

Effect of inorganic salt solutions on consistency limits of kaolinite

Gamze VARANK¹, Ahmet DEMİR¹, Senem YAZICI GÜVENÇ^{1,*},
Mehmet Şükrü ÖZÇOBAN²

¹Department of Environmental Engineering, Faculty of Civil Engineering, Yildiz Technical University, 34220 Davutpasa, Esenler, Istanbul, Turkey

²Department of Civil Engineering, Faculty of Civil Engineering, Yildiz Technical University, 34220 Davutpasa, Esenler, Istanbul, Turkey

Geliş Tarihi (Received Date): 18.07.2017
Kabul Tarihi (Accepted Date): 15.02.2018

Abstract

This study presents the effect of different inorganic salt solutions (KCl, BaCl₂, MgCl₂, KNO₃, Na₂SO₄ and MgSO₄) at different concentrations on geotechnical properties (Atterberg Limits) of kaolinite material which can be used as impermeable bottom liner in barrier systems. Since the use of distilled water or tap water is far from being representative of the in-situ conditions in landfills, salt solutions were used to investigate the leachate effect on liner materials. Additionally, the mineralogical characterization of kaolinite was studied. Atterberg limits, specifically the liquid limit (LL) and plastic limit (PL) that were used for classifying the clayey soil samples according to the Unified Soil Classification System were determined whereas mineralogical studies performed included XRD, BET and FT-IR analyses. Results indicated that all salt solutions have a considerable effect on the consistency limits of kaolinite. The liquid limit values of kaolinite decreased with increasing chemical concentration whereas plastic limit values increased. It is observed that the effects of the divalent and trivalent cations on kaolinite were more apparent than those of monovalent cations. As a result chemical solutions decrease liquid limit values of high plasticity kaolinite materials, tend to reduce the thickness of the DDL and flocculate the kaolinite particles, resulting in reduction of swelling and increasing of hydraulic conductivity.

Keywords: Inorganic salts, consistency limits, kaolinite, landfilling, barrier systems.

Gamze VARANK, gvarank@yildiz.edu.tr, <https://orcid.org/0000-0003-3437-4505>

Ahmet DEMİR, ahmetd@yildiz.edu.tr, <https://orcid.org/0000-0003-4649-3368>

* Senem YAZICI GÜVENÇ, senem.yazici87@gmail.com, <https://orcid.org/0000-0002-2877-0977>

Mehmet Şükrü ÖZÇOBAN, ozcoban@yildiz.edu.tr, <https://orcid.org/0000-0003-3521-0633>

Kaolinitin kıvam limitleri üzerine inorganik tuz çözeltilerinin etkileri

Özet

Bu çalışma, farklı konsantrasyonlardaki inorganik tuzların (KCl, BaCl₂, MgCl₂, KNO₃, Na₂SO₄ ve MgSO₄) bariyer sistemlerde geçirimsizlik alt tabaka dolgu malzemesi olarak kullanılan kaolinitin geoteknik özellikleri (Atterberg Limitleri) üzerine etkileri ortaya koymaktadır. Distile su veya çeşme suyu kullanımı, depo sahası koşullarını temsil edici olmadığından, dolgu malzemeleri üzerine sızıntı suyu etkilerini araştırmak için tuz çözeltileri kullanılmıştır. Ayrıca, kaolinit malzemesinin minerolojik karakterizasyonu incelenmiştir. Killi zemin numunelerinin, Birleşik Zemin Sınıflandırma Sistemi'ne göre Atterberg Limitleri; likit limit (LL) ve plastik limit (PL) değerleri belirlenmiş ve minerolojik çalışmalar için XRD, BET ve FT-IR analizleri gerçekleştirilmiştir. Elde edilen sonuçlar, tüm tuz çözeltilerinin kaolinitin kıvam limitleri üzerinde önemli bir etki olduğunu göstermiştir. Kimyasal konsantrasyon artışı, kaolinitin likit limit değerlerini düşürürken plastik limit değerlerini arttırmıştır. 2 ve 3 değerlikli katyonların kaolinit üzerine etkileri, tek değerlikli katyonlara göre açıkça görülmüştür. Sonuç olarak, kaolinit malzemesi hidrolik iletkenliği artar ve şişme eğilimi azalırken, DDL tabakası incelleme ve kaolinit partikülleri floküle olma eğiliminde iken, kimyasal çözeltiler yüksek plastisiteye sahip kaolinit malzemesinin likit limit değerlerini düşürmektedir.

