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HYDRAULIC PERFORMANCE OF UNTREATED AND POLYMER-TREATED BENTONITE IN INORGANIC LANDFILL LEACHATES

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Abstract—Short- and long-term exposure to inorganic solutions can cause significant degradation of the hydraulic properties of bentonite clay used in geosynthetic clay liners (GCLs). In particular, the increase in hydraulic conductivity due to cation exchange when Na-montmorillonite is subjected to leachates rich in Ca and Mg has caused problems in incinerator ash landfill liners located in wet environments, where large quantities of leachates are generated. Experimental results are presented to evaluate the immediate change in hydraulic conductivity of seven types of GCL clays upon permeation with leachate generated from three ash landfills. The composition of the ash, which is a by-product of the incineration of municipal solid waste (MSW), in turn influences the composition of the resulting leachate. Falling head permeability tests were performed on flexible-wall permeameter specimens, with back-pressure saturation. Chemical analysis shows that the three leachate products contain high, medium, and low concentration Ca and Mg cations. The clay component of GCL materials tested in this study consists of regular and polymer-treated bentonite. Polymer treatment is believed to render the clay non-reactive to many organic and inorganic chemicals. The results of this study indicate that: (1) polymer treatment is generally more beneficial if the clay is first saturated with water and not directly with the leachate; (2) high swell potential of the bentonite is more advantageous than polymer treatment, especially when low hydraulic conductivity is required in the short term and if the clay is pre-hydrated. Experiment setup and special specimen preparation procedures are also discussed.

Key Words—Bentonite, Contaminant Resistant Clay, Geosynthetic Clay Liner, Hydraulic Conductivity, Landfill Leachate, Montmorillonite, Permeability, Polymer Treatment, Swell Index X-ray Diffraction.

INTRODUCTION

In regions where natural low-permeability clay deposits are not available, geosynthetic clay liners (GCLs) may be used for landfill liner and cover systems to replace or augment natural clays as long as the selected GCL is chemically compatible with the liquid being contained. Unless a geomembrane sheet is present, the low hydraulic conductivity of GCLs is solely attributed to the clay component, which consists typically of bentonite or similar low-permeability clays. Mechanical or chemical methods are used to bond the clay component of the GCL to the adjacent geotextile or geomembrane sheets.

The clay component of a GCL material may consist of either untreated or polymer-treated bentonite. In MSW landfills where the inorganic or organic contaminant concentration in the leachate is low, lining systems can rely on natural or untreated bentonites (Egloffstein, 1995; Ruhl and Daniel, 1997). Such GCL bentonites typically contain natural Na-montmorillonite, although

Ca-montmorillonite is sometimes used. In regions where leachates with high concentrations of contaminant are present, the use of polymer-treated or polymer-coated bentonite is beneficial, as it arguably renders the montmorillonite non-reactive towards most organic and inorganic chemical compounds. Laboratory test results (Elhajji *et al.*, 2001) suggest that polymer-treated GCLs maintain their low hydraulic conductivity when hydrated with liquids containing single-species solutions at low concentration. In this paper, laboratory data are presented for seven untreated and treated GCL bentonites permeated with natural landfill leachate solutions with different concentrations. The leachates were obtained from three MSW landfills where large quantities of incinerator ash rich in Ca and Mg are disposed.

BENTONITES IN GEOSYNTHETIC CLAY LINERS

Bentonites, a key component in commercially manufactured GCLs, are natural clays formed as a result of mechanical and chemical weathering of volcanic ash that has been deposited in salt or fresh water. Na- and Ca-montmorillonites, which constitute the main minerals in bentonite clays, are present in salt- and fresh-water

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deposits, respectively. Bentonite exhibits low hydraulic conductivity (up to 10^{-10} cm/s), high fluid adsorption capacity, and can swell to several times its original volume. Its swelling capacity depends on factors such as mineral composition, grain size, aggregate size, cation exchange capacity (CEC), chemical concentration of the permeating liquid, and chemical composition of the first wetting liquid. Extrinsic factors that affect the hydraulic conductivity of bentonite include, among other things, confining pressure, laboratory pressure-saturation conditions, and hydraulic gradient (Shackelford *et al.*, 2000).

