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Novel bentonites for containment barrier applications

Bentonites novatrices pour des applications comme barriers de confinement

Bohnhoff G., Shackelford C.
Colorado State University, Fort Collins, Colorado, USA

Malusis M.
Bucknell University, Lewisburg, Pennsylvania, USA

Scalia J., Benson C., Edil T.
University of Wisconsin, Madison, Wisconsin, USA

Di Emidio G.
Ghent University, Zwijnaarde, Belgium

Katsumi T.
Kyoto University, Kyoto, Japan

Mazzieri F.
Universita` Politecnica delle Marche, Ancona, Italy

ABSTRACT: Sodium bentonite (Na-bentonite) commonly is used in geoenvironmental containment barriers to control liquid flow and contaminant migration, but is known to be thermodynamically unstable in environments where multivalent cations are present. As a result, a host of novel, chemically-modified bentonites designed for improved resilience have been developed. The objective of this paper is to illustrate selected properties of some of these bentonites being considered for containment barrier applications. The bentonites considered in this paper include Na-bentonite polymerized with acrylic acid (bentonite-polymer nanocomposite, or BPN), Na-bentonite amended with sodium carboxymethyl cellulose (HYPER clay), and a propylene carbonate modified Na-bentonite (multiswellable bentonite, or MSB). Engineering properties of the novel bentonites relevant to two specific types of barriers (i.e., cutoff walls and geosynthetic clay liners) are compared and contrasted with those of natural Na-bentonites. The results illustrate the potential for improved hydraulic performance of barriers containing the novel bentonites. However, further research is needed elucidate the mechanisms responsible for differences in behavior and performance among these bentonites.

RÉSUMÉ : La bentonite à base de sodium (bentonite-Na) est utilisée couramment comme barrière de confinement géo-environnementale pour contrôler les flux de liquides et la migration de contaminants ; elle est cependant connue pour être instable thermodynamiquement dans des environnements qui contiennent des cations multivalents. Pour palier cet inconvénient, diverses bentonites novatrices modifiées chimiquement et conçues pour présenter une résistance améliorée aux écoulements ont été développées. L'objectif de cet article est d'illustrer certaines propriétés techniques de quelques unes de ces bentonites étudiées leur utilisation dans les barrières de confinement. Cet article présente les propriétés de la bentonite-Na polymérisée avec de l'acide acrylique (« bentonite-polymer nanocomposite », ou BPN), de la bentonite-Na modifiée avec de la carboxyméthylcellulose sodique (ou « HYPER Clay ») et d'une bentonite-Na modifiée avec du carbonate de propylène (« multiswellable bentonite », ou MSB). On a procédé à une analyse comparative des propriétés des bentonites novatrices correspondant à deux types spécifiques de barrières (c.a.d., parois de séparation et revêtement par des géomembranes synthétiques) et celles des bentonites-Na naturelles. Les résultats illustrent le potentiel d'amélioration de la performance hydraulique que présentent des barrières contenant les nouvelles bentonites. Des recherches supplémentaires sont cependant nécessaires pour comprendre les mécanismes susceptibles d'expliquer les différences de comportement et de performance entre ces bentonites.

KEYWORDS: Bentonite; Bentonite slurry; Cutoff wall; Geosynthetic clay liner; Hydraulic conductivity; Polymerized bentonite

1 INTRODUCTION

Bentonite is a natural clay composed primarily of the mineral montmorillonite, a member of the smectite group of clay minerals. Montmorillonite is characterized by large surface area (100-800 m^2/g), a net negative charge typically on the order of 80 to 150 $cmol_c/kg$, and exchangeable surface cations (Grim 1968). These characteristics, in turn, impart a strong affinity by bentonite for water, resulting in swelling, sealing, and adhesive characteristics (Eisenhour and Brown 2009).

