Evaluation of the long term effect of inorganic leachate on geosynthetic clay liners

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Evaluation of the Long Term Effect of Inorganic Leachate on
Geosynthetic Clay Liners

by

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A dissertation submitted in partial fulfillment
of the requirements for the degree of
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Evaluation of the Long Term Effect of Inorganic Leachate on Geosynthetic Clay Liners

Darwish El-Hajji

ABSTRACT

Because of its low permeability and high swelling characteristics, bentonite is used in various hydraulic barrier systems and in the manufacturing of Geosynthetic Clay Liners (GCLs). Exposure to inorganic solutions containing elevated concentrations of electrolyte can significantly increase their permeability. To enhance the bentonite’s chemical resistance to inorganic solutions, the manufacturers of GCL materials introduced propriety soluble polymeric compounds as an additive to bentonite. The resulting materials are referred to as polymer-treated, chemically-enhanced, or contaminant-resistant clays, and are arguably resistant to a host of inorganic chemicals. In this study, the response of both regular and polymer-treated bentonite clays to ordinary tap water and inorganic landfill leachate is evaluated using permeability tests, index tests and x-ray diffraction (XRD). The results indicate the high dependence of performance on sample preparation techniques, prehydration conditions, and first wetting liquid and, to a lesser extent, polymer treatment. The XRD results indicate that the samples reached chemical equilibrium during the permeation process, as demonstrated by a full shift in $d$-spacing from Na-bentonite to Ca-bentonite. Further, the results show that the cation exchange capacity, the clay plasticity ratio, and the swell index appear to be reliable
indicators of the hydraulic compatibility of bentonite permeated with inorganic chemicals.
Chapter 1 - Introduction

1.1 Background

The issue of environmental protection has been in the forefront for decades. Explosive population growth in states like Florida is placing extreme demands on the groundwater supply. Furthermore, population growth leads to a substantial increase in the solid waste generation and the need to dispose of this waste in an environmentally responsible way. Past waste disposal activities have evolved from simply placing solid waste in pits dug in remote areas to the current state-of-the-art landfills. To protect groundwater supplies from potential contamination by landfill leachate, the environmental permitting agencies mandate landfills to have a bottom containment layer that includes a synthetic liner overlaying a 2-ft thick low permeability layer of compacted clay, with a hydraulic conductivity smaller than $10^{-7}$ cm/s. Low permeability clays are not readily available in many parts of this country and, in many cases, this clay has to be imported at a substantial cost.

To meet the environmental agencies’ regulations, and to provide a cost effective alternate to the Compacted Clay Liner (CCL), the manufacturers introduced a Geosynthetic Clay Liner (GCL) that can be used as an alternate to the CCL. The GCL is manufactured by placing a thin layer of bentonite, mined from the State of Wyoming, between two geotextile filter fabrics. Because of their availability, low permeability, ease
of use and relatively inexpensive construction costs compared to conventional CCLs, the use of GCLs has gained wide acceptance and is becoming the norm in landfill construction (Ashmawy et al. 2002). At the same time, a number of metropolitan areas incinerate their solid waste and dispose of the generated ash in the landfills. During the incineration process, slurry lime is introduced to reduce pollutant emission to the atmosphere. The use of lime causes the ash generated from the incineration process to have elevated concentration of electrolytes typically in the form of calcium salts which end up in the leachate generated by the landfill.

It has been demonstrated that the bentonite’s permeability, upon exposure to calcium salts, tends to increase by orders of magnitude (e.g., Ruhl 1994, Lin and Benson 2000, Shan and Daniel 1991). The manufacturers introduced polymer modified GCL that can maintain its low permeability \((k<10^{-7} \text{ cm/s})\) upon exposure to inorganic solutions containing elevated electrolyte concentrations. The performance of GCL hydrated with non-standard liquids (other than water) has been studied by others including Jo et al. 2001, and Ruhl (1994). However, these studies used simulated liquids rather that actual landfill leachate and, according to the authors, chemical equilibrium was not reached during the permeation process. Without achieving chemical equilibrium, the long-term performance of GCLs when permeated with highly concentrated inorganic solutions remains undocumented. According to Ashmawy et al. 2002 the long-term performance of such materials in aggressive environments has not been fully evaluated.
1.2 Research Scope

The scope of the research program is to evaluate the suitability of the use of GCLs in lining applications, containing aggressive inorganic chemical compounds (high salt concentration), with the following specific objectives in mind:

1. Simulate long-term permeability performance of polymer-treated and untreated soil component of GCLs upon exposure to actual inorganic landfill leachate containing elevated electrolyte concentrations.
2. Assess the effect of the inorganic leachate on the soil’s mineralogical makeup and validate the results.
3. Analyze data and evaluate whether a correlation can be established between index tests as early indicators to the soil’s compatibility with the actual permeating solution.

1.3 Research Objective

The research’s main objective is to establish a procedure that can be adopted to predict the expected permeability performance of the materials upon hydration with inorganic leachate and prior to performing extended duration compatibility tests. Specifically, the hypothesis that there exists a relationship between index test parameters and hydraulic conductivity will be explored. While earlier studies have attempted to establish such a relationship (e.g., Jo et al. 2001; Lee et al. 2005), these studies failed to identify any significant mathematical relationship between index parameters and hydraulic conductivity. In contrast, the principal hypothesis of the current research is the existence of a mathematical relationship between the relative values (ratios) of hydraulic
conductivity and index parameters upon permeation of the bentonite with water and leachate, respectively. As such, the results will be an indicator of the chemical compatibility of the clay with the leachate at hand.

1.4 Materials

The leachate used in this study was obtained from three facilities in Florida: Pasco County, Bay County, and Palm Beach County ash disposal landfills. The soil component of both treated and untreated GCLs was obtained from Colloid Environmental Technologies Company (CETCO) headquartered in Arlington Heights, Illinois and Gundle/SLT Environmental, Inc. (GSE) headquartered in Houston, Texas. These two companies are the primary manufacturers and suppliers of a multitude of geosynthetic lining products in the United States.

The chemical composition of the various leachates was determined through chemical analysis at the University of South Florida (USF) College of Engineering Environmental Laboratory. The composition of the Pasco County leachate (Table 1-1) indicates the presence of calcium cations in high concentration, mainly in the form of calcium chloride. The chemical compositions of the other two leachates (Bay and Palm Beach Counties) exhibited smaller concentrations of these chemicals and are presented later in the dissertation.
This elevated electrolyte concentrations is not typical in ash landfills and is specific to Pasco County. For example, leachate generated from Bay County and West Palm Beach ash landfills has electrolyte concentrations in the range of 4,500 to 6,000 mg/L in the form of calcium chloride. The elevated electrolyte concentration in the Pasco County leachate is due to the end product generated by their leachate treatment facility. This facility implements the use of evaporation technology to remove the dissolved salt from the leachate. The end byproduct of the treatment is water and pure salt crystals. The water byproduct is further treated at the adjacent wastewater treatment plant and the salt crystals are taken in industrial-sized canvas bags and deposited in the active landfill ash cell. Subsequent rain causes the dissolution of the salt crystals back into the leachate collection system. This leaching process, coupled with more salt being leached out of the deposited ash, causes the salt concentrations to be abnormally high. Therefore, the use of this leachate as the permeant for this research clearly represents a conservative condition that will simulate a worst condition scenario.

The GCL manufacturers use propriety polymer treatment processes to enhance the physical properties (mainly permeability) of sodium-bentonite (Na-bentonite) soils mined from the State of Wyoming that are intended for use in a variety of environmental applications. In the geosynthetics industry, the term Contaminant Resistant Clay (CRC)
has become synonymous with these polymer-treated products. A total of ten soil samples were obtained from these manufacturers in 1999. CETCO provided a total of six samples that consisted of five polymer-treated samples and one untreated sample. GSE delivered a total of four samples of which two were polymer-treated and the other two were of the regular (untreated) type. These samples were sent to the USF’s Geoenvironmental Laboratory in multiple shipments. All of the samples that arrived as part of the first shipment were labeled as “Old” and the subsequent shipment were assigned the term “New”, preceded by the manufacturer’s name in order to differentiate between the samples. Concurrently, the polymer-treated materials were assigned the symbol (T) whereas the untreated materials were labeled (U). For example, the first polymer-treated shipment received from GSE was labeled as “GSE Old T” and the subsequent shipment was labeled as “GSE New T”. CETCO provided four polymer-treated samples in the first shipment followed by one treated and one untreated sample. The first shipment was labeled as “CETCO Old T #1, CETCO Old T # 2 and so on, while the subsequent polymer-treatment sample was labeled as “CETCO New T”. The untreated sample was labeled as “CETCO New U”.

Visual inspection of the soils samples revealed varying textures, colors and appearances that ranged from fine to coarse aggregate with colors varying from light gray to charcoal.

1.5 Testing Procedures

The soils samples were tested to measure their permeabilities (hydraulic conductivities), index properties and mineralogical makeup. The results were further
analyzed to determine whether meaningful relationships can be established between the index properties and the materials’ permeability. The index testing included the swell index tests, Atterberg limits (plasticity) tests, and methylene blue adsorption tests.

1.5.1 Permeability Measurements

The permeability measurements were performed in general accordance with the ASTM D5887 method using a flexible wall permeameter. In this dissertation, as in geotechnical practice, the terms permeability, coefficient of permeability, and hydraulic conductivity will be used interchangeably. One of the termination criteria in the ASTM Standards stipulates the termination of the test when two pore volumes of liquid have passed through the sample. This termination criterion was followed for water permeation condition. When leachate was used, the permeation process continued for extended periods that lasted, in some instances, three months with over 100 pore volumes of leachate passing through the samples. The reader is referred to Chapters Two and Four for detailed discussions and results interpretation.

1.5.2 Swell Index Test

The swell index tests measure the swelling properties of a clay mineral in a reagent (water or leachate). The testing procedure is simple and entails the placing of 100 ml of permeant in a graduated cylinder. To that, a total of 2 g of the soil is added gradually and allowed to swell freely for a period of 48 hours. The final soil volume is then recorded and graphically plotted. Bentonite is believed to achieve its low permeability by virtue of its swelling behavior. This swelling constricts the pore spaces available for the permeating liquid to travel through, which in turn reduces the
permeability of the material. The swell index test can provide an insight to the expected permeability performance of the material. However, there have not been any mathematical models that can be used to predict the permeability based on the material’s swelling index.

1.5.3 Liquid Limit

In the United States, the ATSM 423 method is typically used to calculate the index properties, collectively called Atterberg limits, of fine-grained (clay) soils. Because of the high plasticity of the bentonite materials it was not possible to form a standard groove as called for in the testing procedure, and subsequently, this testing method was abandoned. Instead, the British Standard Method BS 1377:1975 was used to calculate the liquid limit. The British Standard specifies the use of a cone penetrometer apparatus, shown in Figure 1-1 below, and requires the mixing of 200 to 250 g of soil with distilled water and then allowing the mixture to cure in a covered container for a period of 24 hours.

![Figure 1-1 Cone penetrometer](image-url)
Following the curing period, the prepared sample is re-mixed for a minimum of ten minutes and placed in a special metal cup. The cup is placed under the cone penetrometer and the initial displacement gauge reading is recorded. The cone is released and allowed to penetrate into the soil sample and held into that position for 20 seconds. The final gauge reading is recorded. This step is repeated by adding more water to the soil to obtain a range of cone penetration from about 15 to 25 mm. The liquid limit versus the displacement values is plotted and the liquid limit value at 2 cm displacement is recorded as the liquid limit (LL).

