



Potential hydraulic barrier performance of cyclic organic carbonate modified bentonite complexes against hyper-salinity[☆]



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ABSTRACT

The effect of adding glycerol carbonate (GC) or propylene carbonate (PC) to sodium (Na)-bentonite on the hydraulic performance of geosynthetic clay liners (GCLs) under hypersaline conditions is examined. Fluid loss (FL), swell index (SI) and solution retention capacity (SRC) measurements were carried out to compare the potential hydraulic performance of these two cyclic organic carbonates (COCs) as bentonite modifiers. A modified FL test enabled quantitative measurement of both the water retention characteristics of untreated and COC modified bentonites as well as calculation of hydraulic conductivity values. Tests under aggressively saline conditions (ionic strength, $I \geq 1$ M of NaCl and ≥ 3 M of CaCl₂) showed that at a mass ratio of 1:1 (GC to bentonite), the FL of a GC-Na-bentonite was ≈ 40 – 104 mL in NaCl and ≈ 61 – 91 mL in CaCl₂. This was about 10–20 mL and 70–200 mL, respectively, lower than that of a comparable PC-Na-bentonite (1:1 PC to bentonite) and untreated Na-bentonite. Greater swelling (SI) and greater solution retention capacity (SRC) was observed for the GC treated Na-bentonite compared to untreated Na-bentonite in all salt solutions, and for PC-Na-bentonite at high ionic strength of both NaCl and CaCl₂ solutions, demonstrating the superior hydraulic barrier performance of COC-bentonites under severely saline conditions. Experiments conducted in flexible-wall permeameters with $I = 3$ M CaCl₂ showed approximately one order of magnitude lower ($\sim 10^{-11}$ m/s vs $\sim 1.9 \times 10^{-10}$ m/s) hydraulic conductivity of GC treated bentonite cake compared to the k value of the untreated Na-bentonite cake. Calculated hydraulic conductivity from fluid loss tests estimated the measured values in a conservative way (overestimation).

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1. Introduction

The Australian mining industry produces tens of thousands of tonnes of saline leachates each year, which needs to be disposed of

safely (Australian Bureau of Statistics, 2012). In this respect, there has been an increasing use of geosynthetic clay liners (GCLs) in disposal impoundments where processing liquors from solid ore wastes are stored (Hornsey et al., 2010; Bouazza and Gates, 2014). The excellent hydraulic barrier performance of GCLs (Egloffstein, 2001; Bouazza, 2002; Guyonnet et al., 2005; Harvey and Lagaly, 2006; Rowe, 2014) have made them important hydraulic barrier components in waste containment facility applications (Egloffstein, 2001; Bouazza, 2002; Rowe et al., 2004; Rowe, 2005; Gates et al., 2009; Lange et al., 2009; Bouazza and Bowders, 2010). While GCLs have superior sealing properties, which provide low permeability to water, other investigations have pointed out the potential chemical incompatibility of these materials when exposed to leachates having extremes of pH or ionic strength (I) (Petrov et al., 1997; Petrov and Rowe, 1997; Shackelford et al., 2000; Jo et al., 2001; Kashir and Yanful, 2001; Kolstad et al., 2004; Jo et al.,

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2005; Benson et al., 2008, 2010; Gates and Bouazza, 2010; Bouazza and Gates, 2014; Liu et al., 2013, 2014, 2015). Chemical incompatibility between Na-bentonite and saline leachates, in part, stems from a loss of crystalline and osmotic swelling of smectite (the active component of bentonite), resulting from the high osmotic strengths of hypersaline leachates (Norrish, 1954; Slade et al., 1991).

As the use of GCLs is not normally limited to be in contact only with clean water during their service life, there exists a need (i) to optimise the hydraulic behaviour of GCLs in contact with nonstandard liquids (Shackelford et al., 2000; Malusis and Shackelford, 2002), and (ii) to develop possible solutions for applications where saline leachates, particularly calcium-enriched leachates, may have significant detrimental effects on the hydraulic performance of GCLs. In addition to the osmotic effect, calcium-enriched leachates may permit ion-exchange on the bentonite, which also has been shown to be detrimental to hydraulic performance under most conditions (Guyonnet et al., 2005; Lee and Shackelford, 2005; Benson et al., 2008). Osmotically induced desiccation and cation exchange reactions, which may occur in bentonites when exposed to saline and hypersaline leachates, can change the microstructure of the bentonite in GCLs by removing the hydration water from the smectite interlayer space (Aylmore and Quirk, 1960; Suquet et al., 1975; Tessier, 1991; Guyonnet et al., 2005; Laird, 2006) and ultimately may result in increased porosity because of the collapse of the smectite gel structure (Norrish and Quirk, 1954; Quirk and Schofield, 1955; Slade and Quirk, 1991; Slade et al., 1991). Such an effect has been identified as important in diminishing the tight seal that swollen bentonite normally has to liquids (Shackelford et al., 2000; Chertkov and Ravina, 2001; Rowe, 2005; Gates and Bouazza, 2010; Bouazza and Gates, 2014).