Bentonite commonly is used to control flow (seepage) and contaminant transport in a variety of hydraulic containment applications, such as soil-bentonite (SB) groundwater cutoff walls, barriers for waste containment (e.g., landfills, wastewater ponds, manure lagoons, nuclear storage, etc.), secondary containment in petroleum tank farms, and seals in monitoring and water supply wells. In all of these applications, sodium bentonite (Na-bentonite) typically is used, meaning that sodium (Na^+) is the predominant cation on the exchange sites of the individual bentonite particles. The preference for Na-bentonite stems from desirable engineering properties, such as low hydraulic conductivity to water, k_w (typically < 10^{-10} m/s), and the existence of semipermeable membrane behavior, the latter giving rise to hyperfiltration, chemico-osmotic flow, and reduced diffusion (Malusis et al. 2003). Unfortunately, Na-bentonite is thermodynamically unstable in environments where multivalent cations (e.g., Ca^{2+}, Mg^{2+}) are present, including most naturally occurring pore waters as well as waste streams containing heavy metals (e.g., landfill leachates, acid mine drainage) or radionuclides. Under such conditions, multivalent

cations replace exchangeable monovalent cations, thereby reducing or eliminating osmotic swelling of the bentonite and the ability of bentonite to function effectively (e.g., Gates et al. 2009).

In response to the susceptibility of natural Na-bentonites to chemical attack and poor hydraulic performance, recent research has been undertaken to investigate novel bentonites that have been chemically modified to achieve greater compatibility with the surrounding environment, such that the desirable engineering properties of the bentonites are not compromised. For example, anionic polymers have been added to Na-bentonite to attain rheological properties needed for drilling fluids, and organobentonites (i.e., bentonites with organic cations on the exchange sites) have been created for applications where an organophilic material is needed for containment of organic compounds (e.g., Lee et al. 2012). Also, Na-bentonites complexed with various organic chemicals (e.g., polymers) have attracted considerable interest as potential substitutes for natural Na-bentonites in containment barriers, such as SB cutoff walls and geosynthetic clay liners (GCLs) (e.g., Katsumi et al. 2008, Mazzieri et al. 2010). The objective of this paper is to illustrate the potential improvement in engineering properties that can be attained with some of these novel bentonites when used in containment barriers. This objective is achieved by comparing engineering properties of the novel bentonites, including hydraulic conductivity to water and chemical solutions, with those of natural Na-bentonites.

2 NOVEL BENTONITES

Three novel bentonites are considered herein. These bentonites include (1) a natural Na-bentonite polymerized with acrylic acid to form polyacrylate polymerized bentonite, referred to as a bentonite polymer nanocomposite or BPN, (2) a propylene carbonate (PC) modified Na-bentonite, referred to as "multiswellable bentonite" or MSB, and (3) Na-bentonite amended with sodium carboxymethyl cellulose (Na-CMC), referred to as "HYPER clay" or HC.

The BPN was supplied by Colloid Environmental Technologies Co. (CETCO, USA), and was produced with polyacrylic acid (PAA) using methods similar to those used for the production of polymer nanocomposites (Bohnhoff 2012, Scalia 2012). A monomer solution was prepared by dissolving acrylic acid in water, and then the solution was neutralized with sodium hydroxide. Next, a natural (unmodified) Na-bentonite was added to the monomer solution in concentrations ranging from 30 to 50 % by weight to form bentonite-monomer slurry, followed by the addition of sodium persulfate, which served as an initiator. Polymerization then was induced by raising the temperature of the slurry above the decomposition temperature of the initiator molecule. Following polymerization, the PAA polymerized bentonite was oven dried, milled, and screened.

The MSB, supplied by Hojun Corp. (Japan), was created by compounding Na-bentonite with PC that expands the clay lattice and forms a hydration shell around the interlayer cations. The resulting Na-bentonite-PC complex can undergo osmotic swell in both fresh water and electrolyte solutions (hence the term "multiswellable"), including sea water (~500 mM NaCl) and solutions containing multivalent cations (e.g., Onikata et al. 1999). The MSB contained 25 % PC by dry weight.