Anahtar kelimeler: *İnorganik tuzlar, kıvam limitleri, kaolinit, düzenli depolama, bariyer sistemler.*

1. Introduction

The sanitary landfilling continues to be widely accepted and used method for the ultimate disposal of solid wastes due to its economic advantages. Besides its economic advantages, landfilling minimizes environmental insults and other inconveniences, and allows waste to decompose under controlled conditions until its eventual transformation into relatively inert, stabilized material. However, the release from a sanitary landfill consist mainly of leachate which has become the subject of recent interest as a strongly polluted and complex wastewater. Leachates are defined as the aqueous effluent generated as a consequence of rainwater percolation through wastes, biochemical processes in waste's cells and the inherent water content of wastes themselves. Leachates may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents consist an important group, as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts [1]. Contamination of groundwater by landfill leachate represents the major environmental concern associated with the landfilling of waste [2].

The liner system is the most important elements of a modern engineered landfill avoiding groundwater contamination. Current landfill liner systems are designed primarily as low permeability structures and commonly incorporate compacted clays or other mineral layers with synthetic membranes in composite or multi-barrier

arrangements [3-5]. Due to its high strength, impermeability, and resistance to chemicals, the high density polyethylene (HDPE) geomembranes are the most widely used components of a modern liner system in solid waste landfills. The other significant component of a modern liner system is soil liner generally comprising clay material. Governments apply enhanced regulation for liner systems. The standard liner recommended by The Ministry of Environment and Urbanisation of Turkey (Regulations of Municipal Solid waste Landfilling) consists of 100 cm of compacted clay liner (CCL) having a hydraulic conductivity $< 1 \times 10^{-9}$ m/s overlain with 2 mm HDPE GM .

Clay liners are commonly used to limit or eliminate the movement of leachate in the landfill site. The liners are exposed there to various chemical, biological and physical events, and they are affected by the resulting leachate. To assess the durability of the liner material, it is important to study the chemical compatibility of the liner material with different pore fluids, or the leachate that the liner may be subject to. When attempting to define the geotechnical characteristics of clay liners, the use of chemicals can be representative in-situ conditions.

In recent years, many researches have been focused on the interaction of clay soils with different types of fluids to determine the effect of landfill leachate on geotechnical properties of liner materials especially on hydraulic conductivity [6-11]. Hydraulic conductivity is the key parameter for barrier soils in contaminant migration from leachate to groundwater. Since the determination of hydraulic conductivity values of soils takes a long time, the effect of fluid on hydraulic conductivity of the barrier soil can be estimated by the consistency limit tests. The consistency limits have been representative indicators of clay behavior. Basically, the liquid limit, plastic limit and the plasticity index parameters are highly influenced by interaction with liquids [6], and the hydraulic conductivity tends to decrease when the liquid limit and the plasticity index are increased [11-13].

In this study, effect of different concentrations (0.01, 0.1 and 1 N) of inorganic salt solutions (KCl, BaCl₂, MgCl₂, KNO₃, Na₂SO₄ and MgSO₄) on the consistency limits of kaolinite was investigated. Mineralogical properties and soil classification of kaolinite were also studied.

2. Material-method

Liner materials were classified according to Unified Soil Classification System (USCS) (ASTM D 2487) and index parameters of the liner materials were determined by Atterberg tests (ASTM 4318) [14]. Solutions of used in the experimental study were prepared by dissolving salt of KCl, BaCl₂, MgCl₂, KNO₃, Na₂SO₄ and MgSO₄ (powdered with purity grade of (95%) in DI water. Each solution was mixed in 1 L flask.

