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# The Effect of Long-Term Permeation of Inorganic Salts on the Geotechnical Characteristics of Bentonite-Amended Clay Liners

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## ABSTRACT

Bentonite-amended clay liners in engineered landfills are designed to prevent groundwater contamination from landfill leachate. Although numerous studies have examined the short-term effect of inorganic salts present in the landfill leachate on geotechnical properties of the bentonite-amended clay liners, studies on their long-term effects remain limited. Therefore, this study evaluated the long-term effects of $CaCl_2$ and NaCl, salts commonly found in landfill leachate, on the plasticity, swelling, compressibility, and hydraulic conductivity of clayey soil amended with 10% and 20% bentonite. These samples were immersed in 1M solutions of $CaCl_2$ and NaCl for 180 days to investigate their long-term effects. In contrast to the results obtained when water was the pore fluid, the liquid limit, plastic limit, free swell index, and free swell of bentonite-amended soils decreased in the presence of $CaCl_2$, $MgCl_2$, and NaCl solutions. Samples immersed in salt solutions showed significantly lower liquid and plastic limits than fresh samples prepared with the same salt solutions. The hydraulic conductivity of all samples exposed to $CaCl_2$ and NaCl decreased with effective stress and reached values less than $1 \times 10^{-9}$ m.s$^{-1}$. The addition of more bentonite did not significantly improve hydraulic conductivity when exposed to different salt solutions in the long term. In conclusion, samples with higher bentonite content are more susceptible to attack by inorganic salts over the long term.

## INTRODUCTION

In many developing countries, waste management has not kept pace with advancements in other areas. As cities grow quickly, often without proper planning, and as consumption and industrial activity rise, existing waste systems become increasingly overwhelmed (Sanoop et al. 2024). Compacted clay soils are commonly used in engineered landfill liners due to their low hydraulic conductivity and high adsorption capacity, which help prevent subsurface contamination from Landfill leachate (Yong 2019, Widomski et al. 2018, Sobti & Singh 2017). Landfill leachates, which contain both dissolved and suspended materials, can pose environmental risks by carrying toxic substances that contaminate surface water and groundwater (Tenodi et al. 2020, Essienubong et al. 2018). Clay soil liners' hydraulic conductivity is typically determined by the size and tortuosity of the pathways through which the free water flows, as well as the percentage of water that is hydraulically mobile. Because a large amount of pore water is 'bound' to the clay surface, which means it stays immobile under a hydraulic gradient, and the free water moves along constricted, winding channels, bentonite in particular has low hydraulic conductivity (Mesri 1971). In the interlayer gaps of montmorillonite particles, bound water molecules build up during hydration and are held tightly in place by electrical forces (Sivapullaiah et al. 1996). Changes in the amount of bound water in the pore space directly impact the amount of free water and the characteristics of the flow paths. Further, due to the high swelling capacity of bentonite, upon absorption of water, bentonite particles swell inside the liner, reducing the porosity of the liner. As a result, bentonite-amended clay liners generally exhibit lower hydraulic conductivity than unamended clay liners (Benson et al. 2018). However, bentonite-amended compacted clays may face challenges related to shrinkage or desiccation cracking, particularly when a significant amount of bentonite is used, which can increase hydraulic conductivity (Dutta & Mishra 2015). Bentonite swells in two stages. The first stage, called interlayer or crystalline swelling, occurs when water is adsorbed in monolayers on the surface of clay particles, forcing the layers of montmorillonite units apart. The repulsion between the diffuse double layers of bentonite particles causes the second stage of swelling. Leachate chemicals with greater cation valence, higher concentration, and lower dielectric constant are more likely to cause the diffuse double layer to shrink. These modifications may impact the compacted clay liner's strength, compressibility, hydraulic conductivity, and plasticity (Rout & Singh 2020). Although numerous studies have examined the short-term effect of inorganic salts present in the landfill leachate on geotechnical properties of the bentonite amended clay liners, studies on their long-term effects remain limited. Therefore, it is crucial to study the geotechnical properties of bentonite-amended compacted clay liners when exposed to the inorganic compounds present in landfill leachate in the long term. The objective of this study is to evaluate how prolonged exposure (180 days) to different inorganic salt solutions affects the plasticity, swelling behavior, and hydraulic performance of bentonite-amended clay liners.

## MATERIALS AND METHODS

### Materials

Several soil samples were collected from various areas in Sri Lanka, and based on the recommended Plasticity Index (10-30%) for the construction of clay liners, soils collected from Kegalle were selected. Commercially available Bentonite was purchased from Loyeds Hardware, Colombo. Selected soil was mixed with 10% and 20% bentonite to prepare bentonite amended soil samples. 1M solutions of $CaCl_2$, $MgCl_2$ and NaCl were used as permanent liquids representing inorganic compounds in landfill leachate, as inorganic compounds interact more with bentonite compared to other constituents in leachate.

