Effect of Inorganic Salt Solutions on the Consistency Limits of Two Clays

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Abstract

Due to its low permeability, a clay liner is the main material used in solid waste disposal landfills. It is exposed there to various chemical, biological, and physical events, and the clay liner is affected by the resulting leachate. For this reason, when attempting to define the geotechnical characteristics of clay liners, the use of distilled water or tap water is far from being representative of the in-situ conditions. In this study, the effects of 4 different salt solutions on the consistency limits of clays were experimentally investigated. The salt solutions used as leachate compounds were ammonium chloride (NH_4Cl), potassium chloride (KCl), copper(II) sulfate ($CuSO_4$), and iron(II) sulfate ($FeSO_4$). Consistency limit tests were conducted on low plasticity (CL-class) and high plasticity (CH-class) commercial clays using both distilled water and these salt solutions. The experimental results indicated that the liquid limit increased when the salt concentration for the CL clay was increased. Moreover, the liquid limit was found to decrease when the salt concentration for the CH clay was increased. The plastic limit of the CL clay increased as the salt concentration increased to a certain value. However, the plastic limit of CH clay decreased at low salt concentrations and increased at high salt concentrations. In addition, the test results indicated that salt solutions at high concentrations changed the soil class of clays. In other words, for the conditions investigated, CL and CH class clays were transformed into low plasticity silt (ML) and high plasticity silt (MH) class soils, respectively, according to the Unified Soil Classification System (USCS).

Key words: Clay, Consistency limits, Landfill, Leachate, Salt solutions, Solid waste

Introduction

Because population increase leads to an increase in consumption, waste disposal has become one of the most serious modern environmental problems in developed and developing countries all over the world. One of the preferred methods of dealing with this kind of environmental problem is to dispose of the waste in sanitary landfills. Because of its low permeability, a clay liner is the main material used in such landfills. Clay liners are now commonly used to limit or eliminate the movement of leachate and landfill gases from the landfill site. The liners are exposed there to various chemical, biological, and physical events, and they are affected by the resulting leachate. For this reason, when attempting to define the geotechnical characteristics of clay liners, the use of distilled water or tap water is far from being representative of the in-situ conditions.

The Atterberg limits have been repeatedly shown to be useful indicators of clay behavior (Jefferson and Rogers, 1998). The liquid limit and plastic limit of a type of soil can be correlated with various engineering properties, such as permeability, shrinking and swelling behavior, shear strength, and compressibility of the soil (Sharma and Lewis, 1994; Abdullah et al., 1999). Consequently, if the consistency limits of the soil are determined, some other geotechnical

properties whose determination may take a long time can be estimated easily with acceptable accuracy. In addition, the evaluation of the consistency limits provides some very basic mechanical data about the soil and also a first insight into the chemical reactivity of clays. Basically, the liquid limit and the plasticity index are highly and mainly influenced by the ability of clay minerals to interact with liquids (Schmitz et al., 2004) and the hydraulic conductivity tends to decrease when the liquid limit and the plasticity index are increased (Alawaji, 1999; Met et al., 2005).

There are a limited number of studies in the literature on the effects of solutions composed of leachate on the consistency limits of clay liners. Most researchers focused on the investigation of the hydraulic conductivity of higher activity clays such as bentonite or geosynthetic clay liners (GCLs). Gleason et al. (1997) examined the effect of different concentration of $CaCl_2$ on the hydraulic conductivity of calcium and sodium bentonite. Jo et al. (2001) investigated hydraulic conductivity and swelling of nonprehydrated GCLs permeated with single-species salt solutions such as LiCl, NaCl, KCl, $CaCl_2$, $MgCl_2$, $ZnCl_2$, $CuCl_2$, and $LaCl_3$. Jo et al. (2005) conducted experimental tests to research the long-term hydraulic conductivity of a GCL permeated with some inorganic salt solutions (i.e. NaCl, KC, and $CaCl_2$). Shackelford et al. (2000) studied the hydraulic conductivity of GCLs permeated with non-standard liquids such as NaCl, $ZnCl_2$, and $CaCl_2$. Lee and Shackelford (2005) and Lee et al. (2005) used $CaCl_2$ as the testing liquid for the determination of the hydraulic conductivity of GCLs. All of the researchers pointed out that the hydraulic conductivity of bentonite clay or GCLs increased when the concentration of salt solutions was increased.