1.5.4 Plastic Limit

The plastic limit was measured in accordance with the ASTM D4318 method which requires the mixing of a small amount of water with the soil material and the rolling of the mixed material into a thread-like shape having a diameter of about 3 mm. The procedure is repeated by adding additional water until the mixed soil material begins to crumble at the 3 mm diameter. The rolled material is weighted and placed in a drying oven to determine the water content of the soil, which represents the plastic limit (PL). The plasticity index (PI) is calculated as the difference between the liquid limit and plastic limit.

1.5.5 Methylene Blue Adsorption

The methylene blue adsorption test (MBA) is a semi-quantitative analysis used to determine the presence of clay minerals in a soil sample and to calculate the soil’s cationic exchange capacity (CEC). The underlying principle is that if the amount of methylene blue adsorbed by the clay is an indication of its CEC and, consequently, of its
swelling potential. According to Taylor 1984, the cations in the MB solution irreversibly replace those on the exterior clay surface as follows:

\[
\text{Ca-Na-Clay + MB hydrochloride} \rightarrow \text{MB-Clay + Ca-Na-Chloride}
\]

The MBA is a simple and relatively quick lab procedure and can be performed using either the “turbidimetric” method or the "spot" method. The turbidimetric method is more accurate, but more complex, than the spot method, and is performed by the mixing of finely ground soil with methylene blue solution and then leaving the mixture for a few days. A spectrometer is then used to determine the amount of methylene blue adsorbed by the clay. The spot method, used for this study, is a titration procedure in which a methylene blue solution is added in measured quantity to a mass of finely ground soil particles, thoroughly mixed, and a single drop is extracted from the suspension using a pipette and dropped on a filter paper. The concentration of the methylene blue in the solution is increased until a blue ring begins to appear around the “spot” on the filter paper. The appearance of such a ring or halo signifies that the clay has reached its maximum adsorption capacity due to cation exchange at the surface. The total amount of methylene blue solution adsorbed is used to calculate the CEC using the following mathematical equation:

\[
\text{C.E.C.} = \frac{\text{MB added (cc)} \times \frac{\text{MB dry wt (g)}}{319.87} \times \frac{1000}{\text{Vol. of MB solution (cc)}} \times \frac{100g}{\text{Clay dry wt (g)}}}{100}\text{ (mEq/100 g)}
\]

The methylene blue used in the current study is available commercially and was acquired from Fisher Scientific.
1.5.6 X-Ray Diffraction

The XRD technique was used to evaluate the soil’s mineralogical makeup in the as-received, water-permeated and leachate-permeated samples in order to determine whether chemical equilibrium was achieved during the leachate permeation process. The XRD analysis can be performed using qualitative, quantitative or semi-quantitative analysis or a combination thereof. The qualitative analysis reveals the mineralogical make up of the minerals present in a sample while the quantitative analysis is used to measure the abundance of those minerals. For this research, the qualitative analysis was used because the intent of the research is to determine whether or not any alteration has occurred in the mineralogical makeup upon leachate permeation. The reader is referred to Chapters Two and Five of this dissertation for an expanded discussion of the XRD technique and conclusions.

1.6 Testing Facilities

The various testing procedures were conducted at the appropriate laboratories situated in the USF. The permeability and index tests were conducted at the College of Engineering Geoenvironmental Laboratory. The permeability test apparatus consists of a state-of-the-art permeability panel with the capability of performing multiple permeability tests concurrently while maintaining the regulated pressure for each permeability cell. At the onset of the testing program, the panel’s burettes were used as influent and effluent chambers. This practice was quickly terminated because the metal components of the panel began to corrode from the highly corrosive leachate.
Subsequently, external clear plastic cells were fabricated at the College of Engineering machine shop and the metal components were coated with epoxy-based paint. These tanks provided large storage capacity for the leachate and enabled the user to suspend the permeability test to obtain samples or add additional leachate without disturbing the sample.

The XRD analysis was conducted at the USF Material Testing Laboratory equipped with a state-of-the-art Philips PW 3040/60 XRD instrument that was directly connected to a PC for data acquisition and control. The lab was equipped with the necessary sample preparation instruments which includes a pestle and mortar, and sieves with varying mesh sizes. Furthermore, XRD results were analyzed using a software system, X’Pert Pro, developed by Philips.

The leachate chemical analysis was performed by a qualified technician at the USF College of Engineering Environmental Laboratory. The lab is fully equipped and has the capabilities of performing various chemical analyses on a host of chemical compounds.

1.7 Dissertation Organization

The organization of this dissertation is mainly based on professional journal papers co-authored by the dissertation author that were either published, submitted for publication consideration, or being submitted for publication. Each of these papers was authored and formatted with the intent of being a standalone document suitable for publication. These papers are incorporated in this dissertation in Chapters Three, Four and Five and as such some of the information included in these papers may be partly
duplicated in other parts of the dissertation. However, the paper sequence provides an integrated view of the topic at hand and serves to support the hypothesis of this dissertation.

Chapter Two provides a review of past literature and discusses related topics that include bentonite origin and characteristics, diffuse double layer theory, cationic exchange capacity, XRD, factors affecting permeability measurement, and related data published by other researchers. This chapter provided a road map to the development and evolution of this research project. Information collected from the various literature sources illustrate the gaps that exist in the published data and lead to the specific goal and objectives of this research program.

Chapter Three provides a discussion of materials index testing that include swell index, liquid limit and plasticity index test, plastic limit and USCS classification. This chapter was published as a journal paper (Ashmawy et al. 2002) and lays out the main shortcomings in the current state-of-the-art by validating some of the empirical testing relations that were proposed or established by others. The findings illustrate the inability of the current models to capture the dependence of bentonite permeability on the various index properties.

Chapter Four includes some of the more recent data that was recently published in ASCE Geotechnical Special Publication No. 142 (Ashmawy et al. 2005). It discusses advection, diffusion, and sorption characteristics of inorganic chemicals in GCL bentonite. This chapter utilizes the data published by Ashmawy et al. 2005 to further examine the bentonite material characteristics and its interaction with inorganic leachate. This chapter forms the core of the dissertation because the data was analyzed with the
purpose of developing mathematical models that can be used to predict the bentonite’s permeability performance and its resistance to inorganic chemicals vis-à-vis what is called “performance compatibility” in engineering practice. The results and the new mathematical models are being prepared for publication in a journal paper.

Chapter Five provides an in-depth discussion of the XRD technique and how it was used to evaluate the changes in lattice parameters or d-spacing of the crystalline component of the bentonite. A detailed discussion of the XRD data collected during the research process is presented. The material contained in this chapter is essential in verifying that chemical equilibrium, that is, full replacement of sodium cations by calcium cations, has been achieved during the permeation process, thus validating the findings in the earlier chapters.

Chapter Six is intended to provide conclusion and present a concise summary of the testing program, and to present the key findings. Further this chapter includes a section on engineering implications intended to provide the engineering community with a useful procedure based on statistical models obtained through nonlinear regression analysis. This statistical regression model can be used by design professionals to make informed decisions, early in the design process, regarding GCL material selection and its suitability and chemical compatibility for the intended use.
Chapter 2 - Literature Review

2.1 Overview

This chapter presents a comprehensive review of the literature published on the permeability performance of geosynthetic clay liners (GCL), XRD technique and material index testing. Throughout this review, the words bentonite, montmorillonite and clay may be used interchangeably despite the subtle technical differences between these terms.

Bentonite is characterized as a low permeability soil having high swelling and “self-healing” potentials. Clays are typically used in environmental containment applications that include landfill bottom liners, lagoon lining and tank farms. Bentonite has a fine powder texture and is usually mixed with local soils using pug mills to reduce their permeabilities. GCLs are manufactured by placing approximately a half centimeter-thick layer of pure sodium or calcium bentonite between two geotextile fabrics. The use of GCLs has evolved from simple water irrigation pond containments to environmental containment applications to separate certain organic and inorganic compounds from the surrounding environment. It has been illustrated that certain organics and inorganic contaminants can substantially increase the permeability values of bentonite material (e.g., Ruhl 1994, Lin and Benson 2000, Shan and Daniel 1991). To counter the negative effects of these contaminants on the permeability of the bentonite material, the
manufacturers introduced a propriety treatment process to enable the bentonite to maintain its low permeability upon exposure to inorganic compounds. Conceptually, the treatment process involves the use of polymer materials to encapsulate the bentonite with a sacrificial layer, thus promoting the ionic exchange within that layer instead of the cations present in the bentonite (McKelvey 1996).

2.2 Bentonite Origin and Characteristics

The term “clay” is used, in the technical sense, to describe fine-grained soils having particle diameter size less than 2 microns (0.002 mm) and characterized by their high plasticity index and swell potential. Bentonite is classified as clayey soil that was formed by ash spewed by volcanic eruptions and deposited into the seas that covered much of Wyoming during the cretaceous age. Wyoming bentonite is composed mainly of the mineral sodium (Na\(^+\)) montmorillonite, which has an intrinsically high swelling capacity and low permeability values. Alternatively, calcium (Ca\(^{2+}\)) montmorillonite, readily available in Europe and parts of the United States, exhibits less swelling and has higher permeability values than the Na-montmorillonite type.

Clay minerals are classified into the following four groups 1) kaolinite; 2) smectite; 3) illite and 4) chlorite. Montmorillonite falls within the smectite group and is characterized as a 2:1 clay mineral composed of one gibbsite sheet held between two tetrahedral sheets as illustrated in Figure 2-1 below.
The soil interlayers for the smectite group are weakly held together and can have a basal spacing \((d\)-spacing\) that varies between 9.6 Angstrom in the dry condition to a complete separation when the soil is treated with Ethylene Glycol (Moore and Reynolds 1989). Because the interlayer is expansible, smectites exhibit high swelling potential. The separation between individual smectite sheets varies and depends on:

1. The interlayer cations type; for example monovalent cations like sodium \((\text{Na}^+)\) cause more expansion than do divalent cations like calcium \((\text{Ca}^{2+})\),
2. The concentration of ions in the surrounding solution, and
3. The amount of water present in the soil.

### 2.3 Water Adsorption and Swelling Capacity

The liquid limit of bentonite is appreciably high because of its high water adsorption capacity which is attributed to its small particles that have a high amount of absorbing surface. According to Egloffstein 1995, it could take up to ninety minutes for sodium bentonite to reach its maximum adsorption capacity while calcium bentonite could reach it in about twenty minutes as illustrated in Figure 2-2 below.

![Figure 2-1 Two-to-one (2:1) clay mineral](image)
Figure 2-2 Water adsorption of Ca-bentonite and Na-bentonite (adopted from Egloffstein 1995)

Sodium bentonite’s high water absorption capacity contributes to its high plasticity, fracture resistance, high swelling and low permeability. The low permeability of bentonite is attributed to its mineralogical make up and swelling capacity. For example, sodium montmorillonite can free swell fifteen to twenty times its original volume while calcium montmorillonite swells to about five times of its original volume (Egloffstein 1995). Upon swelling, the permeability of the soil tends to decrease because the pore spacing becomes constricted, thus reducing the available conduits for the water to travel through the soil’s fabric (Ruhl 1994).
2.4 Diffuse Double Layer Theory

According to Das 1990, clay particles carry a net negative charge on their surface mainly due to isomorphous substitution and break in the structural continuity at its edge. In dry clay, the electrical neutrality is preserved by electrostatic attraction between the exchangeable cations such as Mg$^{2+}$, Ca$^{2+}$, Na$^+$ and K$^+$ and the negatively charged particle surface. The diffuse double layer is formed when water is added to the dry clay whereby the cations and some anions encapsulate the clay particle. Cation concentration decreases with increased distance from the clay particle while anions concentration increases with increased distance. At a known distance from the clay particle, the cations and anions reach equilibrium as illustrated in Figure 2-3 below.