The HC was created by combining Na-bentonite with Na-CMC, the sodium salt form of the anionic polymer CMC that has been used in industrial applications as a thickener (Di Emidio 2010). The base Na-bentonite was combined with a Na-CMC solution to form slurry. The slurry was dried at 105 °C, and the resulting solids were ground with a mortar grinder. The Na-CMC penetrates the interlayer regions between the clay platelets, resulting in expansion of the clay lattice and enhanced osmotic swell. Test results for HYPER clay containing either 2 % Na-CMC or 8 % Na-CMC are presented herein, where the former is designated as HC2 and the latter is designated as HC8. In both cases, the base Na-bentonite was the same base Na-bentonite used to create the MSB (designated herein as NB3).

The index properties of the novel bentonites (BPN, HC2, HC8, and MSB) are summarized in Table 1 and compared with those of two natural Na-bentonites designated as NB1 (NaturalGel®, Wyo-Ben, Inc., USA) and NB2 (Volclay®, American Colloid Company, USA) and the base Na-bentonite, NB3, used to create HC and MSB. Both NB1 and NB2 are commonly used for SB cutoff walls. The unusual behavior that can be exhibited by polymer modified bentonites is illustrated in the case of the BPN, wherein the BPN exhibits the highest free swell and cation exchange capacity (*CEC*), despite the lowest liquid limit (*LL*) and an overall nonplastic behavior. This unusually high swell and *CEC* have been attributed to the high swelling potential of the hydrophilic polymer used in the BPN (also used in baby diapers) and the tendency of the polymer to bind cations (Bohnhoff 2012, Scalia 2012). In contrast, the *CECs* for HC2, HC8, and MSB are only marginally higher than that of NB3. However, HC2, HC8, and MSB exhibited markedly higher *LLs* relative to NB3, presumably due to the addition of Na-CMC and PC, respectively.

Table 1. Index properties of bentonites examined herein (NB1 = NaturalGel®; NB2 = Volclay®; NB3 = Hojun Na-Bentonite; BPN = Bentonite Polymer Nanocomposite; HC2 and HC8 = HYPER Clay with 2 % and 8 % Na-CMC, respectively; MSB = Multiswellable Bentonite).

Property	Standard	Value						
		NB1	NB2	NB3	BPN	HC2	HC8	MSB
Soil classification	ASTM D 2487	CH	CH	CH	CH	CH	CH	CH
Liquid limit (%)	ASTM D 4318	583	420	466	255	650	742	547
Plasticity index (%)		530	381	421	NP	594	681	502
Distilled water swell index (mL/2 g)	ASTM D 5890	35	32	26	73	37[c]	48[c]	29
Montmorillonite content (%)	a	69	91	77	76	78	78	74
Cation exchange capacity, *CEC* (cmol$_c$/kg)	b	83.4	78.0	44.5	143	47.3	46.7	49.8
Exchangeable metals (cmol$_c$/kg):	b							
Ca^{2+}		4.9	28.1	5.6	9	11.4	12.7	7.7
Mg^{2+}		8.8	13.3	7.9	3	6.2	5.5	6.1
Na^+		73.4	34.3	26.3	128	34.2	44.5	33.3
K^+		1.1	1.6	0.2	3	0.3	0.2	0.5
Sum		88.2	77.3	40.0	143	52.1	62.9	47.6

[a] Based on energy dispersive X-ray diffraction analysis;
[b] Procedures for NB1, NB2, and MSB given by Shackelford and Redmond (1995); procedures for BPN given by Scalia (2012); procedures for HC2 and HC8 given by Di Emidio (2010);
[c] Values likely underestimated (see Di Emidio 2010).