The mesopore and micropore size distributions were estimated based on the Barrett–Joyner–Halenda (BJH) and Horwath–Kawazoe (HK) theory, respectively. The Brunauer–Emmet–Teller (BET) surface areas and BJH pore distributions were determined using the Quantachrome Ins. Quadrasorb SI model instrument by the nitrogen adsorption at 77 K. The specific surface area was calculated based on the BET

equation. The micropore surface area and volume were calculated by the t-method. X-ray diffraction (XRD) measurements were performed on a X-ray diffractometer using $\text{CuK}\alpha$ radiation. Qualitative estimation of the surface functional groups was performed by the Fourier transform infra-red spectroscopy (Perkin Elmer Spectrum 100 Model) by the potassium bromide (KBr) pellet method.

3. Results and discussion

3.1. Mineralogical properties of kaolinite material

Kaolinite material used in this study can be classified as CH-Class (LL>50) high plasticity clay according to the Soil Classification System (USCS). To determine the particle size distribution of the kaolinite, tests including sample preparation ASTM D421 (1985)[15], sieve analysis for coarse particles ASTM D422 (1963) [16], and hydrometer tests for fine particles ASTM D422 (1963) [16] were implemented. According to hydrometer test it was determined that kaolinite had high clay content. Clay and silt content of kaolinite was found to be 85% and 15% respectively. High LL value can be attributed to high clay content of the kaolinite resulting high swelling property.

XRD and FT-IR analysis graphs are given in Figure 1 and Figure 2, respectively. XRD analysis gives mineralogical composition of the material. Kaolinite mineralogically contains quartz (SiO_2), kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) and alunite ($\text{KAl}_3(\text{SO}_4)_2\cdot\text{OH}_6$). Quartz represents the purity of the material.

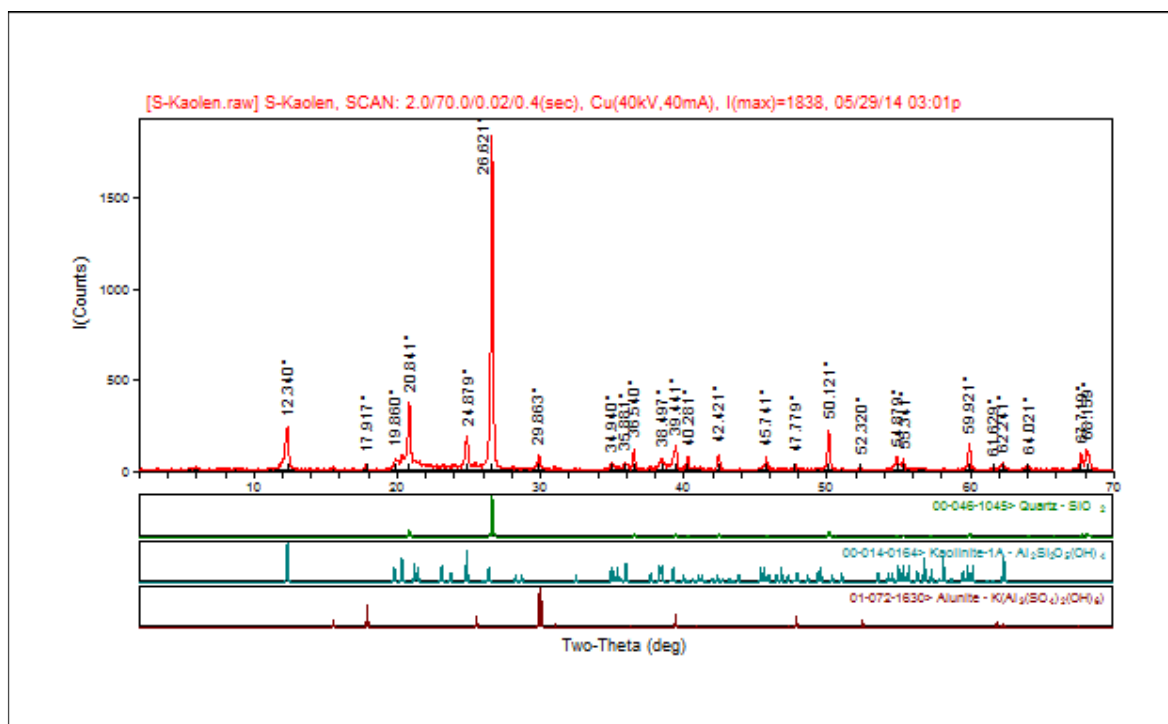


Figure 1. XRD graph of kaolinite.