![Figure 2-3 Diffuse double layer (adopted from Das 1990)](image)

The double layer thickness is directly proportional to the soil’s water adsorption capacity and inversely proportional to its permeability. An increase in the double layer thickness decreases the soil’s permeability by constricting the flow path available to the permeating liquid to travel between the soil’s particles as illustrated in Figure 2-4 below.
The high water adsorption capacity of Na-montmorillonite causes an increase in its double layer thickness and a decrease in its permeability. Alternatively, Ca-montmorillonite has a low water adsorption capacity which causes the diffuse double layer thickness to remain relatively unchanged and the free water flow path unobstructed thus resulting in higher permeability than Na-montmorillonite.

### 2.5 Cationic Exchange Capacity

CEC is used to measure the soil’s activity and interactions with the various chemical compounds and is expressed in milliequivalents per 100 g of dry soil (mEq/100g). It can be measured using the MBA test in accordance with the ASTM C0837 and D2330 methods. The CECs for the various clay minerals vary widely as illustrated in Table 2-1 below.

<table>
<thead>
<tr>
<th>Clay Mineral</th>
<th>CEC (meq/100 g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Montmorillonite</td>
<td>80-150</td>
</tr>
<tr>
<td>Illite</td>
<td>10-40</td>
</tr>
<tr>
<td>Kaolinite</td>
<td>3-15</td>
</tr>
</tbody>
</table>
Based on the above CEC values, soils composed of the mineral montmorillonite tend to have lower permeability than those having illite or kaolinite as the main mineral. Within the same mineral group, the CEC can vary widely. For example sodium montmorillonite has a higher CEC value than that of calcium montmorillonite, as noted by Lin and Benson 2000 and Egloffstein 2002.

The soil mineralogical makeup and its CEC directly affect the soil’s permeability. Cationic exchange is one of the mechanisms responsible for altering the soil’s mineralogical makeup. For instance, when sodium montmorillonite is permeated with inorganic liquids having elevated calcium chloride concentration, a cationic exchange process occurs whereby the higher divalent calcium cations replace the weaker monovalent sodium cations. This cationic exchange process continues until chemical equilibrium is reached whereby the sodium cations are completely exhausted and replaced by those of calcium (Figure 2-5) and resulting in soil having low expansion potential and higher permeability value.

![Figure 2-5 Change from sodium to calcium bentonite](image)

According to Brown and Anderson 1983, the cationic exchange mechanism also affects the soil particle orientation. This alteration increases the clay permeability when its particle arrangement is altered from a well-dispersed state into a flocculated one. The low permeability of Na-montmorillonite is attributed, in part, to its very fine soil particles
that are dispersed and randomly oriented. Because of this particle arrangement, it would take a considerable amount of time for the permeating liquid to travel through the soil’s fabric paths. Upon transformation to Ca-montmorillonite, the soil’s particles become flocculated and oriented, thus providing a direct travel path for the permeating liquid to travel through the soil fabric, resulting in higher permeability as illustrated in Figure 2-6 below.

Flocculation of the soil particles causes an increase in the soil’s permeability, and diminishes with increased thickness of the double layer. In turn, the characteristics of the double layer are influenced by: 1) permeating liquid electrolyte concentration \((n_0)\), 2) temperature \((T)\), 3) ion valence \((v)\) and 4) the permeating liquid dielectric constant \((\varepsilon)\) which is a measure of the ease with which molecules can be polarized and oriented in an electric field (Goldman et al.1998). The double layer thickness \((H)\) can be calculated using the following mathematical relation (Mitchell 1993):

\[
H = \sqrt{\frac{\varepsilon k T}{8 n_0 \pi \varepsilon^2 v^2}}
\]  

(2.1)

where, \(k\) is Boltzman’s constant \((1.38 \times 10^{-16} \text{ erg}^{\circ}K)\) and \(e\) is the unit electronic charge \((1.6 \times 10^{-19} \text{ coloumb})\).
2.6 Factors Affecting Permeability Measurement

The soil’s permeability is influenced by inherent properties that include the soil’s mineralogical makeup, water adsorption capacity, diffuse double layer thickness and CEC. These properties are interrelated, interdependent and are specific to the soil’s makeup. They can be altered by external factors that include permeating liquid chemical make up, test confining pressure and hydraulic gradient. In an effort to establish uniformities while performing permeability measurement, the ASTM D5887 and D5084 methods established certain standards that must be followed during laboratory sample preparation and testing. These standards are specific to sample preparation methods, magnitudes of the confining pressure and hydraulic gradient, length of hydration, permeation, test termination criteria and calculation procedures.

2.6.1 Permeant Type and First Wetting Liquid

To perform laboratory permeability measurements, the ASTM D5084 method recommends a 48-hour hydration period using tap water (first wetting liquid) followed by permeation with the intended chemical compound. Water hydration can substantially increase the thickness of the diffuse double layer, albeit by varying degree depending on the soil’s mineralogy. The increase in the double layer thickness could result in artificially low permeability value. Ruhl 1994 reported a permeability of $2 \times 10^{-9}$ cm/s for a GCL material that was first hydrated with water and then permeated with simulated landfill leachate. When leachate was used to hydrate and permeate the GCL, the measured permeability increased to $2 \times 10^{-5}$ cm/s.
Water, because of its chemical neutrality, is not detrimental to the soil’s low permeability while liquids containing certain organic and/or inorganic compounds can adversely affect the soil’s permeability. Furthermore, organic liquids like aromatic hydrocarbons can increase the montmorillonite’s permeability by order of magnitudes. When Brindley and Brown 1980 permeated bentonite with water, they reported a permeability value of $3.6 \times 10^{-8}$ cm/s and when the same bentonite was permeated with xylene, the permeability increased to $1.76 \times 10^{-4}$ cm/s, almost a four-orders-of-magnitude increase.

Inorganic chemical compounds like calcium chloride, magnesium chlorides and sodium chloride can significantly increase the bentonite’s permeability. Lin and Benson 2000 reported a permeability increase from $5 \times 10^{-9}$ cm/s to $8 \times 10^{-6}$ cm/s for bentonite material permeated with water and then with a solution containing 0.0125 mol/L of calcium.

### 2.6.2 Confining Pressure

Using high confining pressure is not recommended during permeability testing because it could lead to soil consolidation thus artificially lowering its permeability. When Daniel et al. 1997 conducted permeability testing on GCL material using a confining pressure between 5 and 10 kPa, they achieved a permeability of $10^{-9}$ cm/s and upon increasing the confining pressure to 300 kPa, the permeability decreased by one order of magnitude. According to Ruhl 1994, when Shan and Daniel 1991 permeated a GCL material with tap water under a confining pressure of 4 kPa they reported a permeability of $2 \times 10^{-9}$ cm/s and when the confining pressure increased to 140 kPa, the
permeability decreased to $3 \times 10^{-10}$ cm/s. The ASTM D5887 method recommends a maximum confining pressure of 35 kPa for permeability testing with no minimum value specified. The confining pressure should be specific to the intended use and should be consistent with the pressure present under normal field conditions.

### 2.6.3 Hydraulic Gradient

It is the norm to use high hydraulic gradient to perform permeability tests for fine grained soils. Due to their low permeabilities it would take considerable amount of time for the permeant to fully saturate and penetrate the soil layer. Conversely, Jo et al. 2001 recommend the hydraulic gradient be as close as what can be expected in the field under actual conditions. Testing with gradients higher than that could result in an artificially low permeability due to sample consolidation. Although the ASTM D5887 method recommends a maximum hydraulic gradient of 30, researchers and practitioners continue to test low permeability soils using elevated gradients. Hydraulic gradients that ranged from 50 to 550 were reportedly used by others for the same material with no noticeable change of the reported permeability values. Jo et al. 2001 reports that Rad et al. 1994 demonstrated that using a hydraulic gradient of 2800 to permeate a GCL sample with water had no measurable effect on the permeability value, and Petrov et al. 1997 illustrated the same findings as shown in Figure 2-7.
2.6.4 Electrolyte Concentration

According to Sivapullaiah and Savitha 1999 the inter-particle separation of the bentonite particles in water can lead to a higher swell index while electrolyte solutions tend to limit or inhibit the inter-particle separation, thus leading to a lower swell index and a potential decrease in the diffuse double layer thickness. Sykes et al. (1982) reported a permeability of $4.14 \times 10^{-8}$ cm/s when the salt concentration in the permeating liquid was at zero percent, but when the salt concentration increased to five percent, the bentonite permeability increased to $1.31 \times 10^{-7}$ cm/s.

2.7 X-Ray Diffraction

The theory and practical applications of XRD techniques are well documented in the literature (e.g., Brindley and Brown 1980; Whittig 1986; Bueno 2002). XRD is used to determine the mineralogical make up of soil samples in qualitative and quantitative manners, and will be used for that purpose in this dissertation. According to
Whittig 1986 x-rays are electromagnetic radiation generated from the oscillation of electrostatic and electromagnetic fields of a wavelength equal to the interatomic distance in a crystal.

XRD is based on Bragg’s law, and depends on the crystal’s physical properties. Crystalline structures are formed by three dimensional arrangements of atoms situated at a fixed interplanar distance, commonly referred to as $d$-spacing or basal spacing, within a crystal. Diffraction occurs when the electromagnetic waves with a wavelength $\lambda$ bombard an atom causing the incident wave to refract. Successive diffraction enforces the diffracted beam resulting in signal strength sufficient for recording as illustrated in Figure 2-8 below.

The XRD pattern is expressed mathematically using Bragg’s law (Equation 2.2) that must be satisfied for the diffraction process to be successful.

$$n\lambda = 2d \sin \theta \quad (2.2)$$

Figure 2-8 XRD pattern (adopted from Wikipedia)
where \( n \) is an integer, \( \lambda \) is the wavelength of the x-rays, \( d \) is the spacing (basal spacing) between the planes in the atomic lattice, and \( \theta \) is the angle between the incident ray and the refracted beam.

According to Moore and Reynolds 1989, the diffraction process and the resulting diffraction data are highly dependable on the sample preparation techniques. Poor sample preparation could result in a destructive diffraction pattern while a good sample preparation technique can result in a constructive diffraction pattern. Constructive diffraction occurs when two electromagnetic waves are in phase whereby their troughs and peaks line up thus magnifying the amplitude of the refracted wave. Alternatively, destructive diffraction occurs when the waves are 180° out of phase and the resulting troughs of one wave line up with the peaks of the next wave resulting in waves having little or no amplitude as illustrated in Figure 2-9 below.

![Figure 2-9 Constructive and destructive waves (adopted from Wikipedia)](image)

The types of refracted waves have an impact on the XRD results whereas narrow peaks with lower amplitude as an indication that destructive interference was introduced during the diffraction process while wider peaks with higher amplitude is an indication that constructive waves were recorded (Moore and Reynolds 1989).
Depending on the scope of the study, the XRD process can be conducted with a qualitative or quantitative analysis. The qualitative analysis is the simpler of two methods and little experience is required to perform the analysis, whereas the quantitative analysis requires a great deal of experience and, if not used properly, can lead the researchers to wrong conclusions (Moore and Reynolds 1989). The quantitative analysis is conducted using either an internal standard method or an external standard method. According to Brindley and Brown 1980 the choice of method depends largely on the type of material being analyzed. The internal standard method is typically used in analyzing powdered soil specimens and is applied to mineral or materials for which the composition is unknown. Alternatively, the external standard method is used on solid materials like alloys and allows the quantification of one or more components in the material while the direct comparison method is only applicable to fully crystalline mixtures and does not require any standards.