The hydraulic conductivity of bentonite can increase gradually, albeit by a significant amount, due to alterations in its micro-fabric, stemming from a decrease in the diffuse double layer thickness. Such a decrease in thickness can occur upon increasing the solution concentration, or as a result of the replacement of monovalent ions (*e.g.* Na^+) with higher valence cations present in an intruding inorganic solution. Changes in the bentonite hydraulic conductivity by several orders of magnitude have been reported in the literature upon permeation with high-concentration solutions (*e.g.* Petrov *et al.*, 1997; Shackelford *et al.*, 2000; Elhajji *et al.*, 2001).

Recently, several GCL manufacturers introduced products such as contaminant-resistant clays (CRC) or polymer-treated bentonites (PTB) to minimize bentonite degradation upon exposure to contaminants. Polymer treatment processes and formulae used in GCL manufacturing are proprietary and are not documented (Ruhl and Daniel, 1997; Kajita, 1997). According to Theng (1979), clay-polymer interactions can be classified according to the polymer's surface charge; uncharged polymers are electrically neutral whereas anionic and cationic polymers carry net negative and positive surface charges, respectively. The chemical modification mechanism in bentonite clays, which involves the use of cationic polymers, has been described extensively in the literature since the 1970s (*e.g.* Pezerat and Vallet, 1973; Bart *et al.*, 1979).

Table 1. Bentonite properties, as supplied by the manufacturers.

Manufacturer	Treatment	Label	Swell index ^a mL/2 g	Hydraulic conductivity ^b cm/s
A	Untreated	AU	24	5×10^{-9}
A	Treated	AT	24	N/A
B	Untreated	BU	24	5×10^{-9}
B	Treated	BT	24	1×10^{-9}
C	Untreated	CU	24	5×10^{-9}
C	Treated	CT1	24	5×10^{-9}
C	Treated	CT2	24	5×10^{-9}

^a ASTM D 5890

^b ASTM D 5887, based on typical GCL thickness

Two mechanisms exist by which polymer treatment enhances the resistance of bentonite to cation exchange, the main cause of flocculation which, in turn, results in an increase in hydraulic conductivity. The first mechanism involves the replacement, during manufacturing, of the adsorbed Na or Ca ions in the clay by a cationic polymer. This process is irreversible because a single polymer chain contains thousands of cations which would need to be displaced simultaneously if cation exchange is to take place later. The second mechanism relies on a weaker bond (dipole attraction) between the cationic polymer and the Na ions. In this case, Na ions are not replaced, but rather the Na-montmorillonite sheets are 'coated' with the polymer.

EXPERIMENTAL PROGRAM

Description of clays and leachates

In the present study, seven bentonite clays, produced by three GCL manufacturers, were used. Three were described by the manufacturers as 'polymer-treated', although the exact treatment process was not disclosed. Some of the samples were obtained directly in the form of a dry clay powder from the manufacturer while others were extracted for the testing program by cutting a sample of the parent GCL material and extracting the clay component. Table 1 contains a description of the seven bentonites with the nominal properties provided by the manufacturers. The principal mineral in all seven clays is Na-montmorillonite.

The leachates were obtained from incinerator ash disposal facilities in three different counties in the State of Florida, USA. One of the facilities is an ash monofill while the other two facilities use landfill co-disposal where both incinerator ash and MSW are disposed. Table 2 summarizes the relevant chemical characteristics of the leachates. All three leachates were found to possess high levels of Ca and Mg compared to other multivalent cations (*e.g.* Al, Cu and Zn). The abundance of Ca and Mg in the form of chloride, bicarbonate and sulfide, has been traced back to the chemicals added during the waste incineration process. Because of the aggressive nature of the ash chemistry, the concentrations reported here are significantly higher than those reported by earlier researchers for real and simulated MSW leachates and salt solutions (*e.g.* Ruhl and Daniel, 1997; Shackelford *et al.*, 2000; Jo *et al.*, 2001)

Specimen preparation and test parameters

Falling-head permeability tests with back-pressure saturation were performed following the ASTM D 5887 procedure. The single exception entailed the preparation of specimens from the clay component only rather than the intact GCL with the geosynthetic backing. For consistency purposes, 60 g of dry bentonite were tamped to a constant thickness of 6 mm in a specially fabricated mold to produce the 100 mm diameter specimens, which

Table 2. Relevant leachate properties.

