



# Effect of ammonium on the hydraulic conductivity of geosynthetic clay liners



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## ABSTRACT

Hydraulic conductivity and swell index tests were conducted on a conventional geosynthetic clay liner (GCL) containing sodium-bentonite (Na-B) using 5, 50, 100, 500, and 1000 mM ammonium acetate (NH<sub>4</sub>OAc) solutions to investigate how NH<sub>4</sub><sup>+</sup> accumulation in leachates in bioreactor and recirculation landfills may affect GCLs. Control tests were conducted with deionized (DI) water. Swell index of the Na-B was 27.7 mL/2 g in 5 mM NH<sub>4</sub><sup>+</sup> solution and decreased to 5.0 mL/2 g in 1000 mM NH<sub>4</sub><sup>+</sup> solution, whereas the swell index of Na-B in DI water was 28.0 mL/2 g. Hydraulic conductivity of the Na-B GCL to 5, 50, and 100 mM NH<sub>4</sub><sup>+</sup> was low, ranging from 1.6–5.9 × 10<sup>-11</sup> m/s, which is comparable to the hydraulic conductivity to DI water (2.1 × 10<sup>-11</sup> m/s). Hydraulic conductivities of the Na-B GCL permeated with 500 and 1000 mM NH<sub>4</sub><sup>+</sup> solutions were much higher (e.g., 1.6–5.2 × 10<sup>-6</sup> m/s) due to suppression of osmotic swelling. NH<sub>4</sub><sup>+</sup> replaced native Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup> in the exchange complex of the Na-B during permeation with all NH<sub>4</sub><sup>+</sup> solutions, with the NH<sub>4</sub><sup>+</sup> fraction in the exchange complex increasing from 0.24 to 0.83 as the NH<sub>4</sub><sup>+</sup> concentration increased from 5 to 1000 mM. A Na-B GCL specimen permeated with 1000 mM NH<sub>4</sub><sup>+</sup> solution to chemical equilibrium was subsequently permeated with DI water. Permeation with the NH<sub>4</sub><sup>+</sup> converted the Na-B to “NH<sub>4</sub>-bentonite” with more than 80% of the exchange complex occupied by NH<sub>4</sub><sup>+</sup>. Hydraulic conductivity of this GCL specimen decreased from 5.9 × 10<sup>-6</sup> m/s to 2.9 × 10<sup>-11</sup> m/s during permeation with DI water, indicating that “NH<sub>4</sub>-bentonite” can swell and have low hydraulic conductivity, and that the impact of more concentrated NH<sub>4</sub><sup>+</sup> solutions on swelling and hydraulic conductivity is reversible.

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## 1. Introduction

Geosynthetic clay liners (GCLs) are thin (5–10 mm-thick) factory-manufactured hydraulic barriers consisting of a layer of sodium-bentonite (Na-B) sandwiched between two geotextiles (Shackelford et al., 2000; Meer and Benson, 2007; Scalia and Benson, 2010; Bradshaw and Benson, 2013; Bradshaw et al., 2015; Tian et al., 2016). GCLs are widely used in containment applications due to their low hydraulic conductivity (<10<sup>-10</sup> m/s) to water, thinness, ease of installation, and perceived resistance to environmental distress (Jo et al., 2001; Bradshaw and Benson, 2013; Scalia et al., 2014; Bradshaw et al., 2015; Tian et al., 2016).

The Na-B in a conventional GCL consists mainly of Na-montmorillonite. The low hydraulic conductivity of GCLs is attributed to osmotic swelling of the Na-montmorillonite fraction, which results in narrow and tortuous paths for flow (Jo et al., 2001; Bradshaw and Benson, 2013; Tian et al., 2016). Cations in the exchange complex of the montmorillonite must be primarily monovalent (e.g., Na<sup>+</sup>, Li<sup>+</sup>) and the permeant solution must have an ionic strength no more than 300 mM for osmotic swelling to occur (Norrish and Quirk, 1954; Jo et al., 2001). Norrish and Quirk (1954) report that Na-montmorillonite exhibits osmotic swelling in NaCl solutions with concentration less than 300 mM. Jo et al. (2001) indicate that osmotic swell of Na-B is sensitive to valence of cations (e.g., monovalent vs. divalent), regardless of speciation. Osmotic swelling is suppressed when GCLs are exposed to solutions with high ionic strength or a predominance of divalent cations, which results in larger intergranular pores and higher hydraulic

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conductivity (Jo et al., 2001; Kolstad et al., 2004; Meer and Benson, 2007; Scalia et al., 2014; Tian et al., 2016).