There are various potential sources of errors that could occur before, during and after the diffraction process has been completed which could cause the user to misinterpret the data. The most common type of error that occurs before the diffraction process involves sample preparation techniques. The sample must be prepared with great care to ensure that near random orientation of the soil particles is achieved so that each set of crystals can be diffracted. Errors could also result during the diffraction process by the improper mounting of the sample and instrument misalignment. Following the diffraction process, data misinterpretation is the major source of error and usually occurs due to the lack of user’s experience.
Chapter 3 - Index and Permeability Testing

3.1 Overview

In this chapter, 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 calcium and magnesium cations. The results are interpreted using existing models developed by other researchers, and the limitations of such models are discussed. Most notably, the inability of such models to provide a systematic methodology or a general framework for evaluating the compatibility of bentonite with specific inorganic chemicals is demonstrated.

3.2 Introduction

The clay component of GCL materials tested in this study consists of regular and polymer-treated bentonite. As mentioned in Chapters One and Two, polymer treatment arguably renders the clay non-reactive to many organic and inorganic chemicals. In municipal solid waste 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 sodium montmorillonite, although calcium 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 possibly renders the montmorillonite non-reactive towards most organic and inorganic chemical compounds. Laboratory test results (El-Hajji et al. 2001) suggest that polymer-treated GCLs maintain their low hydraulic conductivity when hydrated with liquid containing single-species solutions at low concentration. In this chapter, 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 calcium and magnesium are disposed.

3.3 Bentonite in GCL

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. Sodium and Calcium montmorillonite, which constitute the main mineral in bentonite clays, are present in salt and fresh water 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, 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 several orders of magnitude, 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; El-Hajji et al. 2001).

Over the past several years, GCL manufacturers introduced products such as CRCs or polymer-treated bentonites (PTB) to minimize bentonite degradation upon exposure to contaminants. Polymer treatment processes and formulas 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 1970’s (e.g. Pezerat and Vallet 1973; Bart et al. 1979).

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 sodium or calcium 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 sodium ions. In this case, sodium ions are not replaced, but rather the sodium montmorillonite sheets are “coated” with the polymer.

3.4 Experimental Program

3.4.1 Description of Clays and Leachates

In the present study, seven bentonite clays, produced by two 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 3-1 contains a description of the seven bentonites with the nominal properties provided by the manufacturers. The principal mineral in all seven clays is sodium montmorillonite. The leachates were obtained from incinerator ash disposal facilities in three different counties in the State of Florida: L-1 from Bay County, L-2 from Palm Beach County, and L-3 from Pasco County. The Pasco County facility (L-3) is an ash monofill while the other two facilities use landfill co-disposal where both incinerator ash and MSW are disposed. Table 3-2 summarizes the relevant chemical characteristics of the leachates.
### Table 3-1  Bentonite properties, as supplied by the manufacturers (ASTM D5890\(^a\), ASTM D5887\(^b\), based on typical GCL thickness)

<table>
<thead>
<tr>
<th>Manufacturer</th>
<th>Treatment</th>
<th>Label</th>
<th>Swell Index (^a) ml/2g</th>
<th>Hydraulic conductivity (^b) cm/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>GSE</td>
<td>Untreated</td>
<td>AU</td>
<td>24</td>
<td>5×10(^{-9})</td>
</tr>
<tr>
<td>GSE</td>
<td>Treated</td>
<td>AT</td>
<td>24</td>
<td>N/A</td>
</tr>
<tr>
<td>GSE (Bentofix)</td>
<td>Untreated</td>
<td>BU</td>
<td>24</td>
<td>5×10(^{-9})</td>
</tr>
<tr>
<td>GSE (Bentofix)</td>
<td>Treated</td>
<td>BT</td>
<td>24</td>
<td>1×10(^{-9})</td>
</tr>
<tr>
<td>CETCO</td>
<td>Untreated</td>
<td>CU</td>
<td>24</td>
<td>5×10(^{-9})</td>
</tr>
<tr>
<td>CETCO</td>
<td>Treated</td>
<td>CT(_1)</td>
<td>24</td>
<td>5×10(^{-9})</td>
</tr>
<tr>
<td>CETCO</td>
<td>Treated</td>
<td>CT(_2)</td>
<td>24</td>
<td>5×10(^{-9})</td>
</tr>
</tbody>
</table>

### Table 3-2  Relevant leachate properties, measured at the USF environmental laboratory

<table>
<thead>
<tr>
<th>Leachate</th>
<th>L-1</th>
<th>L-2</th>
<th>L-3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Landfill type</td>
<td>Co-disposal</td>
<td>Co-disposal</td>
<td>Ash monofill</td>
</tr>
<tr>
<td>pH</td>
<td>6.30</td>
<td>6.55</td>
<td>7.24</td>
</tr>
<tr>
<td>Cations (mg/L)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ammonia, (NH_4^+)</td>
<td>60</td>
<td>260</td>
<td>15</td>
</tr>
<tr>
<td>Sodium, (Na^+)</td>
<td>2,200</td>
<td>300</td>
<td>1,640</td>
</tr>
<tr>
<td>Calcium, (Ca^{2+})</td>
<td>1,150</td>
<td>2,625</td>
<td>5,120</td>
</tr>
<tr>
<td>Magnesium, (Mg^{2+})</td>
<td>750</td>
<td>1,525</td>
<td>2,075</td>
</tr>
<tr>
<td>Anions (mg/L)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chloride, (Cl^-)</td>
<td>5,300</td>
<td>3,800</td>
<td>6,836</td>
</tr>
<tr>
<td>Bicarbonate, (HCO_3^-)</td>
<td>227</td>
<td>870</td>
<td>100</td>
</tr>
<tr>
<td>Sulfur Species</td>
<td>920</td>
<td>7</td>
<td>1,100</td>
</tr>
<tr>
<td>Other (mg/L)</td>
<td>33</td>
<td>13</td>
<td>14</td>
</tr>
<tr>
<td>Total Dissolved Solids (mg/L)</td>
<td>10,640</td>
<td>9,400</td>
<td>16,900</td>
</tr>
</tbody>
</table>
All three leachates were found to possess high levels of calcium and magnesium compared to other multivalent cations (e.g., aluminum, copper, and zinc). The abundance of calcium and magnesium, 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).

3.4.2 Specimen Preparation and Test Parameters

Falling head permeability tests with back-pressure saturation were performed following the ASTM D5887 method. The only 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 were then placed in the flexible wall permeameter. Test parameters including confining pressure, back pressure saturation, and hydraulic gradient were followed per the ASTM D5887 method. 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 hours before the initial hydraulic conductivity reading was recorded. However, five additional specimens subjected to leachate L-3 were prehydrated to investigate the influence of the first wetting liquid on the results. Swell Index tests per the ASTM D5890 method were also performed on all clays with all leachates.

3.5 Experimental Results and Discussion

3.5.1 Hydraulic Conductivity

To measure the variation of hydraulic conductivity with quantity of flow, a constant gradient of approximately 150 was maintained, and measurements were recorded at regular intervals. Both prehydrated and non-prehydrated 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. Due to the use of back-pressure saturation technique and equipment limitations, 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 3-3 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.

Table 3-3  Measured hydraulic conductivities in cm/s

<table>
<thead>
<tr>
<th>Clay</th>
<th>Water</th>
<th>Leachate</th>
<th>Prehydrated</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>L-1</td>
<td>L-2</td>
</tr>
<tr>
<td>AU</td>
<td>&lt; 9×10^{-9}</td>
<td>1.3×10^{-5}</td>
<td>4.5×10^{-8}</td>
</tr>
<tr>
<td>AT</td>
<td>&lt; 3×10^{-9}</td>
<td>1.2×10^{-6}</td>
<td>1.0×10^{-8}</td>
</tr>
<tr>
<td>BU</td>
<td>&lt; 1×10^{-8}</td>
<td>1.5×10^{-8}</td>
<td></td>
</tr>
<tr>
<td>BT</td>
<td>&lt; 1×10^{-8}</td>
<td>1.2×10^{-8}</td>
<td></td>
</tr>
<tr>
<td>CU</td>
<td>&lt; 3×10^{-9}</td>
<td>4.0×10^{-9}</td>
<td>8.5×10^{-6}</td>
</tr>
<tr>
<td>CT–1</td>
<td>&lt; 1×10^{-7}</td>
<td>1.4×10^{-6}</td>
<td>2.0×10^{-5}</td>
</tr>
<tr>
<td>CT–2</td>
<td>&lt; 5×10^{-8}</td>
<td>1.6×10^{-8}</td>
<td>7.5×10^{-7}</td>
</tr>
</tbody>
</table>

This implies that the cation exchange process occurs almost instantaneously upon initial saturation. On the other hand, prehydrated specimens exhibited a gradual increase in hydraulic conductivity, but a steady-state condition was always reached beyond a maximum quantity of flow of 3 pore volumes (Figure 3-1). Shackelford et al. 2000 reported a gradual increase in hydraulic conductivity up to at least 40 pore volumes of flow when a diluted CaCl2 solution was used. Ruhl and Daniel 1997 found that diluted 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 approximately $10^{-8}$ 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 3-3 indicate that all materials exhibited hydraulic conductivities higher 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 calcium and magnesium 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.

3.5.2 Effect of Prehydration

The results presented in Table 3-3 indicate a significant difference in hydraulic conductivity, depending on the initial wetting conditions. Prehydrated specimens performed, in general, far better than those exposed directly to the leachate, even at large numbers of pore volumes of flow (Figure 3-2). 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 sodium montmorillonite with a multivalent-rich solution, such as calcium chloride, causes the divalent cations (Ca\(^{2+}\)) to immediately occupy most of the cationic sites originally taken by the sodium. 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 3-3(a)).
Figure 3-2 Effect of prehydration on the hydraulic conductivity of specimens exposed to leachate L-3

In contrast, initial hydration of the monovalent cations (Na⁺) present in sodium bentonite attracts large quantities of water into the interlayer space, thereby creating a dispersed structure, as shown in Figure 3-3(b). 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 3-3(c)). 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 question the effectiveness of prehydration on the long-term hydraulic 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 prehydration are being currently 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 prehydrated L-3 leachate.

### 3.5.3 Relationship to Swell Index and Mineralogy

While performing the permeability experiments, higher 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 3-4. A mathematical model is proposed, together with the model statistics, in Chapter Four of this dissertation to relate swell index and hydraulic conductivity.

![Figure 3-4 Relationship between swell index and hydraulic conductivity for non-prehydrated specimens](image)

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 3-5 support their conclusion that the hydraulic conductivity ratio is constant for free swell ratios larger than 20. Here, the hydraulic conductivity ratio is defined as the ratio
between $k_{\text{leachate}}$ and $k_{\text{water}}$. 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 Figure 3-4 and Figure 3-5), was uncharacteristically high compared to the other materials; values as high as 50 and 90 ml/2g-clay were obtained with leachate L-3 and water, respectively. Based on the data presented, it is recommended that the swell index ratio be used with caution when correlating with the hydraulic conductivity ratio. In other words, the data at hand does not support the implementation of swell index ratio as an early indicator of bentonite compatibility with inorganic chemicals.
To evaluate the influence of the mineral composition on the hydraulic and swelling characteristics, a series of XRD tests was conducted. Aggregated clays were grinded to a size of approximately 5 microns and dried at a low temperature of 50°C to avoid shifting in the diffraction peaks. Unoriented powder samples were prepared following the Poppe et al. (2001) back-loading procedure, and tested in the Philips PW3040 Theta-2 diffractometer. Incident and diffracted beam optics settings were selected based on values recommended by Moore and Reynolds 1989. A more detailed description of the XRD testing program, results, and interpretation is given in Chapter Five.

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 (1989), and Hillier 2000. The results of the analyses are summarized in Table 3-4.