BET analysis gives surface area of mesopores, macropores and micropores, volume of macropores and micropores and diameter of macropores and micropores of the kaolinite. Total surface area, macropore surface area and micropore surface area of kaolinite was determined to be 27.315, 22.049 and 5.266 (m^2/gr), respectively. Macropores volume and micropores volume were found to be 0.120 and 0.006 cc/gr

whereas macropore diameter and micropore diameter were determined to be 19.483 Å and 8.800e+ 00 Å, respectively. It was found that the kaolinite had a remarkable BET surface area, which was primarily contributed by mesopores and macropores. It can also be concluded that kaolinite has a macroporous structure.

Although the information obtained from FT-IR scanning was limited as the concentrations of the functional groups on the sample surface were in fact very low, the absorption spectra provide the evidence of the presence of some surface functional groups. In the FT-IR spectrum of kaolinite peak at 691 and 1026 cm^{-1} indicates Si-O structures on the surface of kaolinite. The peak observed at 795 cm^{-1} can be assigned to Si-O-Si structures associated with pronounced concentration of silicon in the sample corresponds to cellulose on the surface. The peaks at 3691, 3650 and 3620 cm^{-1} could be assigned to hydroxyl groups probably attributed to adsorbed water. The band centered at 911 cm^{-1} in the FT-IR spectra of sewage sludge was assigned to Al-O-H structures in the region and the peak observed at 1003 cm^{-1} indicates Si-H structure.

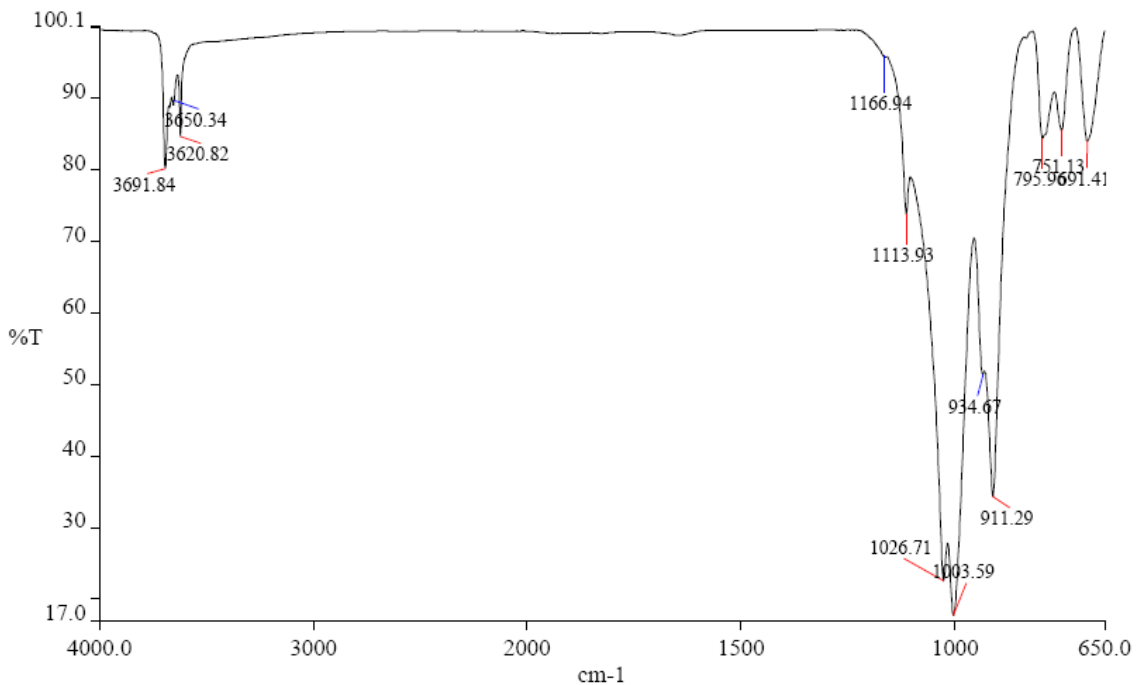


Figure 2. FT-IR graph of kaolinite.