Gleason et al. (1997) also investigated the consistency limits of Ca and Na-bentonite with different concentrations of $CaCl_2$ (varying between 0.01 and 0.735 M), NaCl (varying between 0.01 and 0.1 M), and methanol (pure methanol and 50% methanol in distilled water), and gasoline. They reported that, when mixed with a strong $CaCl_2$ solution, calcium and sodium bentonite had approximately the same liquid limit and plasticity index. Lee et al. (2005) also investigated the effect of $CaCl_2$ solutions with concentration of 5, 10, 20, 50, 100, and 500 mM on the liquid limits of GCLs. Similar studies were carried out by van Paassen (2002), di Maio (1996), and Schmitz et al. (2004). They determined a reduction in liquid limits of bentonite and colclay (i.e.

an industrial smectite clay) when NaCl, KCl, and $CaCl_2$ solutions at different concentrations (varying between 0.01 and 4 M) were added.

Furthermore, Park et al. (2006) studied the effects of surfactants (octylphenol polyoxyethylene, biosurfactant, and sodium dodecyl sulfate) and electrolyte solutions ($NaPO_3$ and $CaCl_2$) on some properties of 2 soil samples (100% kaolinite clay soil and 30% kaolinite + 70% sand). They found that $CaCl_2$ solution did not affect the liquid limit significantly but decreased the plasticity index of kaolinite soil. Similarly, Sivapullaiah and Manju (2005) investigated the same geotechnical properties of a low plasticity soil ($w_L = 38\%$) using NaOH solution. They reported that the liquid limit of the test soil increased with increasing NaOH concentration due to the forming of new swelling compounds.

The present study was undertaken to investigate the effect of some leachate components on the consistency limits of clay liners. Consistency limit tests were carried out on 2 different commercial clays (i.e. low plasticity CL-class and high plasticity CH-class clays). Four different salt solutions (i.e. ammonium chloride (NH_4Cl), potassium chloride (KCl), copper(II) sulfate ($CuSO_4$), and iron(II) sulfate ($FeSO_4$) solutions) were chosen as leachate components in the tests. The tests were repeated at 8 different values of salt solution concentrations (i.e. 0.0001, 0.001, 0.01, 0.05, 0.1, 0.2, 0.5, and 1 M). When selecting the salt solutions and the values of their concentrations, the leachate components given in the literature were taken into consideration (Jo et al., 2001; Schmitz et al., 2004; Lee et al., 2005; Arasan and Yetimoğlu, 2006). Some of the tests were repeated as many as 4 times to assure the repeatability of the results. Test results were compared with those in the literature and discussed.

Materials and Methods

Clays

Two different classes of commercial clay soils were used in the tests. According to the Unified Soil Classification System (USCS), the class of one soil is low plasticity clay (i.e. CL) and the class of the other is high plasticity clay (i.e. CH). The CL and CH clays were supplied from Tokat and Askale, respectively. The same clays were used previously in similar studies (Arasan and Yetimoğlu, 2006; Yılmaz et al., 2008a; Yılmaz et al., 2008b). Some index properties of the clays are given in Table 1 and the grain

size distributions of clays are shown in Figure 1. The CL and CH clays have 55% and 56% silt content, respectively. Moreover, the hydraulic conductivity tests with distilled water were conducted using the rigid-wall compaction mold permeameter in accordance with the procedure described in ASTM D5856. The hydraulic conductivities (k) of CL and CH clays are 1.408×10^{-7} cm/s and 7.641×10^{-9} cm/s, respectively. According to The United States Environmental Protection Agency (USEPA), the compacted soil liner must be at least approximately 60 cm thick and must have a hydraulic conductivity of no more than 1×10^{-7} cm/s (USEPA, 1993). Additionally, according to The Turkish Solid Waste Disposal Regulation the compacted soil liner must be at least 0.6 m thick and must have a hydraulic conductivity of no more than 1×10^{-6} cm/s (Anonymous, 1991). Hence, it could be judged that the clays have appropriate hydraulic conductivities to be used as clay liners.

Table 1. Index properties of clays (Arasan and Yetimoğlu, 2006; Yılmaz et al., 2008a).