Leachate	L-1	L-2	L-3
Landfill type	Co-disposal	Co-disposal	Ash monofill
pH	6.30	6.55	7.24
Cations (mg/L)			
Ammonia, NH ₄ ⁺	60	260	15
Sodium, Na ⁺	2,200	300	1,640
Calcium, Ca ²⁺	1,150	2,625	5,120
Magnesium, Mg ²⁺	750	1,525	2,075
Anions (mg/L)			
Chloride, Cl ⁻	5,300	3,800	6,836
Bicarbonate, HCO ₃ ⁻	227	870	100
Sulfide, S ²⁻	920	7	1,100
Other (mg/L)	33	13	14
Total dissolved solids (mg/L)	10,640	9,400	16,900

were then placed in the flexible wall permeameter. Test parameters including confining pressure, back pressure saturation, and hydraulic gradient were followed per ASTM D 5887. The experimental design randomized block model (Neter *et al.*, 1990) was implemented to minimize the number of experiments. The model permits the selection of specific combinations of parameters in multifactor investigations while avoiding parameter bias due to systematic or subjective selection. Consequently, not all combinations of clays and leachates were tested in the present study.

In order to simulate the worst possible field conditions, the specimens were back-pressure saturated directly with the leachate and allowed to soak for 48 h before the initial hydraulic conductivity reading was recorded. However, five additional specimens subjected to leachate L-3 were pre-hydrated to investigate the influence of the first wetting liquid on the results. Swell Index tests per ASTM D 5890 were also performed on all clays with all leachates.

EXPERIMENTAL RESULTS AND DISCUSSION

Hydraulic conductivity

In order to measure the variation of hydraulic conductivity with quantity of flow, a constant gradient of ~ 150 was maintained, and measurements were recorded at regular intervals. Both pre-hydrated and

non pre-hydrated specimens exhibited an increase in hydraulic conductivity with time. Each test was continued until no trend was observed in the readings, and a steady value was obtained for the hydraulic conductivity, following the criteria proposed by Ruhl and Daniel (1997), Shackelford *et al.* (2000) and Jo *et al.* (2001). Because back-pressure saturation was employed, and due to limitations in the equipment, it has not been possible to sample the influent and effluent for chemical analysis. Therefore, chemical equilibrium was not established during testing, and the tests are not necessarily representative of long-term conditions.

The hydraulic conductivity values given in Table 3 indicate that leachate L-3 was very detrimental, even to polymer-treated clays. Some of these measurements were confirmed through independent testing by a commercial lab in Orlando, Florida, and by one of the GCL manufacturers. Initial saturation with the leachate resulted in high hydraulic conductivities, with the values being stable from the beginning of the test. This implies that the cation exchange process occurs almost instantaneously upon initial saturation. On the other hand, pre-hydrated specimens exhibited a gradual increase in hydraulic conductivity, but a steady-state condition was always reached beyond a maximum quantity of flow of three pore volumes (Figure 1). Shackelford *et al.* (2000) reported a gradual increase in hydraulic conductivity up to at least 40 pore volumes of flow when a

Table 3. Measured hydraulic conductivities (cm/s).

Clay	Water	Leachate			
		L-1	L-2	L-3	L-3 pre-hydrated
AU	$<9 \times 10^{-9}$			1.3×10^{-5}	4.5×10^{-8}
AT	$<3 \times 10^{-9}$			1.2×10^{-6}	1.0×10^{-8}
BU	$<1 \times 10^{-8}$		1.5×10^{-8}		
BT	$<1 \times 10^{-8}$	1.2×10^{-8}			
CU	$<3 \times 10^{-9}$	4.0×10^{-9}		8.5×10^{-6}	3.1×10^{-8}
CT1	$<1 \times 10^{-7}$			1.4×10^{-6}	2.0×10^{-5}
CT2	$<5 \times 10^{-8}$		1.6×10^{-8}		7.5×10^{-7}

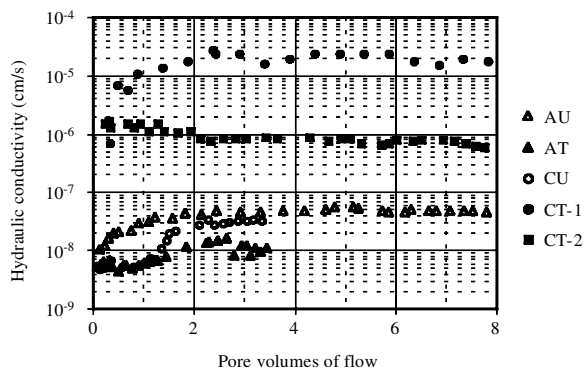


Figure 1. Variation in hydraulic conductivity with pore volumes of flow for pre-hydrated specimens exposed to leachate L-3.