### Table 3-4 Mineralogical composition of clays from XRD analyses

<table>
<thead>
<tr>
<th>Mineral</th>
<th>Weight Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>AU</td>
</tr>
<tr>
<td>Montmorillonite</td>
<td>92</td>
</tr>
<tr>
<td>Illite</td>
<td>15</td>
</tr>
<tr>
<td>Quartz &amp; Cristobalite</td>
<td>3</td>
</tr>
<tr>
<td>Feldspar</td>
<td>4</td>
</tr>
<tr>
<td>Calcite</td>
<td>&lt;2</td>
</tr>
<tr>
<td>Gypsum</td>
<td>&lt;2</td>
</tr>
<tr>
<td>Siderite, Mica, and others</td>
<td>&lt;2</td>
</tr>
</tbody>
</table>

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 3-6 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 3-6 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, possibly because of the lower concentration of calcium in solution. 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 3-3, as well as with data published by Petrov et al. 1997 and Shackelford et al. 2000.

![Figure 3-6 Relationship between montmorillonite content and hydraulic conductivity of GCL clays](image)

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 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.

3.6 Summary

Hydraulic conductivity tests were conducted to evaluate the response of untreated and polymer-treated bentonites to water and ash fill leachates, and to compare the results to those published in earlier studies. The results were interpreted using methods and models available in the literature. The preliminary findings 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 prehydrated, 3) prehydration is generally more beneficial than the use of treated bentonites for up to at least eight pore volumes of flow, and 4) a relationship exists between the swell index and the hydraulic conductivity, as suggested in earlier studies. 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. Compared to untreated GCL clays with similar mineralogical composition, polymer-treated clays exhibit only limited improvement in terms of reduction in hydraulic conductivity.
Chapter 4 - Evaluation and Modeling of Permeability-Plasticity Relationship

4.1 Overview

In this chapter, the geotechnical index properties of the bentonite clay are measured, and their use as early indicators of chemical compatibility of bentonite is explored. The specific geotechnical index properties addressed in this study are liquid and plastic limits (Atterberg limits) and swell index. In addition, the advection, diffusion, and sorption characteristics of untreated and polymer-treated bentonite clays are examined in this chapter. Long-term advection properties are determined from the flexible and rigid wall permeability tests described in Chapter Three, where both chemical and hydraulic equilibrium are established. In this chapter, it is demonstrated that the cation exchange capacity, the swell index, and the clay plasticity ratio appear to be more reliable than the swell index ratio and the plasticity index as indicators of the hydraulic performance and chemical compatibility of bentonite permeated with inorganic chemicals.

4.2 Background

Little work has been conducted to relate the permeability of bentonite to its plasticity behavior. In a most recent study by Lee et al. 2005, no significant relationship was found between the plasticity index of bentonite clay and its hydraulic conductivity when permeated with inorganic solutions. In addition, only a few studies have been
conducted to evaluate the sorptive, diffusion, and retention characteristics of bentonite materials within the context of their exposure, as GCL components, to chemicals abundant in landfill leachates. While Egloffstein 1995 provided a general overview of candidate test methods for comprehensive evaluation of the chemical and hydraulic compatibility of GCLs, little research has since been conducted in this regard.

In this chapter, the experimental results presented in earlier chapters are complemented with additional tests to develop a relationship between permeability and index tests using a new framework. The results provide a broad perspective of the factors relevant to long term design of GCL-based leachate containment systems, while the model presents a specific methodology to be followed as a means of evaluating the chemical compatibility of a GCL material. The same untreated and polymer-treated bentonite materials described in earlier chapters were tested for hydraulic conductivity, ionic sorption characteristics, and index properties, while diffusion and retention data on only untreated bentonite are presented. In addition, rigid wall tests were also performed to verify the influence of sample preparation procedures and test methods on the results.

4.3 Materials

Eleven types of commercially available Wyoming sodium bentonite, with different index properties, were tested and are reported in this chapter, including the seven materials presented in Chapter Three. One additional untreated bentonite material (CU-2), and one polymer-treated bentonite (CT-3) were provided by the same manufacturers. While bentonite, in a general sense, is composed mostly of montmorillonite, the type and relative abundance of other minerals such as quartz, illite,
calcite, and feldspar, can have a significant influence on its engineering properties. Moreover, the properties of bentonite depend not only on the source and composition, but also on preparation processes such as wetting and drying, heating, chemical modification, and mechanical grinding. Engineering properties of identical GCL materials obtained from the same source that may be altered during manufacturing include consistency limits, swell properties, hydraulic conductivity, and long-term chemical compatibility.

4.4 Equipment and Specimen Preparation

Permeability tests were conducted using both flexible and rigid wall permeameters. The specimens were back-pressure saturated with water (prehydrated) or permeant (non-hydrated) before being subjected to a hydraulic gradient of 250. Special interconnected buffer tanks with different capacities were used to store the influent and effluent solutions, and were equipped with three-way valves to sample the solution for chemical analysis, as shown in Figure 4-1.

![Figure 4-1 Schematic of flexible wall permeability setup](image)
By sampling the leachate while the test is running, this setting allowed for verification of steady-state electrical conductivity and pH conditions (chemical stability) as an additional condition for terminating the tests. The void ratio in the rigid wall tests was controlled by maintaining a constant specimen height.

Specimen preparation is a crucial component of GCL testing, and the preparation procedure recommended in the ASTM D5887 and D6766 methods resulted in a non-uniform specimen thickness. Post-testing inspection of the permeated specimens indicated that the thickness was reduced near the edges due to swelling and material loss while cutting the sample. Channels and cracks around the specimen edges were also observed. To ensure homogeneity of the specimens, the bentonite component was first removed from the GCL, and a given weight of dry bentonite was tamped to a constant height of 7.5 mm into a 101-mm diameter rigid mold within the permeameter, in a procedure similar to that used to prepare sand samples. The rigid mold was removed before testing, and the specimen shape was preserved by applying vacuum before removing the mold. The specimens prepared using this method gave repeatable results, and post-testing visual inspection showed no sign of cracking. The only drawback of the method is that only the clay component of the GCL is tested as the geotextile backing is discarded. However, the method allows for unbiased comparisons between the hydraulic conductivities of different clays since the initial mass and thickness are constant. Figure 4-2 illustrates the importance of sample preparation on the measurement of hydraulic conductivity of GCL bentonite.
Samples of five different clay types prepared using the ASTM method exhibited hydraulic conductivity values up to two orders of magnitude greater than the USF method at an effective confining pressure of 30 kPa. The effect was particularly prominent when the samples were permeated with non-hydrated high concentration leachate from an ash landfill. The leachate contained calcium and magnesium chlorides at concentrations close to the saturation limit of the solution (Ca\(^{2+}\) at 5000 mg/L and Mg\(^{2+}\) at 2000 mg/L).

### 4.5 Sorption Characteristics

Exposure to inorganic chemicals causes gradual degradation of the hydraulic performance of bentonite, until the total sorptive capacity of the clay is reached. Sorption of inorganic cations occurs mainly through ionic bonding by replacement of the existing cations – usually Na\(^{+}\) – in the diffuse double layer of the clay with higher valence cations such as Mg\(^{2+}\) and Ca\(^{2+}\). The long-term resistance of bentonite to inorganic chemicals can
be enhanced by sorbing polymers or other organic compounds during preparation (Soule and Burns 2001; Ashmawy et al. 2002).

Depending on their chemical composition, sorption of organics occurs through a combination of ion exchange and adsorption by van der Waals forces (Soule and Burns 2001). The sorption of organic compounds in this case is viewed as beneficial, as the organic sorbents shield the clay surface from interactions with other detrimental organic or inorganic chemicals. However, the addition of large quantities of organics can cause the total sorptive capacity of the clay to be reached, and the hydraulic conductivity of the clay to consequently increase. This can be prevented by mixing small quantities of treated or amended clay into a larger quantity of pure bentonite. The treated fraction of the material adds to the chemical resistance, while the pure bentonite helps to maintain the low hydraulic conductivity.

The sorptive capacity of bentonite is related to its CEC, which can be measured directly or indirectly by a variety of methods including sodium saturation (Wentink and Etzel 1972), XRD analysis (Kaufhold et al. 2002), infrared chromatography (Hwang and Dixon 2000), or chemical adsorption of a wide array of chemicals. MB adsorption is widely accepted as one of the most reliable methods to obtain information on the properties of clay minerals, including CEC (Grim 1968; Santamarina et al. 2002). It is also used as an indirect quality indicator for swelling activity of clay materials, even though minerals which do not swell might also adsorb MB. The CEC of relatively pure montmorillonite, measured by MB adsorption, typically ranges from 70 to 130 mEq/100g. However, the presence of other minerals in bentonite can cause its CEC to fall within a lower range. The eleven GCL clays presented in this chapter exhibited CEC
values between 45 and 90 mEq/100g, as measured by MB adsorption. The relationship between CEC and hydraulic conductivity is discussed later.

Other indicators of sorptive capacity include swell index, XRD descriptors, and consistency limits (LL and PL). Because of its extremely high plasticity, it is often difficult to cut a groove into a bentonite paste, which makes it impossible to accurately determine the LL using the ASTM Standard (Casgrande) Method. The European (fall cone) method was used instead. The eleven materials presented in this chapter exhibited a wide range of plasticity behavior (Figure 4-3), but the majority fell within the typical range for montmorillonite.

![Figure 4-3 Liquid limit vs plasticity index of GCL bentonite materials](image)

The swell index was measured in both water and leachate. The relationship between hydraulic conductivity and both swell index and plasticity limits is discussed later. XRD tests were also conducted to observe the shift in diffraction angle or $d$-
spacing upon permeation with water and a high concentration calcium and magnesium chloride leachate from an ash landfill. All tests were conducted on air-dried bentonite powder specimens prepared using the back-loading method. While dry-prepared XRD specimens may reflect preferred particle orientations, the results provide an unbiased comparison between different specimens. Further description and analysis of the XRD test data is provided in Chapter Five.

The results shown in Figure 4-4 indicate no significant difference in the diffraction pattern between the as-received and water-permeated bentonites. In contrast, the bentonite underwent a distinct shift and broadening of the 7º peak upon permeation with the leachate. The peak at or around 7º is the characteristic peak for montmorillonite, and the shift signifies a change in the thickness of the interparticle spacing due to the associated cation exchange process. This shift from 7º to 6º corresponds to a change in $d$-spacing of approximately 17%, and the broadening of the peak indicates a larger range of interparticle spacing, possibly due to a partial exchange. Similar behavior was observed with all other bentonite samples, with the exception of T4 which exhibited a shift in the 7º peak and intensity upon permeation with water.
Figure 4-4  XRD pattern of T1 bentonite: (a) as received, (b) permeated with water, and (c) permeated with high concentration inorganic leachate

4.6 Hydraulic Conductivity vs Swell Index Relationship

Swell index and LL have been proposed as indicators of the hydraulic conductivity of bentonite (e.g., Shackelford et al. 2000; Jo et al. 2001). While the present data supports the argument that the hydraulic conductivity to water and leachate depends on the swell index, there is a clear distinction in behavior between treated and non-treated bentonites. This is due to the difference between clay and clay-polymer surface chemistries. The nature of clay-polymer-water interaction depends on the composition, ion size, and surface charge of the polymer. For instance, very high swell index values have been associated with high permeability values and vice versa for polymer-treated bentonite (Schenning 2004).
Figure 4-5  Relationship between hydraulic conductivity and swell index

The data presented in Figure 4-5 indicate that there is a strong dependency between the hydraulic conductivity and the swell index. To this end, non-linear regression analysis was performed on the data set using a number of candidate statistical functions, including logarithmic, exponential, and polynomial equations. Polynomial equations were quickly discarded as they do not provide reasonable boundary conditions such as asymptotic values at very low and very high swell index. Correlation coefficients were determined for the various candidate functions, and the mathematical model presented in Equation 4.1 resulted in the highest correlation.

\[
k = \left( -605.2 + \frac{2424}{S_i} \right)^{1/3}
\]

(4.1)

where \(k\) is the hydraulic conductivity in cm/s, and \(S_i\) is the swell index in ml/2g. The corresponding regression statistics are listed in Table 4-1.
Table 4-1  Regression statistics – hydraulic conductivity vs swell Index

<table>
<thead>
<tr>
<th></th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Correlation Coefficient (R)</td>
<td>0.891</td>
</tr>
<tr>
<td>Adjusted (R^2)</td>
<td>0.761</td>
</tr>
<tr>
<td>Standard Error</td>
<td>85.146</td>
</tr>
<tr>
<td>Observations</td>
<td>15</td>
</tr>
<tr>
<td>Standard Error in Intercept</td>
<td>± 36.2</td>
</tr>
<tr>
<td>Standard Error in Slope</td>
<td>± 359</td>
</tr>
</tbody>
</table>

4.7  Hydraulic Conductivity vs Plasticity Ratio Relationship

The plasticity ratio is a newly introduced parameter as part of this research, and is essentially an indicator of the relative change in soil plasticity when tested with leachate as opposed to water. To calculate the plasticity ratio, the consistency limits (LL and PL) of the sample are first measured using water, and the soil is mapped on the plasticity chart, as shown in Figure 4-3. The relative plasticity, a measure of how far the soil is from the U-line, is then calculated by dividing the plasticity index of the soil by the corresponding ordinate on the U-line. The LL and PL are then measured by mixing the soil with the leachate, instead of water, and the relative plasticity is calculated accordingly. The ratio between both relative plasticity values (leachate and water) is then defined as the plasticity ratio. This parameter can be viewed as the relative shift away from the U-line due to sample exposure to leachate.