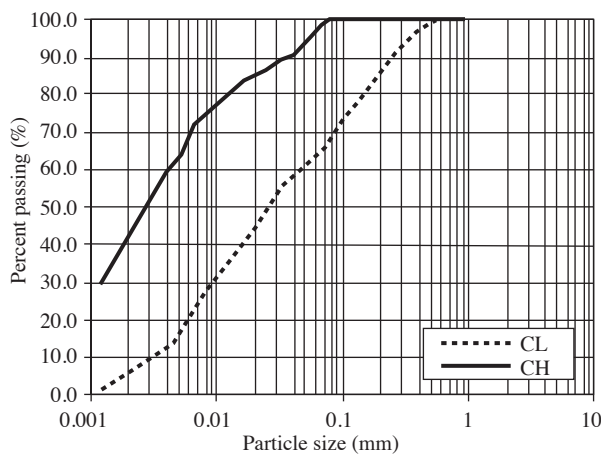|		CL Clay	CH Clay
Clay Content,	< 0.002 mm (%)	7	43
Finer Content,	<0.075 mm (%)	62	99
Specific Gravity,	G_S	2.76	2.79
Liquid Limit,	w_L (%)	26	113
Plastic Limit,	w_P (%)	19	38
Plasticity Index,	I_P (%)	7	75
Activity,	A	1.00	1.74

Figure 1. Grain-size distribution of the clays.

Salt solutions

Leachate may be defined as liquid that has percolated through solid waste and has extracted, dissolved, or suspended materials. In most landfills, leachate is composed of the liquid that has entered the landfill from external sources, such as surface drainage, rainfall, groundwater, and water from underground springs and the liquid produced from the decomposition of the wastes, if any. When water percolates through solid wastes that are undergoing decomposition, both biological materials and chemical constituents are leached into solution. The constituents of leachate can be divided into subgroups of organic carbon compounds, nitrogen compounds, anion, and metal groups. The chemical composition of the leachate will vary greatly depending on the age of the landfill and the events before the time of sampling (Tchobanoglous et al., 1993). Many researchers have studied the chemical composition of landfill leachate (Ehrig, 1988; Tchobanoglous et al., 1993; Vadillo et al., 1999; Kjeldsen et al., 2002; Jorstad et al., 2004). Typical data on the composition of leachate suggested by different researchers are summarized in Table 2.

When evaluating clay soils in their application as impervious liners in landfills, it is necessary to study the properties of the clays not only with distilled water but also with leachate compounds. The solutions used in this study were selected so as to cover as many types and high concentrations of leachate components given in Table 2 as possible. Ammonium chloride (NH_4Cl), potassium chloride (KCl), copper(II) sulfate ($CuSO_4$), and iron(II) sulfate ($FeSO_4$) salt solutions were chosen to represent nitrogen, alkali metal, and heavy metal components of leachate.

Distilled water was used as the reference solution. Salt solutions for the tests were prepared in 8 different concentrations (i.e. 0.0001, 0.001, 0.01, 0.05, 0.1, 0.2, 0.5, and 1M) by dissolving powdered salts in distilled water at solubility temperature. The concentration values used widely in the literature were accounted for when selecting the concentration range used in the present study. Some properties of the salt solutions are shown in Table 3.

Consistency Limit Tests

Consistency limit (i.e. liquid limit and plastic limit) tests were carried out following the procedure in BS

1377, Part 2, 1990. The cone penetrometer (fall cone) method was used to determine the liquid limit. For the liquid limit tests, specimens were prepared by mixing an air-dried clay mass (passing the 425-μm (no. 40) sieve) with each salt solution. Salt solutions were added to the clay mass in small amounts and mixed very slowly and thoroughly for more than approximately 30 min until the clay mass became a homogeneous paste. To let the solutions permeate through the clay mass, the paste was allowed to stand in an airtight container for about 24 h prior to testing. After this tempering, the paste was remixed with each salt solution thoroughly for at least 15 min before performing the first trial. A minimum of 4 trials were performed for each salt solution concentration.

The plastic limit tests were performed on material prepared for the liquid limit test. The average of the water contents (trial plastic limits) determined in the 4 tests was taken as the plastic limit. If the difference between the trial plastic limits was greater than the acceptable range, the plastic limit test was repeated. Both liquid and plastic limit tests were conducted at room temperature.