3.2. Effect of inorganic salt solutions on consistency limits of kaolinite material

It is reported that the geotechnical properties (especially, hydraulic conductivity) of clay were significantly changed due to dispersion and deflocculation of clay as the clay interacted with chemicals [17, 18]. Salt solutions might also cause to form new swelling compounds that increases the liquid limit of CL clay as well as dispersion of the clay particles [19].

Contrasts to the findings of CL (low plasticity) clays, some researchers have indicated that the liquid limit decreased with increasing salt concentration for CH clays [6, 14, 18, 20-28]. It could be said that the salt solutions tended to reduce the thickness of the DDL and flocculate the CH clay particles, resulting in a reduction of liquid limit of CH clay. This behavior has been explained with the flocculation mechanism of kaolinite (nonswelling) clays and Diffuse Double Layer (DDL) theory by the authors [29].

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 interlayer zones of expandable clays, which subsequently became gritty or granular [21]. Also, Sharma and Lewis (1994) [30] 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.

Shackelford et al. (2000) [31], Jo et al. (2001) [32] and Kolstad et al. (2004) [33] reported that the effects of the divalent and trivalent cations on the clays were different from those of monovalent cations. Shackelford et al. (2000) [31] 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) [34] reported that the divalent cations were more effective than the monovalent cations. Hamutcu et al. (2008) [35] indicated that the variation in liquid limit with increasing concentration is insignificant for CL clays. However, Kurt et al. (2007) [36] indicated that the liquid limit of CH clay decreased with increasing concentrations of NaCl and KCl salt solutions. Additionally, It could be said that the temperature and salt solutions tended to reduce the thickness of the Diffuse Double Layer (DDL) and flocculate the CH clay particles, resulting in a reduction in liquid limit of CH clay.

Liquid limit values of kaolinite decreases as the concentration of the inorganic salt solutions increases (Figure 3 and Figure 4). The decrease in liquid limit values were found to be higher as the valence of the cation increases. When permeated with distilled water, liquid limit, plastic limit and plasticity index values of the kaolinite were determined to be 71, 24 and 47, respectively. As the concentrations of salt solutions increase in the range of 0.01-1 N, liquid limit values of kaolinite decreased from 71 to 65 as permeated with KCl, from 71 to 59 as permeated with BaCl₂, from 71 to 57 as permeated with MgCl₂. It can be seen from Figure 3 and 4 that divalent cations has more significant effect on liquid limit values of kaolinite. Similarly, liquid limit values of kaolinite decreased from 71 to 59 as permeated with Na₂SO₄ and from 71 to 52 as permeated with MgSO₄. KNO₃ solution decreased liquid limit values of kaolinite from 71 to 68. Results obtained are found to be consistent with the studies performed by [6, 14, 18, 20-28]. It can be seen from Figure 3 and Figure 4 that plastic limit values of kaolinite increases and plasticity index values decreases as ion concentration increases. Additionally it is observed that the increase in plastic limit values were found to be higher for divalent cations.

Consequently, it could be said that, decrease in liquid limit values causes increase in hydraulic conductivity when the concentration of the salt solutions was increased. This can be attributed to the decrease in the thickness of DDL, resulting in flocculation of the clay particles. The thickness of the DDLs is an important controlling factor for the structural development, hydraulic conductivity, and other physico-chemical and mechanical properties of soils [18]. As indicated by Gouy-Chapman theory, the

thickness of the DDL decreases result in larger pore channels through which flow can occur [25, 37, 38].