Figure 4-6 presents the relationship between the plasticity ratio and the hydraulic conductivity ratio. A strong correlation is evident from the data presented on the graph. The conductivity ratio for each bentonite type is defined here as the ratio between the hydraulic conductivity to leachate and the hydraulic conductivity to water. A non-linear regression analysis was performed on the plasticity and hydraulic conductivity ratios.
resulting in the development of a mathematical model (Equation 4.2) that can be used as an early indicator to the permeability performance of non-prehydrated bentonite upon permeation with inorganic leachate solution. This model was, again, developed by testing nine different statistical functions (logarithmic, exponential, and polynomial) and selecting the one that provided the highest correlation coefficient.

\[ k_r = 0.2232 + \frac{0.4831}{(P_r - 0.35)^2} \]  

(4.2)

where \( k_r \) is the hydraulic conductivity ratio \( (k_{\text{leachate}} / k_{\text{water}}) \) and \( P_r \) is the plasticity ratio.

Table 4-2 lists the regression statistics associated with the non-linear regression analysis.

<table>
<thead>
<tr>
<th>Correlation Coefficient ((R))</th>
<th>0.997</th>
</tr>
</thead>
<tbody>
<tr>
<td>Adjusted ( R^2 )</td>
<td>0.993</td>
</tr>
<tr>
<td></td>
<td>(adjusted for number of observations)</td>
</tr>
<tr>
<td>Standard Error</td>
<td>3.71</td>
</tr>
<tr>
<td>Observations</td>
<td>9</td>
</tr>
<tr>
<td>Standard Error in Intercept</td>
<td>1.5</td>
</tr>
<tr>
<td>Standard Error in Slope</td>
<td>0.0147</td>
</tr>
</tbody>
</table>

Figure 4-6  Relationship between hydraulic conductivity and plasticity ratio
Figure 4-7 shows a general decrease in hydraulic conductivity to landfill leachate as the CEC increases, regardless of polymer treatment, although some level of scatter exists at high CEC values.

![Relationship between hydraulic conductivity and CEC](image)

**Figure 4-7  Relationship between hydraulic conductivity and CEC**

While extreme care was taken during sample preparation and testing, the reproducibility of the hydraulic conductivity values has only been verified for a limited number of tests. The initial results indicate that the range of variability on hydraulic conductivity is within 100% for flexible wall tests, and 50% for rigid wall tests. A consistent trend was, however, observed where flexible wall tests resulted in hydraulic conductivity values larger than rigid wall tests for specimens prepared at the same initial void ratio. The increase was as low as 60% for specimens permeated with water and as high as 450% for high concentration single species solutions. This difference is attributed to the inability of rigid wall specimens to swell vertically and laterally during
testing, which causes the void ratio to remain constant throughout. The hydraulic conductivity values reported in Figure 4-2 are for flexible wall tests while the hydraulic conductivity data reported in Figure 4-5 is for a flexible wall permeameter tests.

**4.8 Diffusion and Sorptive Retention Characteristics**

The diffusivity and retention of cations in bentonite depend on the void ratio, CEC, solution concentration, and cation type. Because of the small thickness of GCLs, diffusion plays an important role in solute transport, and can cause the bentonite to reach its sorption capacity within relatively short periods of time. Chemical diffusion can also cause substantial degradation to the hydraulic conductivity of the liner. Diffusion tests were conducted on pure Wyoming bentonite in conjunction with 1M, 2M, and 5M solutions of NaCl, CaCl₂, and MgCl₂. The range of values of effective diffusion coefficient, \( D^* \), and retardation factor, \( R_d \), is given in Table 4-3.

<table>
<thead>
<tr>
<th>Solution</th>
<th>CaCl₂</th>
<th>MgCl₂</th>
<th>NaCl</th>
</tr>
</thead>
<tbody>
<tr>
<td>Effective diffusion coefficient, ( D ) (m²/s)</td>
<td>1.8-5.5( \times )10⁻¹²</td>
<td>6.6( \times )10⁻¹²</td>
<td>5.2-8.0( \times )10⁻¹²</td>
</tr>
<tr>
<td>Retardation factor, ( R_d )</td>
<td>1.6-6.3</td>
<td>17.1</td>
<td>5.6-18.5</td>
</tr>
</tbody>
</table>

The low retardation factor values in the case of calcium indicate a rapid cation exchange process, resulting in collapse of the double layer and increase in the space available for ions to diffuse. This finding is important in understanding the rate of cation of exchange upon permeation with calcium and magnesium leachates. The low retardation factors support the notion that chemical equilibrium is reached within a short permeation duration, possibly within a few pore volumes. This conclusion was also verified through XRD testing as described later in Chapter Five.
4.9 Summary

Results were presented from studies on the hydraulic conductivity, diffusion, and sorption characteristics of polymer-treated and untreated GCL bentonite exposed to inorganic solutions. Hydraulic and chemical equilibrium were verified throughout each of the tests to ensure the simulation of long-term conditions. Polymer-treated bentonites retained their hydraulic conductivity after exposure to high concentration inorganic leachate, with the exception of one sample that exhibited high $k$ values when permeated with both water and landfill leachate. Swell index and plasticity ratio proved to be better indicators of the performance of polymer-treated clays than CEC. This may be attributed to the fact that the surface chemistry of polymer-treated clays does not follow the classical Gouy-Chapman double-layer theory. Changes to the double layer chemistry in both polymer-treated and untreated bentonite can be characterized by the shift in bentonite characteristic peak in the XRD trace. A statistical regression model was proposed to relate hydraulic conductivity with swell index. A second model was developed to correlate the conductivity ratio with a newly introduced parameter, the plasticity ratio. The second model is of immense value in evaluating the compatibility of GCL bentonite with inorganic leachates.
Chapter 5 - X-Ray Diffraction

5.1 Overview

In this chapter, all ten samples of both treated and untreated bentonite materials, permeated with ordinary tap water and inorganic ashfill leachate during hydraulic conductivity tests, were exhumed at the end of the experiment and subjected to XRD testing. The results indicate that the samples permeated with leachate underwent full ionic exchange whereby the higher valance calcium cations exchanged with the lower valance sodium cations thus resulting in a calcium montmorillonite. This chapter will present testing procedures, discuss results and provide recommendations.

5.2 Background

As part of the current research, a separate task was undertaken to determine whether the leachate permeation altered the soil samples’ mineralogical makeup. Specifically, the use of XRD analysis technique was undertaken to determine whether the samples have achieved chemical equilibrium, and whether significant alteration to the mineral structure has occurred upon permeation. The XRD process is highly reliable, albeit a destructive test in the sense that the permeability test must be terminated, the sample extracted and processed for the diffraction measurement. The inorganic leachate, obtained from the Pasco County facility, was analyzed for chemical composition at the USF Environmental Laboratory, and the results are summarized in Table 5-1 below.
Table 5-1  Leachate chemical composition

<table>
<thead>
<tr>
<th>Alkalinity</th>
<th>Conductivity</th>
<th>pH</th>
<th>Total Hardness</th>
<th>Ca^{++}</th>
</tr>
</thead>
<tbody>
<tr>
<td>72 mg/L as CaCO₃</td>
<td>37.6 mS/cm</td>
<td>6.52</td>
<td>20,000 mg/L as CaCO₃</td>
<td>12,800 mg/L as CaCO₃</td>
</tr>
</tbody>
</table>

5.3  Bentonite Soil Mineralogy

The contraction and expansion of the interlayer spacing in bentonite clay directly affects the permeability of the soil material. Highly swelling bentonites tend to have lower permeability than those with lower swelling capacity. In general, the smectites’ high swelling capacity is attributed to the weak interlayer attraction force which is insufficient to keep the interlayer spacing intact (Moore and Reynolds 1989).

Ca-montmorillonite and Na-montmorillonite are the most common minerals found in bentonite soils. Na-montmorillonite typically has a permeability that is an order of magnitude lower than that of Ca-montmorillonite. Because of its weak surface charge, Na-montmorillonite is vulnerable to atomic exchange when exposed to solutions containing exchangeable cations having higher valence than the sodium cations. For example, when sodium bentonite is permeated with a solution containing electrolytes in the form of calcium chloride, a cationic exchange process occurs whereby the higher valence calcium cations exchange with the lower valance sodium cations thus altering the mineralogical makeup of the soil and its inherent low permeability. The purpose of the XRD testing is to identify such changes in clay mineralogy and to establish whether or not full exchange between Na⁺ and Ca²⁺ has occurred during permeation.
5.4 XRD Principles

A basic overview of XRD theory was presented with the literature review in Chapter Two. In this Chapter, the theory and implementation are reviewed in more detail. X-rays are electromagnetic radiation of a wavelength equal to the size of an atom and are generated from the oscillation of electrostatic and electromagnetic fields perpendicular to each other and to the plane of propagation through space. XRD occurs when Bragg’s Equation 5.1 is satisfied.

\[ n\lambda = 2d \sin \theta \]  

(5.1)

where \( n \) is an integer, \( \lambda \) is the wavelength, \( d \) is the spacing between the lattice plane, sometimes referred to as basal spacing, and \( \theta \) is the diffraction angle. Figure 5-1 shows a schematic of the diffraction process, where the x-rays are refracted upon colliding with the atoms in the lattice structure.

![Figure 5-1 XRD pattern (reproduced from Whittig 1986)](image)

In three dimensional spacing, the interatomic spacing of each mineral is distinct producing a unique diffraction angle for each mineral. These diffraction angles can be used to identify the minerals present in the material being diffracted. For the XRD
process to be perfectly successful, the incident rays should be perfectly parallel, atoms must be perfectly ordered and the crystal should be perfectly oriented. When the incident beam waves hit the atoms and refract in phase, their amplitudes are amplified and form a refracted beam of higher amplitude and wider peak in what is known as constructive interference. Alternatively, when the incident beam waves are out of phase, they form smaller amplitudes which in turn results in a narrow peak due to destructive interference (Moore and Reynolds 1989).

