of monovalent cations to the square root of the total molarity of multivalent cations at a given ionic strength) rather than to the ionic strength. In other words, the hydraulic conductivity increased as RDM decreased, regardless of whether the ionic strength was high or low. Moreover, the authors point out the presence of cracks to any solution used. In addition, they evidenced that cracks formed during desiccation did not heal for low RDM values, leading to an increase of GCLs permeability.

Modified bentonites have been developed to improve bentonite performance in aggressive environments (Katsumi et al., 2008; Mazzieri and Pasqualini, 2006; Di Emidio, 2010; Razakamanantsoa et al., 2012; Bohnhoff and Shackelford, 2014; Malusis and McKeehan, 2013; Scalia et al., 2013). In this research, the HYPER clay technology has been investigated. HYPER clay is a polymer-treated bentonite created by combining natural Na-bentonite with sodium carboxymethyl cellulose (Na-CMC). 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).

This study is part of a broader research to evaluate the performance of the recently patented polymer-amended HYPER clay subjected to wet-dry ageing in contact with seawater (De Camillis et al., 2016). The effect of wet and dry cycles with seawater on the swelling ability, crack formation and hydraulic conductivity is investigated. In particular, the efficiency of untreated bentonite is compared to that of HYPER clay. Seawater was used to simulate highly concentrated solutions or seawater infiltration if the disposal facility was constructed in a coastal area. The present paper provides new insights into the self-healing capacity and crack formation through the technique of μCT scanning. By focusing on the core of the specimens it is possible to quantify the amount of cracks present in untreated bentonite and HYPER clays in three dimensions, and the different cycles. This information is combined with a traditional evaluation of the swelling and hydraulic performance of the clays.

2. Materials

Sodium bentonite (NaB) was used in this study in its natural form and treated according to the HYPER clay procedure (as proposed by Di Emidio, 2010). Physical properties of the bentonite and HYPER clay are listed in Table 1. The HYPER clay treatment consists of mixing powder Na-bentonite (NaB) with a polymeric solution using a mechanical stirrer for 30 min. The polymeric solution is previously prepared by dissolving the anionic polymer, sodium carboxymethyl cellulose (Na-CMC) in water. This paste is then oven dried at 105 °C for 16 h to irreversibly adsorb the polymer. The HYPER clay is then ground first

Table 1
Material characterization.

Property	NaB	HYPER clay 2%	HYPER clay 8%
Specific gravity [−]	2.66	2.53	2.25
Liquid limit [%]	649.7	696.4	988.8
Plastic limit [%]	48.3	87.2	157.2
Plasticity index [−]	601.4	609.1	831.4
CEC* [meq/100 g]	70.17	75.32	87.56
Exchangeable cations**			
Ca^{2+}	43.25	45.32	39.73
K^{+}	1.52	2.01	1.85
Mg^{2+}	15.56	14.81	13.31
Na^{+}	40.28	50.69	62
Chemical composition***			
Al_2O_3%	17.3	17.31	16.74
CaO%	2.05	2.05	1.98
Fe_2O_3%	5.04	5.06	4.87
K_2O%	0.63	0.65	0.62
MgO%	2.35	2.34	2.25
MnO%	0.09	0.09	0.08
Na_2O%	1.85	2.04	2.63
P_2O_5%	0.06	0.06	0.06
SiO_2%	55.96	55.79	53.77
TiO_2%	0.21	0.2	0.2
LOI	14.64	14.63	17.59

* CEC is measured using ammonium acetate method.
** Exchangeable cations values include soluble cations.
*** Chemical composition is measured using ISO14869-2:2002.

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