The objective of this study was to examine the use of glycerol carbonate (GC) modified bentonite, in an effort to minimise adverse effects associated with osmotic dehydration as well as calcium (Ca^{2+}) for sodium (Na^+) exchange of smectite in hypersaline leachates. Osmotic dehydration in this sense is defined herein as the loss of water from the interlayer space of smectite due to the osmotic suction imposed by saline leachates. GC is a non-toxic cyclic organic carbonate solvent having good solubility in water and high dielectric permittivity (Chernyak, 2006). Moreover, GC can be relatively inexpensively synthesized by green chemical methods (Turney et al., 2013). A structurally similar cyclic organic carbonate, propylene carbonate (PC), has previously been studied for similar purposes by other researchers (Onikata and Kamon, 1996; Onikata et al., 1999; Katsumi et al., 2008; Malusis et al., 2010; Bohnhoff et al., 2013; Mazzieri et al., 2013; Di Emidio et al., 2015). However, partly as a result of its greater dielectric permittivity (~110) compared to PC (~66) (Chernyak, 2006), GC offers a stronger interaction with interlayer Na^+ as well as other exchangeable cations that may be present within the interlayer of the smectite. Because of its higher dielectric permittivity, GC more efficiently penetrates smectite interlayer spaces thereby forming H-bonds either with the hydration water surrounding the interlayer cations or directly with the cations when hydration water is removed (Gates et al., 2016). In addition, GC has a free hydroxyl group which may assist in the retention of water through hydrogen bonding. In this study the hydraulic performance of an untreated Na-bentonite and the same bentonite with GC and PC modifications were compared. A modified version of fluid loss (FL), swell index (SI) and solution retention capacity (SRC) tests were used to assess the hydraulic performance of the untreated and COC-modified bentonites. These tests provided quick, reliable, and representative assessments on the water retention characteristics and swelling properties of each system. In particular, the FL and SRC

tests provide quantitative information that may be as reliable as the longer-term standardized hydraulic conductivity tests. Hydraulic conductivity tests were performed to assess the long-term performance of untreated and GC modified bentonite and to compare measured hydraulic conductivity values with k values calculated from FL tests.

2. Materials and methods

2.1. Cyclic organic carbonate treated bentonite

A natural Na-bentonite (Trugel, Miles, Queensland, Australia), widely used in Australian made GCLs, was studied in the present investigation. Its mineralogical and chemical properties are summarized in Table 1. Smectite makes up nearly all of the <0.2 μm fraction, which itself comprises 50% of the bulk material. The methylene blue (bulk) CEC is based on the method of Wang et al. (1996) and is performed on the bulk bentonite material. It assumes that for each unit of charge, one MB molecule is adsorbed by the surface. The Ba-CEC method (CSIRO Land and Water, in-house method) is an X-ray Fluorescence method which measures the amount of Ba^{2+} retained by the smectite portion (i.e. the <0.2 micron size fraction) of the bentonite. As such the Ba-CEC method does not provide an estimate of bulk CEC, but rather more closely approximates the layer charge of the smectite minerals. This latter information is important in gauging the effectiveness of COCs in inhibiting osmotically induced desiccation of the bentonite.

GC was synthesized by the procedures outlined in Turney et al. (2013). PC (anhydrous, 99.7%) was purchased from Sigma–Aldrich. Basic chemical properties of GC and PC are summarized in Table 2.

To prepare the modified bentonite, known amounts of either GC or PC and bentonite (with approximately 8% gravimetric water content) were mixed by hand with a spatula for approximately 3 min, followed by a 24 h rest prior to testing. The 3 min mixing time was sufficient to fully wet the samples and to break up any obvious agglomerates. The resting time allowed the liquid cyclic organic carbonates (COCs) to intercalate the bentonite homogeneously as indicated by X-ray powder diffraction (XRD) measurements (Fehervari et al., 2016; Gates et al., 2016). GC and PC were each added to the bentonite at two ratios by weight: 1:2 (50 wt%) and 1:1 (100 wt%) COC: bentonite. A 1:1 ratio was used for the FL tests. When performing the SI test a 1:2 ratio was chosen, because at higher COC content, the clay particles aggregated making the separation of the required uniform spreading of about 0.1 g portions of the material onto the surface of the test leachate very cumbersome. SRC test was used as an alternative to SI test and hence for comparison purposes the same COC:bentonite (1:2) ratio

Table 1
Mineralogical and chemical properties of the bentonite.

Smectite content (% of bulk)	73
<0.2 μm content (% of bulk)	52
Smectite in <0.2 μm (%)	98
MB-CEC bulk (mol/kg) ^a	0.85
Ba-CEC <0.2 μm (mol/kg) ^b	1.04
Na^+ (% of bulk CEC) ^c	83
Ca^{2+} (% of bulk CEC) ^c	2
Mg^{2+} (% of bulk CEC) ^c	9
K^+ (% of bulk CEC) ^c	6

^a The methylene blue CEC (MB-CEC) tests were performed on bulk materials by CSIRO Land and Water (Adelaide, South Australia) following the tetra-sodium pyrophosphate (TSPP) pre-treatment method of Wang et al. (1996).

^b The Ba-CEC tests were conducted using X-ray fluorescence on oriented films of the <0.2 μm fractions by CSIRO Land and Water following in-house methods.

^c Calculated from cation released from NH_4^+ displacement at pH 7.0 (Method 15A of Rayment and Higginson, 1992).

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