Several studies have investigated the hydraulic conductivity of Na-B GCLs to municipal solid waste (MSW) leachates (Petrov and Rowe, 1997; Ruhl and Daniel, 1997; Rauen and Benson, 2008; Guyonnet et al., 2009; Bradshaw and Benson, 2013; Bradshaw et al., 2015). Bradshaw and Benson (2013) report that the hydraulic conductivity of Na-B GCLs permeated with conventional MSW leachates can be as much as 6 times higher than the hydraulic conductivity to deionized (DI) water. MSW leachates have modest impact on hydraulic conductivity because they have relatively low ionic strength (typical = 80 mM) and a high fraction of monovalent cations (e.g.,  $\text{Na}^+$  and  $\text{NH}_4^+$ ). Bradshaw et al. (2015) evaluated the long-term hydraulic conductivity of Na-B GCLs to leachates from bioreactor or recirculation landfills. The  $\text{NH}_4^+$  fractions in these leachates ranged from 0.082 to 0.43 [i.e., charge ratio,  $[\text{NH}_4^+]/[\text{Na}^+ + \text{NH}_4^+ + \text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+]$ ] and the ionic strength of these leachates ranged from 95 to 187 mM. Hydraulic conductivity of the Na-B GCLs to leachates from the bioreactor and recirculation landfills ( $1.0\text{--}2.0 \times 10^{-11}$  m/s) was comparable to hydraulic conductivity of the GCL to DI water ( $2.2 \times 10^{-11}$  m/s). Exchange complex analysis of Na-B in the GCLs after permeation illustrated that the native cations in the exchange complex were replaced by  $\text{NH}_4^+$ .

Benson et al. (2007) and Barlaz et al. (2010) report that  $\text{NH}_4^+$  accumulates in leachates from bioreactor and recirculation landfills over time. They report a maximum  $\text{NH}_4^+$  concentration of 89 mM (computed assuming the  $\text{NH}_3\text{-N}$  in Barlaz et al., 2010 was in the form of  $\text{NH}_4^+$ ), but also indicate an increasing trend in the total nitrogen concentration and attribute this to absence of a denitrification pathway under anaerobic conditions. Elevated  $\text{NH}_4^+$  concentration can induce cation exchange when GCLs are permeated by leachates, resulting in a higher fraction of  $\text{NH}_4^+$  in the exchange complex and transforming the Na-B to a “ $\text{NH}_4\text{-B}$ ” (Pivato and Raga, 2006; Guyonnet et al., 2009; Bradshaw et al., 2015). Gautier et al. (2010) indicate that only one layer of water molecules forms in the interlayer space during hydration of  $\text{NH}_4\text{-montmorillonite}$ , reducing swelling of  $\text{NH}_4\text{-montmorillonite}$  compared to Na-B. Accumulation of  $\text{NH}_4^+$  also increases the ionic strength of the leachate, resulting in reduced osmotic swelling (Norrish and Quirk, 1954; Jo et al., 2001). This reduction in swelling of the bentonite potentially can lead to higher hydraulic conductivity (Jo et al., 2001, 2005; Scalia and Benson, 2010; Tian et al., 2016).

This study was conducted to evaluate the sensitivity of hydraulic conductivity to  $\text{NH}_4^+$  concentration. A commercially available Na-B GCL was permeated with 5, 50, 100, 500, and 1000 mM ammonium acetate ( $\text{NH}_4\text{OAc}$ ) solutions specifically to evaluate the impact of  $\text{NH}_4^+$  on the hydraulic conductivity of conventional Na-B GCLs, recognizing that actual leachates will contain other cations that may also alter hydraulic conductivity. Swell index tests were conducted on the bentonite with the same solutions to determine how  $\text{NH}_4^+$  concentration affects swelling. The exchange complex of the bentonite was evaluated before and after permeation to assess cation exchange associated with  $\text{NH}_4^+$  in the permeant solutions.