dilute CaCl₂ solution was used. Ruhl and Daniel (1997) found that dilute leachates from MSW facilities did not cause significant degradation in the hydraulic properties of untreated GCLs, even up to nine pore volumes of permeation. In contrast, the results presented here, as well as those published by Jo *et al.* (2001) suggest that significant degradation and steady values can be obtained at smaller pore volumes. This discrepancy can be attributed to the low concentration of the solutions used by the former researchers.

According to the current state of practice worldwide, hydraulic conductivities of ~10⁻⁸ cm/s are typically specified as the minimum acceptable for GCLs in landfill liner applications. A more specific value for the acceptable hydraulic conductivity can be calculated by demonstrating hydraulic compatibility of the GCL with a 300 mm thick clay liner. The results in Table 3 indicate that all materials exhibited hydraulic conductivities greater than those published by the manufacturers, even though the difference is small in most cases. Both untreated and treated bentonites were marginally acceptable under L-1 and L-2. In contrast, L-3 caused significant degradation in the majority of the cases. Specifically, all specimens directly saturated with leachate L-3 were well above the acceptable hydraulic conductivity. Even though this is clearly attributed to the

unusually aggressive nature of the leachate, it is interesting to note that even polymer-treated clays did not perform as expected. Chemical tests on leachate samples obtained several months apart from the L-3 landfill indicate that the Ca and Mg concentrations were consistently high as a result of the ash monofill practice at the waste disposal facility and therefore pose a possible threat to the existing landfill liner.

Effect of pre-hydration

The results presented in Table 3 indicate a significant difference in hydraulic conductivity, depending on the initial wetting conditions. Pre-hydrated specimens performed, in general, far better than those exposed directly to the leachate, even at large numbers of pore volumes of flow (Figure 2). Petrov *et al.* (1997) and Shackelford *et al.* (2000) reported similar findings. The interpretation of such behavior can be drawn from information available in various literature sources on the particle arrangement of clay particles (van Olphen, 1977; Mitchell, 1993; Theng, 1979; Pusch, 1998). In general, initial saturation of Na-montmorillonite with a multivalent-rich solution, such as calcium chloride, causes the divalent cations (Ca²⁺) to immediately occupy most of the cationic sites originally taken by the Na⁺. This abrupt transformation limits water migration into the interlayer space because the electrostatic forces between the cation and the clay particle surface are larger than hydration forces of the divalent cation. Consequently, an aggregated structure with macro-voids is achieved (Figure 3a). In contrast, initial hydration of the monovalent cations (Na⁺) present in Na-bentonite attracts large quantities of water into the interlayer space, thereby creating a dispersed structure, as shown in Figure 3b. The interlayer spacing in this case can be as large as tens of nanometers, which causes extensive swelling of the clay. Upon permeation with a leachate containing multivalent cations, the spacing between the platelets should gradually decrease due to changes in the double layer thickness, but the dispersed and uniform arrangement of the clay fabric is retained (Figure 3c). The homogeneous arrangement of the clay platelets in this case results in a lower hydraulic conductivity than when the particles are aggregated. Shackelford *et al.* (2000), however, appropriately questioned the effectiveness of pre-hydration on the long-term hydraulic

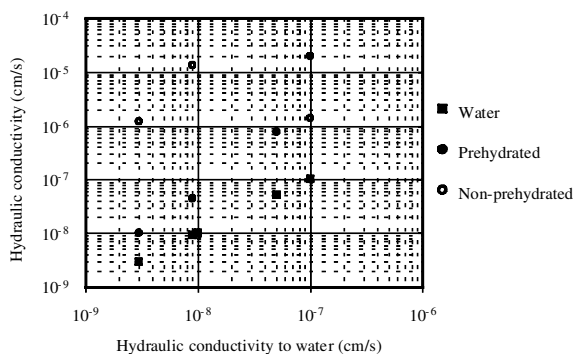


Figure 2. Effect of prehydration on the hydraulic conductivity of specimens exposed to leachate L-3.