5.5 XRD Methods

XRD can be performed using either a qualitative method, quantitative method or a combination of both. The qualitative method is used to identify the minerals that make up the sample with no regard to their quantities. The quantitative method is used to determine the relative concentrations of those minerals. For this research program, the qualitative method was used because the intent of the research is to: 1) identify the type of mineral present in the sample, 2) determine whether the sample has reached chemical equilibrium during permeation, and 3) determine whether the mineralogical makeup was altered following the permeation process.

5.6 Qualitative Analysis Method

The qualitative analysis is based on identifying the minerals associated with the diffraction maxima obtained from a diffracted sample. Identification can be accomplished wither by: 1) using the standard comparison method, or 2) comparing the diffraction spacing with known spacings of standard minerals. In the direct comparison method, the unknown mineral can be identified by comparing the resulting strongest
diffraction peak(s) with those of known mineral standards. Successive association of the peaks with minerals serves to identify the mineralogical make up of the soil sample. For this research program, the diffraction spacing was used to identify the minerals. Each mineral has a unique \( d \)-spacing value that serves to identify that mineral. Sodium montmorillonite, for example, has a \( d \)-spacing value of 12.4 Å (Moore and Reynolds 1989) while the \( d \)-spacing for calcium montmorillonite is 15 Å (Shang et al. 2002).

Following the diffraction process, commercial software was used to identify the minerals associated with the strongest peaks, followed by validation of the identified minerals by the operator. The software is a very effective means for identifying the minerals; on the other hand, it also tends to identify all possible minerals that could be associated with the peaks, resulting in an exhaustive list of common and uncommon minerals. The operator post-identification validation relies heavily on the experience and judgment of the user and serves to narrow down the list of identified minerals to a more plausible roster based on the geology and source of the material.

5.7 Quantitative Analysis Method

The quantitative analysis method requires deliberate and careful sample preparation, good quality data and a detailed understanding of the material being analyzed. This method is difficult to perform on clayey soils due to the variations in their chemical composition, random particle orientation and particle sizes.

The quantitative analysis can be conducted using either an internal standard method, or an external standard method. The choice of method depends largely on the type of material being analyzed. The internal standard method is typically used in
analyzing powdered soil specimens and is typically applied to mineral or materials for which the composition is unknown. The external standard method is used on solid materials like alloys and allows the quantification of one or more components in the material while the direct comparison method is only applicable to fully crystalline mixtures and does not require any standards.

The internal standard method requires a known quantity and weight of a standard substance to be added to the unknown mixture to be analyzed. The substance selected must have the same adsorption characteristics as the sample being analyzed. The mass absorption of a mixture need not be known in advance and any number of constituents in a mixture may be independently quantified. Unlike the external standard method, the mass absorption coefficient must be known in advance which may require either full elemental chemistry or prior knowledge of the chemistry, as in the case of alloys. All of the components in the mixture must be quantified for solution.

5.8 XRD Equipment

An x-ray diffractometer, in its simplest form, is composed of an x-ray source and a detector that picks up and records the diffracted rays as schematically illustrated in Figure 5-2 below.
The Philips PW 3040/60 X'Pert Pro XRD system, located in the College of Engineering Material Testing Laboratory at the USF, was used to perform the XRD analyses. The diffraction system is composed of an upper console that houses the instrumentation and a lower part containing the power supply and the measuring and control electronics. The soil specimen is held in a stationary horizontal position and the angle of incidence of the x-rays is varied by rotating the goniometer as illustrated in Figure 5-3 below.
To ensure complete identification of the soil minerals, the incident beam was set
to bombard the specimen at a scanning rate of $2\theta$/min over an angular range between $4^\circ$
and $70^\circ$ degrees. The $2\theta$ values were used to calculate the $d$-spacing by applying Bragg’s
law.

5.9 Sample Preparation

There is a strong dependency between the quality of the XRD results and the
sample preparation techniques. The specimen powder mass must contain large numbers
of crystals so that enough crystals will be oriented to ensure that every set of lattice
planes will be diffracted, and the crystals must be correctly oriented with respect to the
incident beam. Both of these conditions can be achieved with the proper specimen preparation.

For quantitative analysis, the specimen particles need to be pulverized to produce extremely fine particles in the range 1 to 5 μm (Brindley and Brown 1980) which will ensure that enough particles are “participating” in the diffraction process. For routine qualitative evaluation the recommended particle size is finer than a 325 mesh sieve (44 μm). Achieving the recommended particle size requires sample grinding using either a manual or mechanical method. Excessive grinding should be avoided so as not to distort the lattice planes or alter the mineralogy by excessive friction-induced heat. Mechanical grinding of the soil material should be used as a last resort because it could cause the disorder of the crystalline materials which will reduce the diffracted intensities.

Quantitative analysis relies heavily on the intensity ratios in identifying mineral concentrations, thus requiring the particles to be randomly oriented in the sample being diffracted. Preferred particle orientation causes distortion of the intensities and should be avoided. In qualitative analysis it is also desirable to have randomly oriented particles to produce the maximum number of overlapping reflection.

Randomly oriented specimen preparation can be achieved by using either vibrating table, side packing or back packing. Other advanced preparation methods include the coatings of minerals with various solutions like aerosol plastic spray, acetone and thermoplastic organic cement. Achieving a totally random orientation is extremely difficult and requires a great deal of experience whereas near-random orientation is more manageable.
5.10 Particle Size and Sample Grinding

Particle size distribution plays an important factor in achieving a meaningful XRD data. Coarse particles will produce “blemished” diffraction data and therefore a very fine particle distribution is desirable. Fine powders will include a sufficient number of particles to provide the maximum number of diffraction when bombarded with the incident beam. Achieving 44 μm particle size is very tedious and may involve a combination of wet sieving and gravity or centrifugal sedimentation. Thus, preparing the sample by crushing of the sample using pestle and mortar is the preferred method, at least at the start of the grinding process. The finer particles are separated by sieving the crushed material through No. 200 mesh sieve. The sieved material can be crushed further and sieved through finer mesh sieves until the desired particle size is attained. Achieving particles finer than No. 325 sieve would require special techniques such as grinding under water. Because of equipment limitations, particles passing the 200 mesh sieve (75 μm) were used to perform for the XRD for this research program.

5.11 Specimen Preparation

The bentonite materials were classified into the following three groups: 1) as-received bentonite; 2) bentonite permeated with water; and 3) bentonite permeated with leachate. Following the permeation process, the permeated samples were allowed to air dry at room temperature and then crushed using pestle and mortar and sieved through the No. 200 mesh. The sieved material was placed inside glass sample holders, securely capped and labeled accordingly.
There are various XRD sample preparation techniques that include back-loading, front-loading, side-loading and spray-drying. Achieving randomly oriented crystals is crucial to the quality of the XRD results and as such, smoothing of the specimen surface during preparation should be avoided to minimize preferential crystal orientation. Under this research program, the back-loading technique was used to prepare the samples. The technique entailed the placing of the sample holder over a strip of glass followed by randomly placing the crushed and sieved soil sample in the sample holder and the excess material was removed using a straight edge. The metal backing plate was placed on top of the soil and securely snapped into place on the sample holder. The sample holder was then carefully inverted and the glass strip was carefully removed so as not to disturb the soil material. The prepared sample was immediately placed in the XRD machine. The sample preparation procedure is detailed in Figure 5-4.

5.12 XRD Results and Discussion

The $d$–spacing of the diffracted samples was calculated using Bragg’s law and the 20 diffraction angle values; for the Phillips XRD instrument, the wavelength $\lambda = 1.5406$. The $d$-spacing values were graphically plotted for all samples as illustrated in Figure 5-5 through Figure 5-14.
Figure 5-4  Sample preparation technique for XRD testing
Figure 5-5  XRD pattern for BT (GSE new treated)

Figure 5-6  XRD pattern for AT (GSE old treated)
Figure 5-7 XRD pattern for BU (GSE new untreated)

Figure 5-8 XRD pattern for AU (GSE old untreated)
Figure 5-9  XRD pattern for CT-1 (CETCO old treated #1)

Figure 5-10  XRD pattern for CT-2 (CETCO old treated #2)
Figure 5-11 XRD pattern for CT-3 (CETCO old treated #3)

Figure 5-12 XRD pattern for CT-4 (CETCO old treated #4)
**Figure 5-13** XRD pattern for CT-5 (CETCO new treated #1)

**Figure 5-14** XRD pattern for CU (CETCO untreated)
5.12.1 Intensity and Peak Widths

The above figures were plotted with the diffraction intensity versus the basal spacing ($d$-spacing) to show the location of the peak position which is used to identify the mineral of concern. The term “intensity” as used in this qualitative XRD analysis refers to the diffraction intensity which is directly proportional to the instrument’s receiving slit, (detector) width. Increasing the detector’s width will result in sharper peaks and will have a direct impact on the length of the scanning time. The diffraction intensity data merely provides an insight to the kind of atoms and their position within a unit cell and is not an indication to the quantity of that mineral (Moore and Reynolds 1989).

It is important to understand that narrow peaks are caused by destructive interference during the XRD process while wider peaks are indicative of constructive interference. The peak’s width is a conceptual indication as to the quality of the diffraction data and has no relation to the quantity of the mineral in the diffracted sample. The relative width of the peak is also a measure of the uniformity and purity of the mineral.

5.12.2 Data Analysis

The data reveals that the tested samples, with the exception of CETCO Old Treated #3 and #4, contained Na-montmorillonite as the main mineral when diffracted in the as-received and water-permeated conditions. Upon leachate permeation followed by diffraction analysis, the data illustrate a distinct shift in the basal spacing ($d$-spacing) of the strongest peak from 12.4 Å (Na-montmorillonite) to 15 Å (Ca-montmorillonite). The CETCO Old Treated #3 sample appeared to contain Ca-montmorillonite under all
diffracted conditions which are an indication that the bentonite source used in the fabrication process contained an abundance of calcium cations. The CETCO Old Treated #4 appeared to contain Na-Montmorillonite in the as-received condition and Ca-Montmorillonite when permeated with water and leachate. Since water is chemically neutral and contains no exchangeable cations, it is not possible for the sample’s mineralogical makeup to have significantly changed upon water permeation. The reason for such a disparity remains inconclusive, although it is possible that the clay contained a calcium salt or hydroxide that was dissolved upon permeation with water, causing the calcium ions to replace the sodium in the clay double layer.

The samples that achieved a basal spacing shift from the 12.4 Å to the 15 Å imply that: 1) these samples have achieved chemical equilibrium during the permeation process, and 2) the final calculated permeability value is accurate and will remain constant for the life of the liner system because the exchangeable cations were completely exhausted. Figure 5-15 provides a comparison of the samples’ permeability values following permeation with water and leachate and prior to the XRD process.
The permeability data reveals that samples achieved low permeability values upon water permeation. Alternatively, the permeabilities increased upon permeation with leachate. The magnitude of permeability increase varies from one sample to another. For example, the permeability of the CETCO Untreated sample increased by as much as two orders of magnitude. Further, it appears that the permeabilities of the treated samples remained relatively low after extended leachate permeation, except for CETCO Old Treated #4. This conclusion supports the manufacturers claim that the polymer treatment will enable the bentonite to maintain its low permeability upon permeation with high concentration of inorganic liquids. The CETCO Old Treated #4 sample was the exception and achieved the highest permeability values upon leachate permeation. This could be attributed to its mineralogical makeup which indicated that the sample contained
a calcium compound to begin with. Similar observations hold true for the CETCO Old Treated #3 sample, which also contained Ca-Montmorillonite and achieved the second highest permeability values of all treated samples.

5.13 Summary

The XRD has proven to be an accurate and reliable test that can be performed to determine the mineralogical changes to clay specimens upon permeation with inorganic leachate. On the other hand, XRD is a time-consuming analysis that requires a good deal of experience and familiarity with sample preparation techniques, instrument operating procedure and results interpretation. Sample preparation is the most crucial step in the analysis and is the main source of errors. For example if the sample preparation results in a preferred particle orientation, the results will be erroneous and will not provide for a true determination of the sample’s mineralogical makeup.