Table 2. Typical compositional properties of leachate in landfills.

		Kjeldsen et al. (2002)	Ehrig (1988)	Tchobanoglous et al. (1993)	
				New landfill (less than 2 years)	Mature landfill (greater than 10 years)
pH		4.5-9	4.5-9	4.5-7.5	6.6-7.5
Constituent		Concentration (mg/l)			
Organic	COD	140-152,000	500-60,000	3000-60,000	100-500
Carbon	BOD_5	20-57,000	20-40,000	2000-30,000	100-200
Constituent	TOC	30-29,000	-	1500-2000	80-160
Nitrogen	Org.N	14-2500	10-4250	10-800	80-120
Constituent	NH_3-N	50-2200	30-3000	10-800	20-40
Anions	Cl^-	150-4500	100-5000	200-3000	100-400
	PO_4^{2-}	0.1-23	-	5-100	5-10
	SO_4^{4-}	8-7750	10-1750	50-1000	20-50
Metals	Na	70-7700	50-4000	200-2500	100-200
	Mg	30-15,000	40-1150	50-1500	50-200
	K	50-3700	10-2500	200-1000	100-200
	Ca	10-7200	10-2500	200-3000	100-400
Heavy Metals	Mn	0.03-1400	0.03-60	-	-
	Fe	3-5500	3-2100	50-1200	20-2000
	Cr	0.02-1.5	0.03-1.6	-	-
	Ni	0.015-13	0.02-2.05	-	-
	Cu	0.005-10	0.04-1.4	-	-
	Zn	0.03-1000	0.03-120	-	-
	Cd	0.0001-0.4	0.0005-0.14	-	-
	Pb	0.001-5	0.008-1.02	-	-

Table 3. Some properties of the salts used in the tests.

Chemicals	Molecular Weight (g)	Solubility [g/100 g H_2O (°C)]
$FeSO_4.7H_2O$	278.03	40.3 (40)
$CuSO_4.5H_2O$	249.69	29.0 (40)
KCl	74.5	34.2 (20)
NH_4Cl	53.5	37.6 (20)

Results and Discussion

In the following sections, the effects of salt solutions at varying concentrations on the liquid and plastic limits of CL and CH clays are presented. The findings of the experimental tests are compared with those of other studies in the literature and discussed.

Effect of salt solutions on the consistency limits of CL clay

The variation in the liquid limit with the concentration of the salt solutions for CL clay obtained from the tests is given in Figure 2. For all salt solutions used in the tests, it is seen that the liquid limit generally increases when the concentration is increased. The increase is more pronounced for salt concentrations up to approximately 0.2 M, beyond which the rate of increase in the liquid limit is less significant. In other words, increasing the concentration of the salt solution beyond 0.2 M would not significantly increase the liquid limit. Figure 2 also indicates that, for NH$_4$Cl, FeSO$_4$, CuSO$_4$, and KCl salt solutions, the way the liquid limit varies with concentration is only slightly different for each solution. In other words, almost the same liquid limits are obtained from the tests at the same concentration values for the salt solutions used in the present study.

Similarly, the test results indicated that the plastic limits of CL clay increased with increasing salt concentrations up to around 0.2 M (Figure 3). It should be pointed out that the plastic limit test could not be performed at higher salt concentrations. Salt concentrations greater than 0.2 M appeared to damage the clay fabric; hence, the clay behaved as a non-plastic soil.

It should be pointed out that there has not been a general consensus regarding the effect of leachate compounds on the consistency limits of clays. Sridharan et al. (2002), studying 3 marine clays, reported that the liquid limit of Isahaya clay increased with increasing ion concentration, whereas the liquid limit of the other 2 clays decreased as concentration increased. This different behavior has been explained by the flocculation mechanism of kaolinite (nonswelling) clays and diffuse double layer (DDL) theory by the authors. Similarly, Bowders and Daniel (1987) stated that the many chemicals tended to reduce the thickness of the diffuse double layer (DDL), causing the soil skeleton to shrink and decrease in repulsive forces, thus promoting flocculation of clay particles, and to dehydrate inter-