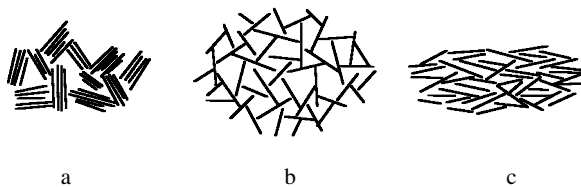


Figure 3. Clay fabric under different saturation conditions: (a) initial saturation with multivalent cations; (b) initial saturation with water; and (c) pre-hydration followed by permeation with multivalent cations.

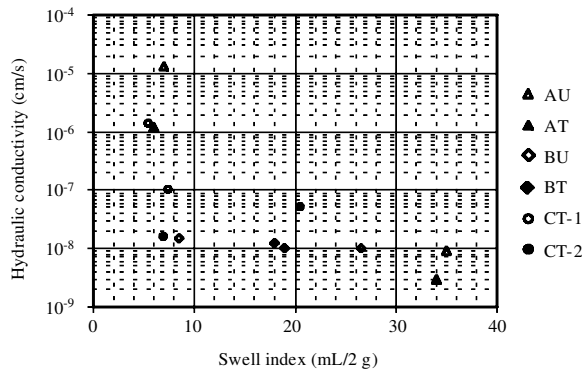


Figure 4. Relationship between swell index and hydraulic conductivity for non pre-hydrated specimens.

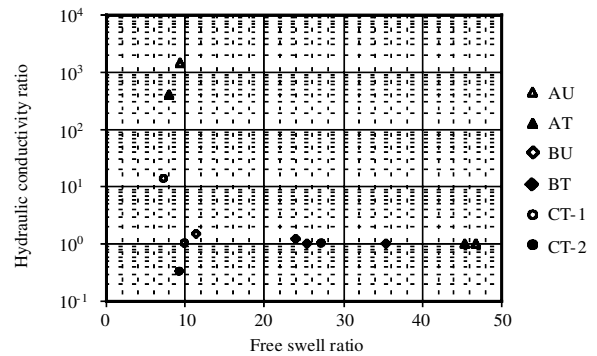


Figure 5. Relationship between free swell ratio and hydraulic conductivity ratio for non pre-hydrated specimens.

conductivity, especially that the tests available in the literature do not go beyond a limited number of pore volumes. Such long-term tests as well as procedures to investigate the mechanism of improvement due to pre-hydration are currently being researched by the authors.

A large variability in results was observed between samples obtained from the same source. Samples CT-1 and CT-2 were both intended to be marketed as the same brand polymer-treated GCL product. However, upon visual and manual inspection of the samples it was evident that they were different, namely in terms of color and aggregation. Large bentonite agglomerations, in general, flocculate upon saturation, especially when cations are present in the solution (Shackelford *et al.*, 2000). In turn, much higher hydraulic conductivities are obtained. Sample CT-1 indeed contained a high percentage of bentonite agglomerations, and exhibited unusually high conductivity with both water and pre-hydrated L-3 leachate.

Relationship to swell index and mineralogy

While performing the permeability experiments, greater swelling was observed in conjunction with specimens with lower hydraulic conductivity, and the opposite was true for highly permeable specimens. The relationship between swell behavior and hydraulic conductivity has been studied by Jo *et al.* (2001). A strong correlation was found to exist in their study and was attributed to the fact that similar mechanisms control both the swelling behavior and the hydraulic

conductivity. A similar trend was observed in the present study, as shown in Figure 4. Although normalization of the swell index and the hydraulic conductivity per Jo *et al.* (2001) did not produce a significant correlation, the results presented in Figure 5 confirm their conclusion that the hydraulic conductivity ratio is constant for free swell ratios >20. Here, the hydraulic conductivity ratio is defined as the ratio between $k_{leachate}$ and k_{water} . The free swell ratio is defined as the free swell index divided by the volume of solids contained in 2 g of clay.

The level of scatter in the data was very high for low-swell combinations, mainly because the hydraulic conductivity is sensitive to changes in soil fabric. Polymer-treated clays consistently produced slightly lower swell indexes compared to their untreated counterparts. Overall, a swell index of 25 or higher always resulted in hydraulic conductivities within the acceptable range. However, the free swell of the CU clay (not shown in Figures 4 and 5), was uncharacteristically high compared to the other materials; values as high as 50 and 90 mL/2 g of clay were obtained with leachate L-3 and water, respectively. It is therefore recommended that the swell index test be used with caution when correlating with the hydraulic conductivity.