The permeabilities results indicate that the amount of pore volume that was introduced into the samples during the permeation process was adequate to ensure chemical equilibrium. Simple calculations can demonstrate that such large pore volumes will take a number of years to be achieved, if ever, under actual field conditions. As such, it is the conclusion of this author that the treated samples will achieve low permeabilities upon exposure to highly concentrated inorganic liquids. It also appears that the treatment process achieved its desirable effect. However, the untreated samples also performed relatively well under the same conditions. Therefore, the decision whether or not to use a treated GCL for a nominal advantage in performance over an untreated GCL material rests solely on the design professional.
Chapter 6 - Conclusion and Recommendations

6.1 Summary

Alterations to the clay’s mineralogical makeup upon permeation with inorganic solution containing elevated concentration of electrolyte increase its permeability. The overall intent of this research was to validate whether the polymer-treated bentonite can maintain its low permeability upon permeation with aggressive inorganic solution, to compare its performance with that of the untreated bentonite, and to develop a rational procedure for evaluating the chemical compatibility of bentonite with inorganic chemicals based on index testing. To that end, permeability measurements were conducted using tap water and leachate generated from ashfill disposal facilities. Permeation with tap water was performed to establish a permeability base value that was used as a baseline to further evaluate the permeability performance of the leachate permeated soils.

The ultimate objective of this research was to explore whether an empirical model can be developed and used as an early predictor of the bentonite’s permeability performance prior to performing extended permeability duration tests. To that end, the index properties (swell index, plasticity index and MBA) of the various bentonite materials were measured and the resulting data was analyzed for meaningful correlation with the permeability. Analysis of the testing results concluded in the development of
two empirical models that can be used to predict the permeability of the bentonite material upon permeation with inorganic leachate prior to performing extended duration tests. These models use the swell index and the soil properties to predict either the permeability or the increase in permeability due to the presence of inorganic substances in the leachate. The developments of these models are addressed in Chapter Four and their practical applications are discussed in Section 6.4 below.

6.2 Conclusions

The results clearly indicate the existence of a strong correlation between the plasticity ratio and the permeability ratio, which can be used to establish the extent to which a particular bentonite is able to maintain its low hydraulic conductivity upon exposure to inorganic leachate. A mathematical relationship was also established between the swell index and the permeability value. Furthermore, the results indicate that the polymer treatment of the bentonite enables it, to a limited extent, to maintain its low permeability upon exposure to highly concentrated electrolyte solution. This finding should be qualified by further stating that the results are specific to the leachate chemical composition and the GCL liners used in this study. Actual results may vary when testing with leachate having different concentrations.

The results indicate that the water-permeated samples, both regular and polymer-treated, achieved comparable permeability values that remained constant for the permeation duration. This is a clear indication that the polymer treatment, in the case of water permeation, did not affect the permeability of the bentonite either adversely or favorably. However, when leachate was used as the permeant, the permeability of both
types of bentonite increased, albeit, by varying degrees. Overall, with the exception of one material (CETCO Old Treated #4), the polymer-treated bentonite samples achieved lower final permeability values than those of the untreated type. It was also concluded that Ca-montmorillonite is a more stable product than Na-montmorillonite in that its permeability is not adversely affected when permeated with leachate containing elevated concentrations of sodium, magnesium, and calcium cations.

### 6.3 Recommendations for Future Work

The findings of this research provided valuable information that can be used to further our understanding of the long-term permeability performance of the GCL materials. However, there remains a need to perform additional research on these materials in addition to further validate and generalize the findings established in this study. The scientific and the engineering communities can greatly benefit from the following research topics in the future:

1. There is a need to evaluate the long-term effect of concentrated organic compounds on regular and polymer-treated bentonite. This can be achieved by collecting leachate generated from landfills known to have high concentrations of organics that are typically detrimental to the permeability performance of the soil component (bentonite) of the GCL. The testing methods and analyses can parallel those followed in this study which include extended permeation with no prehydration of the soils with the organic leachate followed by XRD of the permeated sample, index tests (swell index, plasticity index, and MBA). The goals of the study should be to verify
whether or not the statistical models presented in this study are applicable in the case of permeation with organic leachate – as opposed to inorganic – and to evaluate whether there is a benefit in using the polymer-treated bentonite in those applications.

2. It is imperative to evaluate the use of Ca-montmorillonite as an alternate to Na-montmorillonite in applications containing elevated concentrations of electrolytes. The testing program can, again, parallel the steps implemented in this research, with the ultimate goal of defining the resistance of Ca-montmorillonite to ashfill leachate.

3. Additional swell index and permeability tests need to be carried out using inorganic leachate with varying inorganic concentrations to further evaluate the statistics of the mathematical models established in this study for the relationship between the swell index and permeability and for the relationship between the plasticity ratio and permeability ratio. This can be accomplished by testing bentonite against inorganic leachate from various ash monofills to cover a wide spectrum of electrolyte concentrations and establish the appropriate levels of confidence and error of the current empirical models.

4. The geotextile fabric used in the GCL permeated with inorganic and organic leachates have not been evaluated to determine whether these fabrics will experience any degree of degradation upon extended exposure to these contaminants. While the vast majority of existing data suggests that geotextiles can withstand inorganic chemicals, the resistance to specific organic compounds has not been adequately established. This can be
accomplished by directly submerging these fabrics in leachate for extended duration while observing changes, if any, to their physical composition.

5. It would be of great practical value to simulate the performance of the GCL under actual field conditions using a scaled landfill cell. This can be accomplished by using plexiglas panels to form a rectangular box equipped with leachate detection system overlain by GCL topped with a layer of bottom ash generated from the incineration of MSW. The box should remain uncovered and placed in an open environment to allow for the percolation of rainwater into the material. To further simulate actual field conditions, additional layers of ash could be added at pre-determined intervals. The rate of leachate flow from the detection system can be observed over time and the variation of the flow can be used to evaluate the performance of the GCL. The chemical composition of the leachate at the start of the testing should be determined and used as a basis to determine when chemical equilibrium is achieved. A representative sample of the GCL can then be obtained to measure the permeability value in the lab and to examine the changes in mineralogy using XRD.

6. Throughout this research, the need became evident to devise a practical and cost-effective method to identify when chemical equilibrium is achieved during the permeation process. One possibility is to add and mix a known quantity of inert dye material to the bentonite prior to the permeation process and observe the change in the effluent color. A calibration process could be undertaken to determine the rate of leachability of the dye under actual testing
conditions and its relationship with chemical influent-effluent equilibrium. This determination can be correlated to the number of pore volumes introduced into the permeating sample.

6.4 Engineering Implications

The short-term duration index tests used in this study along with the mathematical models established are valuable tools that can be used by the design professional at an early stage in the design process to determine whether further testing and evaluation of the material under design consideration is warranted. These tools will greatly benefit the engineering community by saving valuable time and by reducing the costs associated with lab testing.

It should be noted that these short-duration tests are not intended to replace the permeability test and the design professional should perform the extended duration permeability tests after the initial screening of the material through the index testing. For example, in extremely environmentally sensitive engineering projects, the design professional may want to use XRD analysis to have a high level of confidence as to the composition and mineralogical makeup of the material. Extended duration permeability tests under leachate-specific conditions may need to be carried out if the long-term compatibility of the GCL bentonite is in question.

One of the key contributions of the current research is the development of methods for early determination and/or quality control of GCL bentonite compatibility. To this end, the design professional should obtain a sample of the GCL material from the manufacturer and associated certification. Upon receipt of the sample, the bentonite components of the GCL should be separated from the geotextile fabric and securely
capped in laboratory-approved air tight containers. A certified geotechnical lab can run both the swell index and plasticity index testing on the bentonite using both water and actual landfill leachate. If the design is for a new facility, a leachate sample can be obtained from existing landfills with similar waste characteristics. When the results are available, the design professional can use the mathematical models presented in this dissertation to decide whether further testing of the material is warranted.

For the swell index results, the following relationship can be used to predict the permeability performance of the GCL upon permeation with the intended leachate:

\[
k = \left( -605.2 + \frac{2424}{S_i} \right)^{1/3}
\]  

(6.1)

where \( k \) is the hydraulic conductivity in cm/s, and \( S_i \) is the swell index in ml/2g. Based on the mathematical model and the results obtained in this study, bentonite material having a swell index of 25 ml/2g or higher should have a final permeability in the range of \( 10^{-8} \) cm/s and lower, which is acceptable by USEPA and all state-EPA standards. The design professional can also plot the results of the plasticity index tests for both water and leachate on a standard Unified Soils Classification chart with the U-line and A-line clearly depicted. The material’s plasticity ratio (leachate/water) can then be calculated by measuring the relative distance of the plotted data points to the U-line for both water and leachate conditions and taking the ratio between those values, as described in Chapter Four.
For example, consider the data presented in Figure 6-1 for a bentonite material tested with water and leachate. The liquid limit and plasticity index of the material, as determined using water and leachate, are (130, 90) and (110, 50), respectively. To calculate the plasticity ratio of the material shown in Figure 6-1, the following steps are implemented:

Calculate the ordinate to the U-line corresponding to the “water” liquid limit of 130. The equation of the U-Line is given by Equation 6.1:

\[ PL_u = 0.9 \times (LL - 8) \]  \hspace{1cm} (6.1)

Where \( PL_u \) is the plasticity index (y-coordinate) of the U-line corresponding to a liquid limit \( LL \). Thus for a liquid limit of 130, the Plasticity Index coordinate for the U-line equals to 109.8. The relative distance to U-Line (relative plasticity) is then calculated as the ratio (90/109.8) which is equal to 0.820. The same procedure is repeated using the “leachate” liquid limit and plasticity index values. The coordinates corresponding to the
material tested with leachate from Figure 6-1 above are (110, 50). From Equation 6.1, for a liquid limit of 110, the Plasticity Index coordinate is equal to 91.8. The relative distance of the material to the U-Line (relative plasticity) is calculated as (50/91.8), which is equal to 0.545. The plasticity ratio is then calculated by dividing the relative plasticity for the leachate by the relative plasticity for the water. For this material, the plasticity ratio is calculated at 0.545/0.820=0.66.

It has been established through the models described by Equation 6.2, which was presented earlier in Chapter Four, that the higher the material’s permeability ratio, the smaller the permeability ratio:

\[ k_r = 0.2232 + \frac{0.4831}{(P_r - 0.35)^2} \]  

where \( k_r \) is the hydraulic conductivity ratio (\( k_{\text{leachate}} / k_{\text{water}} \)) and \( P_r \) is the plasticity ratio. A material with a plasticity ratio of 1.0 is expected to undergo minimal changes upon permeation with the leachate, whereas a material with a plasticity ratio close to 0.4 is expected to exhibit a drastic increase in permeability when permeated with leachate, compared to water. Based on the mathematical model presented by Equation 6.2, the plasticity ratio for the example above (0.66) results in a permeability ratio of 5.25.

In general, a permeability ratio of 10 should be deemed acceptable since it signifies a potential increase of one order of magnitude upon permeation with leachate compared to water, provided that the requirement for maximum acceptable permeability as specified by the regulatory agency is satisfied. In most cases, the permeability of GCL bentonite as provided by the GCL manufacturers is smaller than \( 10^{-9} \) cm/s. As such, a
permeability ratio of 10, corresponding to a plasticity ratio of approximately 0.6, will result in a hydraulic conductivity of $10^{-8}$ cm/s, which is acceptable by all federal and state EPA standards. Therefore, as a first indicator of quality of GCL, it is recommended that a plasticity ratio of 0.6 or higher be achieved.
References


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