3 RESULTS

3.1 *Soil-Bentonite Vertical Cutoff Walls*

Soil-bentonite (SB) cutoff walls are commonly constructed in the US using the slurry trench method in which a trench is excavated and filled with bentonite-water slurry (typically 4-6 % bentonite) to maintain trench stability, the trench spoils are mixed with dry bentonite (as needed) and slurry to create a homogeneous, high-slump SB backfill, and the backfill is placed into the trench to create the wall. The slurry viscosity must be sufficiently high to maintain trench stability, yet sufficiently low to be easily displaced by the backfill. The slurry also should form an adequate filter cake along the trench sidewalls to minimize slurry loss during construction. Recommended slurry properties include a Marsh viscosity of 32-40 s and a filtrate loss of < 25 mL (Evans 1993). Also, the backfill must exhibit a low hydraulic conductivity, typically ≤ 10^{-9} m/s for geoenvironmental containment applications.

The influence of bentonite content on the Marsh viscosity and filtrate loss (API 13A-B) of slurry containing untreated bentonite (NB1, NB2) or treated bentonite (HC8, BPN, MSB) is illustrated in Fig. 1. Slurries containing 3-5 % NB1, NB2, or MSB exhibit viscosities within the range of 32-40 s (Fig. 1a). For these clays, a bentonite content of 5 % likely would be selected to obtain a greater slurry density and reduce filtrate loss (Fig. 1b). In contrast, the viscosities of slurries containing ≥ 3 % HC8 or BPN were > 40 s and increased drastically with increasing bentonite content due to thickening caused by the polymer. Thus, slurry containing 2 % HC8 or BPN would be appropriate for slurry trench construction based on viscosity. Finally, the filtrate losses for 2 % HC8 and BPN are equal to or lower than the filtrate losses for 5 % MSB or NB1.
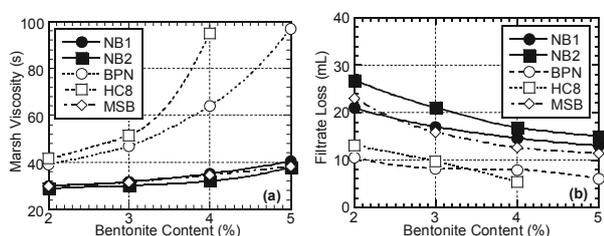

Figure 1. Properties of bentonite-water slurries as a function of bentonite content: (a) Marsh viscosity; (b) filtrate loss (NB1 and MSB data from Malusis et al. 2010; NB2 and BPN data from Bohnhoff 2012).

Hydraulic conductivity and chemical compatibility of model SB backfills comprised of sand and NB1, NB2, MSB, or BPN were investigated in recent studies by Malusis and McKeehan (2012) and Bohnhoff (2012). Although the sands used in both studies were clean and poorly graded, the sand used by Malusis and McKeehan (2012) in the NB1 and MSB backfills was a fine sand (D_{50} = 0.20 mm) whereas the sand used by Bohnhoff (2012) in the NB2 and BPN backfills was a medium sand (D_{50} = 0.45 mm). In both studies, the specimens were tested in flexible-wall cells at low confining stresses (≤ 34.5 kPa). The specimens were permeated with tap water until a steady k_w was achieved, and then were permeated with CaCl$_2$ solutions (5-1,000 mM) until termination criteria for chemical equilibrium between the influent and effluent were achieved (see cited references for further details).

The measured k_w values from these studies are presented in Fig. 2a along with k_w values measured recently at Bucknell University for sand-bentonite backfill specimens containing HC8 and the same fine sand used by Malusis and McKeehan (2012). The backfills containing the polymer-modified bentonites (BPN or HC8) generally exhibited lower k_w relative to the backfills containing similar percentages of MSB or Na-bentonite (NB1 or NB2), indicating that less BPN or HC8 is

needed to create backfill with an acceptable k_w (i.e., ≤ 10^{-9} m/s).