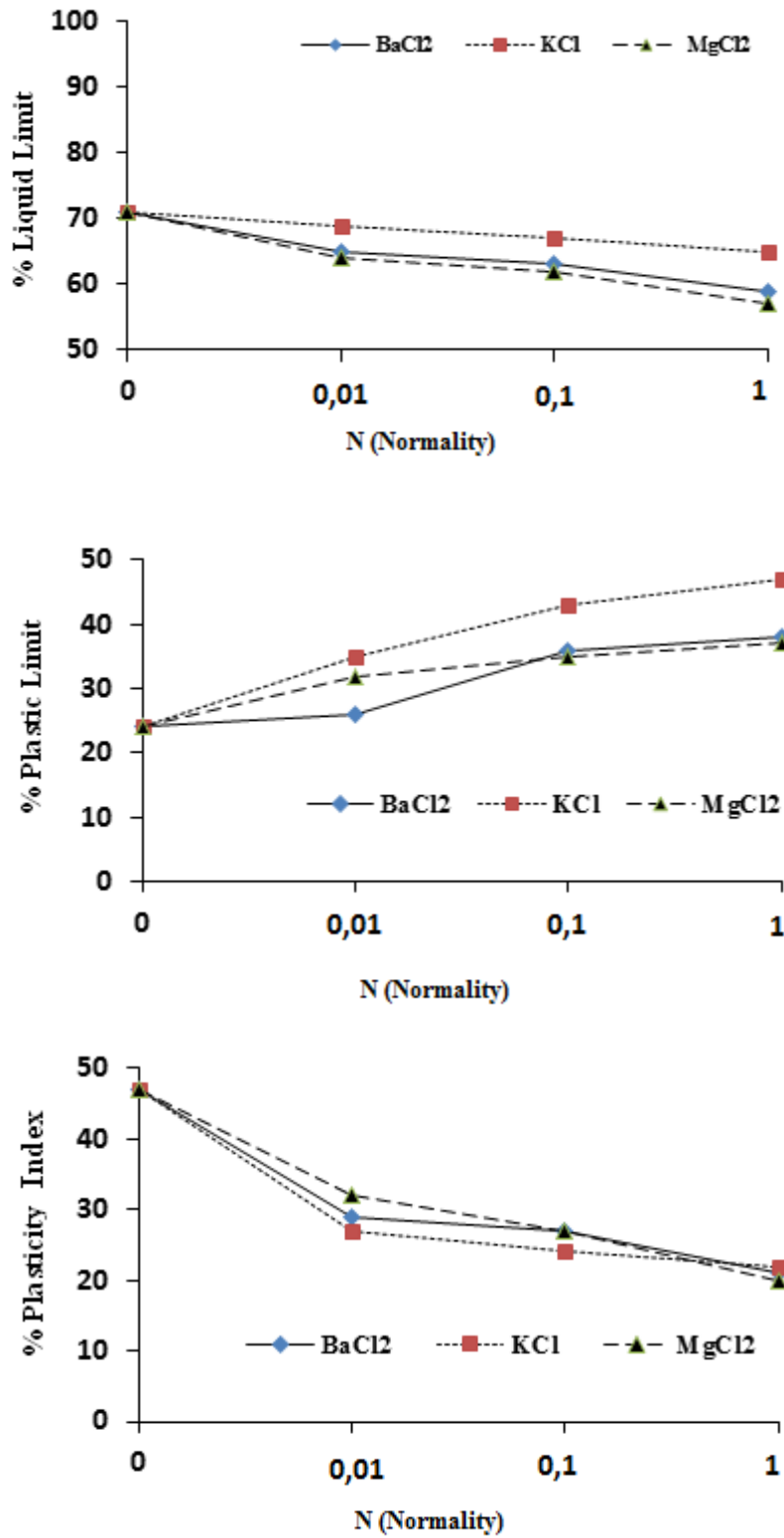


Figure 3. Effect of solutions of KCl, BaCl₂, MgCl₂ on consistency limits of kaolinite.