In order to evaluate the influence of the mineral composition on the hydraulic and swelling characteristics, a series of X-ray diffraction tests was conducted. Aggregated clays were ground ~5 μm and dried at a low temperature of 50°C to avoid shifting in the diffraction peaks. Unoriented powder samples were prepared

Table 4. Mineralogical composition of clays (from XRD analyses).

Mineral	Wt. %						
	AU	AT	BU	BT	CU	CT-1	CT-2
Montmorillonite	92	91	73	78	56	49	74
Illite			15	10	22	39	12
Quartz and cristobalite	3	4	4	7	12	8	6
Feldspar	4	5	6	6	6	3	8
Calcite			<2	<2	<2	<2	<2
Gypsum					<2		
Siderite, mica and others	<2	<2					

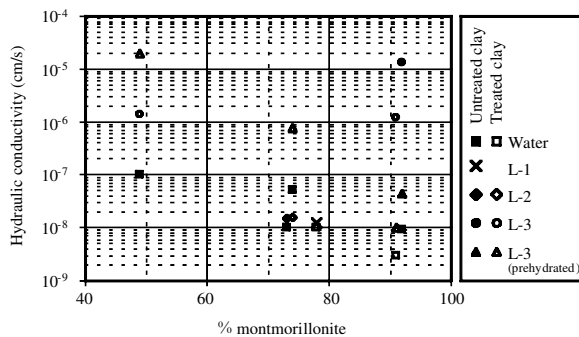


Figure 6. Relationship between montmorillonite content and hydraulic conductivity of GCL clays.

following the Poppe *et al.* (2001) back-loading procedure, and tested in a Philips PW3040 Theta-2 Theta diffractometer. Incident and diffracted beam optics settings were selected based on values recommended by Moore and Reynolds (1997). For quantitative interpretation of the diffraction patterns, the reference intensity ratio method was employed, with mineral intensity factors reported by Burnett (1995), Moore and Reynolds (1997), and Hillier (2000). The results of the analyses are summarized in Table 4.

Because of the abundance of montmorillonite in bentonites, as well as its large specific surface and surface charge, it is often identified as the main mineral affecting the hydraulic and swelling properties of clays. In addition, because of severe changes in the montmorillonite double-layer chemistry upon exposure to different leachates, significant changes can occur in the hydraulic conductivity. Figure 6 shows the relationship between montmorillonite content and hydraulic conductivity for all samples tested in this study, with the exception of CU which, as mentioned earlier, exhibited uncharacteristically high free swell and low hydraulic conductivity. The results plotted in Figure 6 indicate that the hydraulic conductivity decreases as montmorillonite content increases for (1) samples permeated with water and (2) prehydrated samples permeated with L-3 leachate. The data available for L-1 and L-2 leachates are insufficient for establishing a pattern, but the values are comparable to those of water. On the other hand, the hydraulic conductivity appears to be insensitive to montmorillonite content when the samples are initially saturated with leachate L-3. This is in agreement with the mechanism described in Figure 3, as well as with data published by Petrov *et al.* (1997) and Shackelford *et al.* (2000). The results also indicate that polymer treatment is not significantly beneficial when the clay is permeated with water or low-concentration solutions (e.g. L-2). When high-concentration solutions are used (e.g. L-3), and at a given montmorillonite content, polymer-treated GCL clays exhibit, at best, hydraulic conductivities one order of magnitude smaller than the corresponding untreated clay. Although such a level of improvement may be adequate in certain situations, the

resulting hydraulic conductivities are generally higher than typically specified for landfill GCLs. Therefore, acceptance of polymer-treated clays as hydraulic barriers in aggressive inorganic environments should not be recommended without proper verification of their chemical compatibility.

CONCLUSIONS

Hydraulic conductivity tests conducted to evaluate the response of untreated and polymer-treated bentonites to water and ash fill leachates indicated that pre-hydration is generally more beneficial than the use of treated bentonites, for up to at least eight pore volumes of flow. Even though all leachates contained relatively high concentrations of divalent cations, both untreated and treated bentonites produced acceptable results, except when extremely high-concentration solutions were used. Swell index tests confirmed earlier findings that a relationship exists between the swelling behavior and the hydraulic conductivity. Compared to untreated GCL clays with similar mineralogical composition, polymer-treated clays exhibit only limited improvement in terms of reduction in hydraulic conductivity.

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