The influence of CaCl$_2$ on the k of backfill specimens containing 5.7 % NB1 or 5.6 % MSB (Malusis and McKeehan 2012) and specimens containing 7.1 % NB2, 2.4 % BPN, and 5.5 % BPN (Bohnhoff 2012) is shown in Fig. 2b. All of the specimens were susceptible to an increase in k, i.e., $k_c/k_w > 1$, where k_c = hydraulic conductivity to the CaCl$_2$ solution, when permeated with ≥ 10 mM CaCl$_2$ solutions. The increases varied from approximately two-fold to 15-fold depending, in part, on the bentonite content. For example, the specimen containing the most bentonite (7.1 % NB2) exhibited the highest k_c/k_w (~15) of all the specimens. Also, the 2.4 % BPN specimen exhibited a lower k_c/k_w relative to the 5.5 % BPN specimen permeated with the same CaCl$_2$ solution (50 mM). However, k_w for the 5.5 % BPN backfill (2 x 10^{-12} m/s) was well below the typical regulatory limit (10^{-9} m/s), whereas k_w for the 2.4 % BPN backfill was unacceptably high (10^{-7} m/s; see Fig. 2a). Thus, the lower BPN content (2.4 %) was advantageous in terms of chemical compatibility, but was insufficient for achieving regulatory compliance in terms of k.

For the specimens with similar bentonite contents (i.e., 5.7 % NB1, 5.6 % MSB, and 5.5 % BPN), the 5.6 % MSB specimens exhibited the greatest resilience. The higher values of k_c/k_w for the 5.5 % BPN specimens relative to 5.6 % MSB and 5.7 % NB1 specimens permeated with the same CaCl$_2$ solution were attributed to two primary factors, viz., the greater reactivity of the 5.5 % BPN specimens, as reflected by the lower k_w for this backfill relative to those containing 5.6 % MSB or 5.7 % NB1 (see Fig. 2a), and the use of a coarser (i.e., more permeable) sand in the BPN backfills relative to the MSB and NB1 backfills. However, the lower k_w for the 5.5 % BPN backfill also allowed for a greater increase in k to occur without exceeding the typical regulatory limit of 10^{-9} m/s.
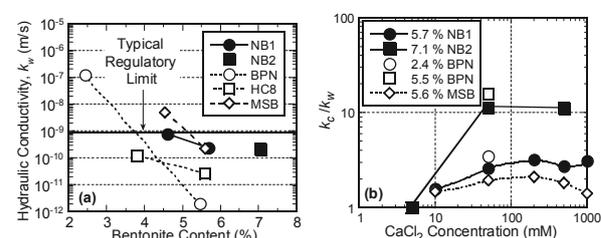

Figure 2. Permeation results for sand-bentonite backfills: (a) hydraulic conductivity to water, k_w, as a function of bentonite content; (b) ratio of hydraulic conductivity to CaCl$_2$ solution, k_c, relative to k_w as a function of CaCl$_2$ concentration (NB1 and MSB data from Malusis and McKeehan 2012; NB2 and BPN data from Bohnhoff 2012).

3.2 *Geosynthetic Clay Liners (GCLs)*

Values of k_c for BPN, HC2, and MSB specimens representing a typical GCL are shown in Fig. 3a. Data for specimens of Na-bentonite taken from actual GCLs (Bentomat® DN, CETCO, USA) are included in Fig. 3a for comparison. All specimens were permeated in flexible-wall cells under low effective stresses (14 to 30 kPa) until the hydraulic termination criteria of ASTM D 5084 were satisfied. Also, most of the specimens were permeated until chemical equilibrium (defined as the ratio of outflow and inflow electrical conductivity within 1.0 ± 0.1) was achieved, with the exceptions being the specimens permeated with deionized water (DIW) and the HC2 specimens. The results reflect a "worst-case" testing condition in that the specimens were not prehydrated prior to permeation (Shackelford et al. 2000). Permeation with DIW (plotted at 0.1 mM CaCl$_2$ in Fig. 3a) resulted in low k_w (i.e., 4.2 x 10^{-12} to 3.4 x 10^{-11} m/s) regardless of the bentonite type. However, the BPN, HC2, and MSB exhibited superior hydraulic behavior (i.e., lower k_c) relative to the GCL bentonites. These results illustrate the potential advantage of novel bentonites in solutions typically

incompatible with conventional GCLs. Although higher $CaCl_2$ concentrations resulted in $k_c > k_w$ for HC2 and MSB, the increases were lower than those for the GCL bentonites. The BPN specimens exhibited similar increases in k_c relative to k_w for 5-50 mM $CaCl_2$, but exhibited $k_c < k_w$ for 500 mM $CaCl_2$.