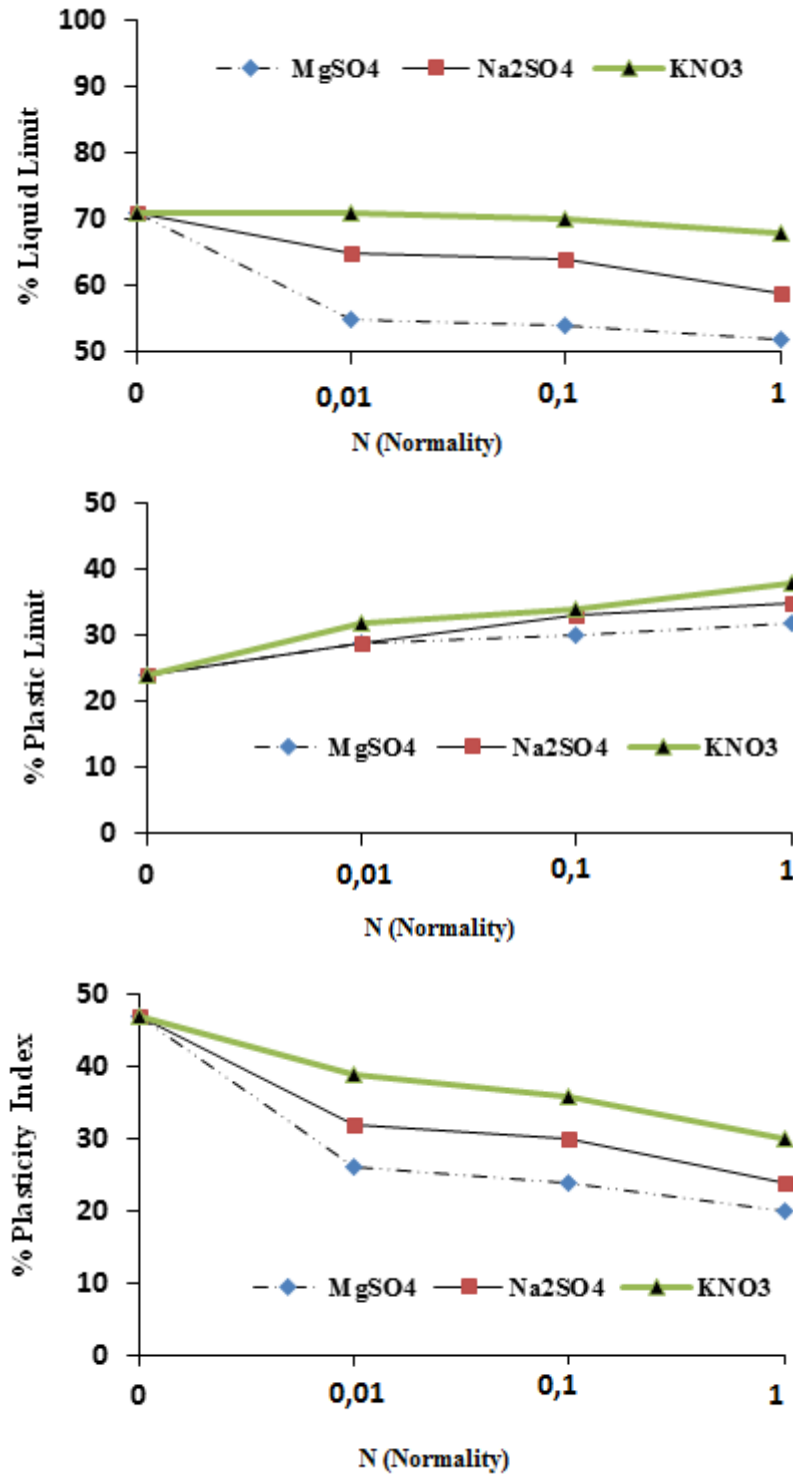


Figure 4. Effect of solutions of KNO₃, Na₂SO₄ and MgSO₄ on consistency limits of kaolinite.

4. Conclusion

In this study, the effect salt solutions (KCl, BaCl₂, MgCl₂, KNO₃, Na₂SO₄ and MgSO₄) on consistency limits of kaolinite was investigated. Experimental study was performed by distilled water and salt solutions at 3 different concentrations in the range of 0.01-1

N. Additionally, mineralogical characterization of kaolinite was determined by XRD, BET and FT-IR analysis. Results indicated that Kaolinite material used in this study can be classified as CH-Class (LL>50), high plasticity clay according to the Soil Classification System (USCS). Kaolinite mineralogically contains quartz (SiO_2), kaolinite ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) and alunite ($\text{KAl}_3(\text{SO}_4)\cdot\text{OH}_6$) and macroporous surface area. The chemical solutions tended to reduce the thickness of the DDL and flocculate the clay particles, resulting in reduction of liquid limit. It is determined that divalent and trivalent cations had shown more significant effect than monovalent cations. It can be estimated that leachate has negative effect on geotechnical properties of CH class liner materials by decreasing the DDL thickness and increasing the hydraulic conductivity.

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