layer zones of expandable clays, which subsequently became gritty or granular. Furthermore, Sharma and Lewis (1994) reported that the net electrical forces between clay mineral layers were affected by the concentration and valence of cations. They indicated that increasing cation concentration or cation valence would result in a decrease in net repulsive forces, hence causing clay particles to flocculate.
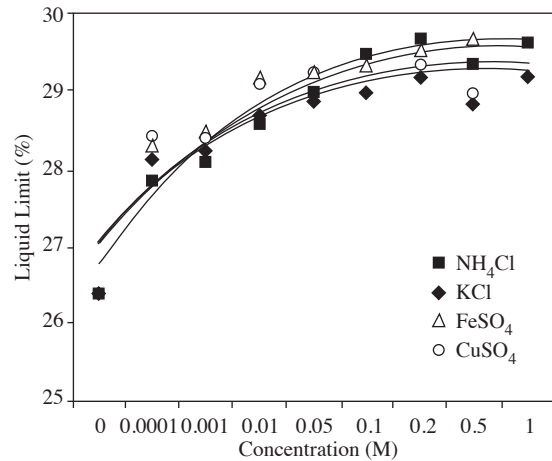

Figure 2. Effect of salt concentrations on the liquid limit of CL clay.
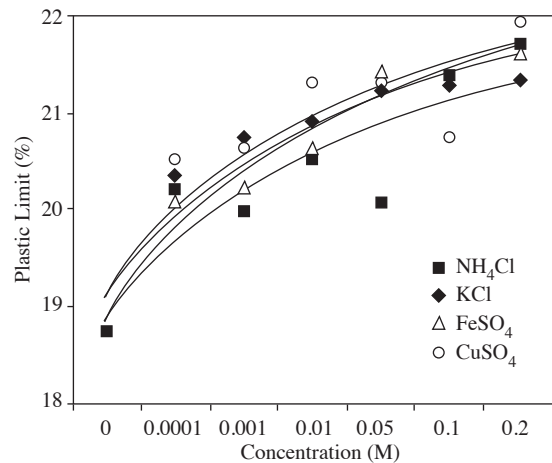

Figure 3. Effect of salt concentrations on the plastic limit of CL clay.

Similar to the findings of the present study, Arasan and Yetimoğlu (2006), based on their experimental study on a CL clay, pointed out that both the liquid limit and the plastic limit somewhat increased when the concentration of salt solutions was increased. On the other hand, Park et al. (2006) indicated that CaCl$_2$ solution did not

significantly affect the liquid limit of low plasticity kaolinite clay. Rao and Mathew (1995), based on their experimental study with marine clay, indicated that the clay particles were dispersed when the clay interacted with chemicals. Due to dispersion and deflocculation of clay, the geotechnical properties (especially, hydraulic conductivity) of clay were significantly changed. Hence, the increase in consistency limits could be attributed to dispersion of the clay particles when CL clay was permeated with salt solutions. Additionally, salt solutions might cause the formation of new swelling compounds and these new compounds might have increased the liquid limit of CL clay as indicated in Sivapullaiah and Manju (2005).

Effect of salt solutions on the consistency limits of CH clay

The test results indicated that for CH clay the effects of nitrogen and alkali metal salts (i.e. NH_4Cl and KCl) on the liquid limit were somewhat different from those of heavy metal salts (i.e. $FeSO_4$ and $CuSO_4$). Figure 4 shows the variation of the liquid limit with NH_4Cl and KCl concentrations. It can be seen that the liquid limit was drastically decreased as the NH_4Cl and KCl concentrations increased up to around 0.2 M. For concentrations greater than 0.2 M, the change in the liquid limit was insignificant.

On the other hand, the test results indicated that the liquid limit decreased proportionally with increases in the concentration of heavy metal salt solutions up to approximately 0.2 M, beyond which the liquid limit tended to increase when the concentration was increased. The variation in the liquid limit with $FeSO_4$ and $CuSO_4$ concentrations is given in Figure 5.

The effect of the salt solutions on the plastic limit of CH clay is shown in Figure 6. It is seen that all salt solutions exhibit almost the same variation. The plastic limit decreased for salt concentrations between 0.0001 and 0.001 M, and then increased for salt concentrations between 0.001 and 0.2 M. For concentration values higher than approximately 0.2 M, salt solutions did not significantly affect the plastic limit, and the plastic limit values approached those of the raw clay sample.