The relationship between k and swell index, SI (ASTM D 5890), in the same solution (DIW or $CaCl_2$) for the bentonites in Fig. 3a is illustrated in Fig. 3b. The treated bentonites generally exhibited lower k for a given SI than the GCL bentonites, including MSB and HC2, which exhibit superior hydraulic behavior because of the activation of osmotic swelling (Di Emidio et al. 2011). However, the BPN exhibited both a low k_c and a low SI (8 mL/2 g) in 500 mM $CaCl_2$, illustrating that the hydraulic behavior of BPN was decoupled from swell. The BPN exhibited higher swelling in DIW relative to the other bentonites (see Table 1) due to the presence of the super-swelling polymer (Scalia 2012). However, polymer swelling does not account for the low k_c of BPN permeated with 500 mM $CaCl_2$, given the SI of only 8 mL/2 g. This atypical behavior of BPN illustrates that SI is not necessarily an accurate indicator of hydraulic conductivity for chemically modified bentonites.
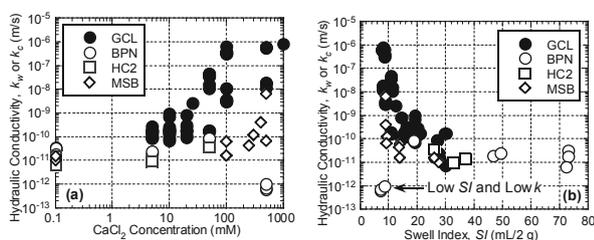

Figure 3. Hydraulic conductivity of bentonite specimens as a function of (a) $CaCl_2$ concentration in the permeant liquid, and (b) swell index in the same solution (GCL data from Jo et al. 2001, Jo et al. 2005, Lee and Shackelford 2005, Lee et al. 2005; MSB data from Lin and Benson 2000, Katsumi et al. 2008; HC2 data from Di Emidio et al. 2011; BPN data from Scalia 2012).

4 CONCLUSIONS

The hydraulic conductivity to water (k_w) and $CaCl_2$ solutions (k_c) of three novel (chemically modified) bentonites (BPN, HC, MSB) were compared with those of three natural Na-bentonites commonly used in geoenvironmental containment applications. The overall hydraulic performance is a function of not only the magnitude of chemical resistance (k_c/k_w) but also the baseline value of k_w. In terms of SB backfills, the use of BPN or HC8 generally exhibited lower k_w compared to MSB or Na-bentonites, implying that less BPN or HC8 is required to create backfills with an acceptable k_w. However, the backfill containing 5.5 % BPN exhibited the highest k_c/k_w, whereas the backfill containing 5.6 % MSB indicated low k_w and a k_c/k_w that was lower than that for the 5.5 % BPN backfill. Overall, the product of k_w and k_c/k_w for the backfills containing 5.5 % BPN and 5.6 % MSB resulted in superior hydraulic performance in terms of the providing the lowest values of k_c. In terms of GCLs, all three novel bentonites (BPN, HC, MSB) showed not only low k_w but also far superior resistance to chemical attack than the natural bentonites. Thus, the potential use of chemically modified bentonites in applications involving SB backfills and GCLs is promising. However, the differences in behaviors among the novel bentonites illustrated in this paper highlight the need for further research into the specific mechanisms affecting the performance of such novel bentonites.

5 ACKNOWLEDGEMENTS

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