## 2. Background

### 2.1. Bentonite exchange complex, swelling, and hydraulic conductivity of GCLs

Na-B used in GCLs consists primarily of Na-montmorillonite, which has high cation exchange capacity (CEC), large specific surface area, and high swelling capacity (Grim, 1968; Bradshaw and Benson, 2013; Tian et al., 2016). Swelling of Na-B occurs primarily in the interlayer of the Na-montmorillonite in two phases: the crystalline phase and the osmotic phase (Norrish and Quirk, 1954;

Jo et al., 2001; Kolstad et al., 2004; Bouazza and Bowders, 2010; Scalia et al., 2014; Tian et al., 2016). During crystalline swelling, up to four discrete layers of water molecules associate with the mineral surface depending on the cation species in exchange complex. Osmotic swelling follows crystalline swelling, and only occurs if monovalent cations (e.g.,  $\text{Na}^+$ ,  $\text{Li}^+$ ) are predominant in the exchange complex (Kolstad et al., 2004; Scalia et al., 2014; Tian et al., 2016). The magnitude of osmotic swelling is inversely proportional to the ionic strength of the pore water (Norrish and Quirk, 1954).

Na-montmorillonite is susceptible to cation exchange because polyvalent cations are preferred relative to monovalent cations to satisfy the negative charge on the montmorillonite surface, as indicated in the lyotropic series (low to high preference):  $\text{Li}^+ < \text{Na}^+ < \text{K}^+ < \text{NH}_4^+ < \text{Mg}^{2+} < \text{Ca}^{2+} < \text{Al}^{3+}$  (Domenico and Schwartz, 1998). Since  $\text{Na}^+$  is near the lower preference end of the lyotropic series, the native  $\text{Na}^+$  in Na-B is frequently replaced by other cations when permeated with solutions containing polyvalent cations (e.g.,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) (Bradshaw and Benson, 2013; Scalia et al., 2014; Bradshaw et al., 2015; Tian et al., 2016).

For these reasons, permeant solutions having high ionic strength or predominantly divalent cations can suppress osmotic swelling of Na-B, resulting larger pores and higher hydraulic conductivity (Jo et al., 2001, 2005; Kolstad et al., 2004; Meer and Benson, 2007). For example, Chen et al. (2015) report hydraulic conductivities from  $5.4 \times 10^{-8}$  to  $1.2 \times 10^{-6}$  m/s for two conventional Na-B GCLs permeated with trona ash leachate, which has high ionic strength (780 mM) and suppresses osmotic swell (swell index = 8–10 mL/2 g). For the same GCLs, the hydraulic conductivity to DI water ranged from  $2.66 \times 10^{-11}$  to  $3.6 \times 10^{-11}$  m/s at the same effective confining stress (20 kPa) and the swell index was approximately 28–36 mL/2 g. Similarly, Tian et al. (2016) report that the Na-B in conventional GCLs permeated with synthetic low-level radioactive waste leachate containing primarily  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  cations was transformed to a Ca-Mg-B GCL with hydraulic conductivity approximately 10–20 times higher than hydraulic conductivity to DI water ( $1.3\text{--}2.8 \times 10^{-10}$  m/s vs.  $8.4 \times 10^{-12}\text{--}2.2 \times 10^{-11}$  m/s).

Jo et al. (2001) and Kolstad et al. (2004) indicate that the hydraulic conductivity of GCLs is inversely proportional to the swell index of Na-B. Key parameters affecting swell index of Na-B include ionic strength, relative abundance of monovalent cations and polyvalent cations, and pH of the permeant solution (Jo et al., 2001; Kolstad et al., 2004). Ionic strength ( $I$ ) is defined as:

$$I = \frac{1}{2} \sum_{i=1}^n C_i Z_i^2 \quad (1)$$

where  $n$  is the number of ionic species,  $C_i$  is the concentration of the  $i$ th species, and  $Z_i$  is the valence of  $i$ th species. Kolstad et al. (2004) quantified the relative abundance of monovalent and polyvalent cations using the parameter RMD:

$$\text{RMD} = \frac{M_m}{\sqrt{M_d}} \quad (2)$$

where  $M_m$  = total molar concentration of monovalent cations and  $M_d$  = total molar concentration of polyvalent cations.

Kolstad et al. (2004) indicate that the hydraulic conductivity of GCLs increases as ionic strength increases or RMD decreases, regardless of cation species. For a given ionic strength, GCLs permeated with solutions having lower RMD (i.e., polyvalent cations predominant in permeant solution) have a higher hydraulic conductivity (Kolstad et al., 2004). In addition, the hydraulic

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