Similar to the findings of the present study, several researchers have reported that chemical solutions at low concentrations are more effective than at high concentrations for CH clays (Gleason et al.,

1997; Alawaji, 1999; Jo et al., 2001). Similarly, some researchers have also indicated that the liquid limit decreased with increasing salt concentration for CH clays (Gleason et al., 1997; Schmitz et al., 2004). Therefore, it appears that the salt solutions tended to reduce the thickness of the DDL and flocculate the CH clay particles, resulting in a reduction in the liquid limit of CH clay.
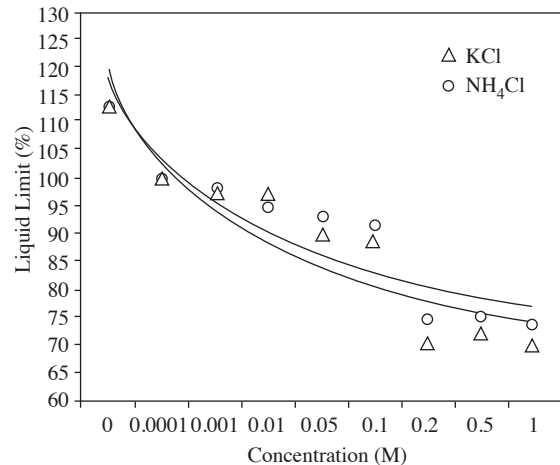

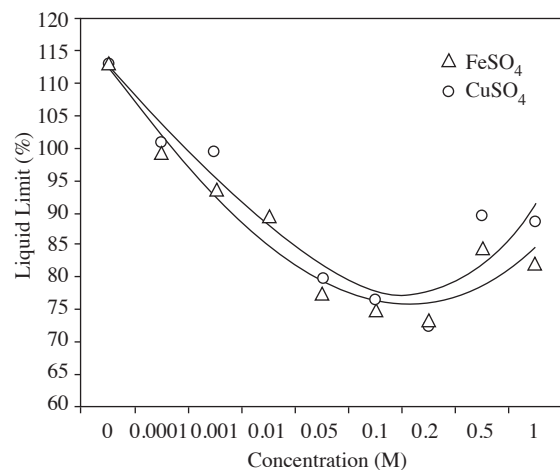**Figure 4.** Effect of NH_4Cl and KCl solutions on the liquid limit of CH clay.

Figure 5. Effect of $CuSO_4$ and $FeSO_4$ solutions on the liquid limit of CH clay.

The difference between the effect of heavy metal salts and the other salts is most likely due to the difference in the cation valence. Shackelford et al. (2000), Jo et al. (2001), and Kolstad et al. (2004) reported that the effects of the divalent and trivalent cations on the clays were different from those of

monovalent cations. Shackelford et al. (2000) indicated that the thinnest double layer and the smallest swell were obtained with trivalent cations, while monovalent cations had little effect on the thickness of the double layer and the swelling. Similarly, Mishra et al. (2005) reported that the divalent cations were more effective than the monovalent cations from the standpoint of permeability and compressibility of mixtures of ballast soil and bentonite permeated with NaCl and $CaCl_2$ solutions.
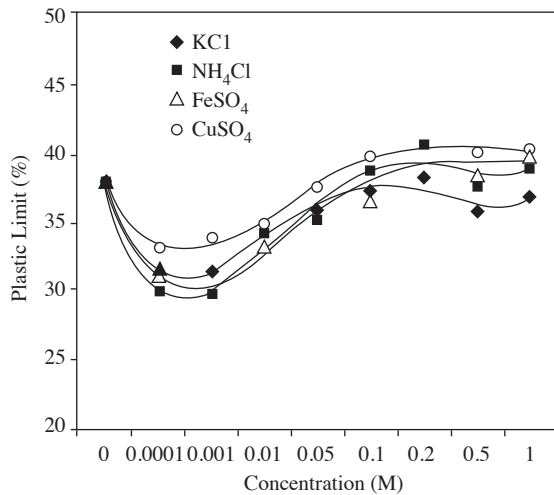

Figure 6. Effect of salt solutions on the plastic limit of CH clay.

Effect of salt solutions on the clay classification

The consistency limit test results were marked on the Cassagrande Plasticity Chart in order to determine the new soil classification according to the Unified Soil Classification System (USCS). Figure 7 shows the change in soil class after the tests. It is clearly seen that the points representing soil class come close to the A-line, and then drop under the A-line when the concentration of all salt solutions is increased. Consequently, the CL class clay was transformed to the ML (i.e. low plasticity silt) class soil at high salt concentrations (i.e. more than 0.2 M).

On the other hand, for the CH class clay, the points representing soil class firstly dropped under the A-line at low concentrations (i.e. less than 0.2 M) of all salt solutions, and then came close again to the A-line at high concentrations. It is to be noted that although the points on the chart come close to the A-line at high concentrations no further soil class transformations are observed (see Figure 7). For the

conditions investigated in this study, the CH class clay was transformed to MH (i.e. high plasticity silt) class soils. The changes in soil classes are attributed to the flocculation of clays permeated with the salt solutions.
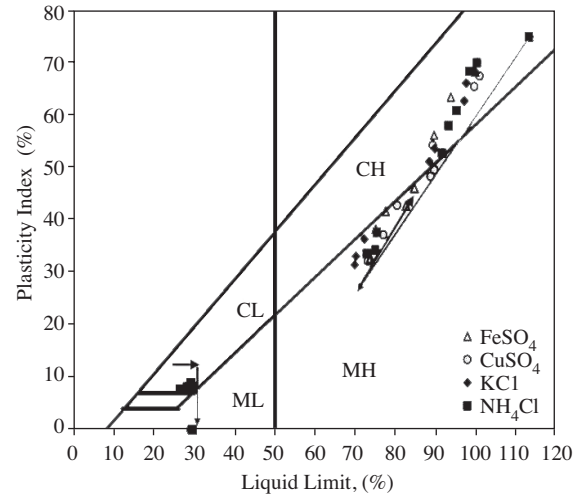

Figure 7. Change in soil class after the tests.

Conclusions

A study was undertaken to investigate the effect of 4 different salt solutions (i.e. ammonium chloride (NH_4Cl), potassium chloride (KCl), copper(II) sulfate ($CuSO_4$), and iron(II) sulfate ($FeSO_4$)) as leachate compounds on the consistency limits of clays. Consistency limit tests were conducted on low plasticity (CL class) and high plasticity (CH class) commercial clays. Tests were performed using both distilled water and these salt solutions at 8 different concentrations varying between 0.0001 and 1 M.

The following conclusions are made based on the test results and on the discussion presented in this study:

1. For CL clay, the liquid and plastic limits increased with increases in the salt concentrations up to around 0.2 M. The salt solutions at a concentration greater than 0.2 M seemed to damage the clay fabric; thus, the clay behaved as a nonplastic soil and the plastic limit could not be obtained.

2. All of the salt solutions with a concentration up to approximately 0.2 M significantly reduced the liquid limit of CH class clay. For NH_4Cl and KCl concentrations greater than around 0.2 M, the liquid limit of CH clay remained

more or less constant. However, for FeSO$_4$ and CuSO$_4$ metallic salts, the liquid limit of CH clay tended to increase at concentration values higher than approximately 0.2 M.

3. For CH clay, the plastic limit decreased at low salt concentrations (i.e. between 0.0001 and 0.001 M), and then increased with increases in the salt concentrations between 0.001 and 0.2 M. For concentration values higher than approximately 0.2 M, the plastic limit was not significantly affected by the salt solutions, and plastic limit values approached those of the raw clay sample.

4. Both CL and CH clays flocculated and formed clusters as the salt concentration increased, and the clay class tended to be changed. For the conditions investigated, CL and CH class clays were transformed into ML and MH class soils, respectively, according to the Unified Soil Classification System (USCS).

It should also be pointed out that further studies on the geotechnical properties of CL and CH clays interacted with chemicals (e.g., XRD, XRF, DTA, TG for mineralogy and cation exchange capacity, zeta potential for electrokinetic properties) are needed to make more reasonable judgments. Furthermore, in these studies, the behavior of the low plasticity clays (i.e. CL clay) may be explained more reasonably in terms of dispersion and flocculation mechanism.

Acknowledgments

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