### Methods

The summary of the experiment conducted is presented in Table 1.

Table 1: The summary of Experiments.

| Experiment | Sample | Test solution |
|---|---|---|
| Liquid Limit, Plastic Limit* | Bentonite<br>Soil<br>S+10%<br>S+20% | Water, 1M $CaCl_2$, 1M NaCl, 1M $MgCl_2$ |
| Free Swell Index | Bentonite | Water, 1M $CaCl_2$, 1M NaCl, 1M $MgCl_2$ |
| Non-load free swell test** | S+10%<br>S+20% | Water, 1M $CaCl_2$, 1M NaCl, 1M $MgCl_2$ |
| Consolidation and hydraulic conductivity tests** | S+10%<br>S+20% | 1M $CaCl_2$, 1M NaCl |

* For fresh samples and samples soaked in salt solution for 180 days
**For samples soaked in salt solution for 180 days

### Physical Properties of Clay and Bentonite

Atterberg limit tests were performed according to BS 1377 Part 2 for bentonite, unamended soil, and soil mixed with 10% (S+10%) and 20% (S+20%) bentonite.

### Free Swell Index

Free swell index (FSI) tests were carried out according to ASTM D 5890. 2 g of dried and finely ground bentonite/clay is dispersed into a 100 mL graduated cylinder in 0.1g increments. For the clay to fully hydrate and settle to the bottom of the cylinder, at least 10 minutes must elapse between additions. Until the full 2 g sample has been put into the cylinder, these procedures are followed. The level of the settled and swelled clay is then measured to the nearest 0.5 mL after the sample has been covered and shielded from disturbances for 16 to 24 h. The free swell index of bentonite and bentonite-amended clays was determined in water and 1 M solutions of $CaCl_2$, $MgCl_2$, and NaCl.

### Non-load Free Swell Test

To understand the swell behavior of bentonite upon exposure to different ion solutions, the oedometer non-load swelling test was conducted under the condition of lateral restraint and no vertical load (Xiao et al. 2021). Samples were prepared in a consolidation ring with an initial water content of 48% and a dry density of 1.1 $g.cm^{-3}$. The samples were fully submerged in the various ion solutions that were injected into the tank. For the first hour of the experiment, a 2-minute time interval was employed to create a non-load swelling curve. The break was then extended to thirty minutes. After 24 h, a 2-hour interval was employed. The experiment ought to be stopped if the deformation during six h is less than 0.01mm.

Table 2: Engineering properties of clay and bentonite.

| Property | Clay | Bentonite |
|---|---|---|
| Liquid limit [%] | 39.41 | 546.52 |
| Plastic limit [%] | 14.14 | 100.20 |
| Plasticity index [%] | 25.27 | 446.32 |
| Specific Gravity [G] | 2.382 | 1.456 |
| pH | 5 | 8 |
| Free Swell Index [$\text{mL.2g}^{-1}$] | 5 | 32 |

### Consolidation and Hydraulic Conductivity Tests

One-dimensional consolidation tests were conducted to determine compressibility characteristics and to calculate the hydraulic conductivity of different mixtures immersed in different salt solutions. To prepare the test samples, samples sieved through a 2 mm sieve and combined with the appropriate salt solution (1M NaCl or 1M $CaCl_2$) were placed and compacted in a metal ring of 60 mm diameter and 20 mm high in the remolded state. The sample was then loaded in increments of vertical stress selected as 25, 50, 100, 200, 400 and 800 kPa. The hydraulic conductivity was calculated from the following equation: $k = C_v \times m_v \times \gamma_w$, where $C_v$ is the coefficient of consolidation, $m_v$ is the coefficient of volume compressibility, and $\gamma_w$ is the unit weight of water. CV values were calculated using Taylor's square root of time fitting method.

## RESULTS AND DISCUSSION

### Physical Properties

The physical properties of clay and bentonite used for this study are given in Table 2.

### Plasticity Characteristics

The variation of liquid limit (LL), plastic limit (PL), and plasticity index (PI) of bentonite, soil mixed with 10% bentonite (S+10%), and soil mixed with 20 % bentonite (S+20%) under permeation of $CaCl_2$, $MgCl_2$, and NaCl are shown in Fig. 1. The liquid limit of bentonite under permeation of water is 546.52 %, but significantly decreased to 70%, 100%, and 79% under permeation of CaCl2, MgCl2, and NaCl, respectively. According to these results, CaCl2 significantly affects the bentonite's plasticity properties. Higher temperatures and cation valence in pore fluids cause the double layer's thickness to decrease and the repulsive forces that induce the clay particles to flocculate to decrease. (Hafiz et al. 2017).

The liquid limit, plastic limit, and plasticity index of S +10% and S+20% under permeation of water, $CaCl_2$, $MgCl_2$ and NaCl are presented in Fig. 2(b) and Fig. 2(c). In S+10%, the highest liquid limit of 74% was observed under permeation of water, and it was decreased to 46%,

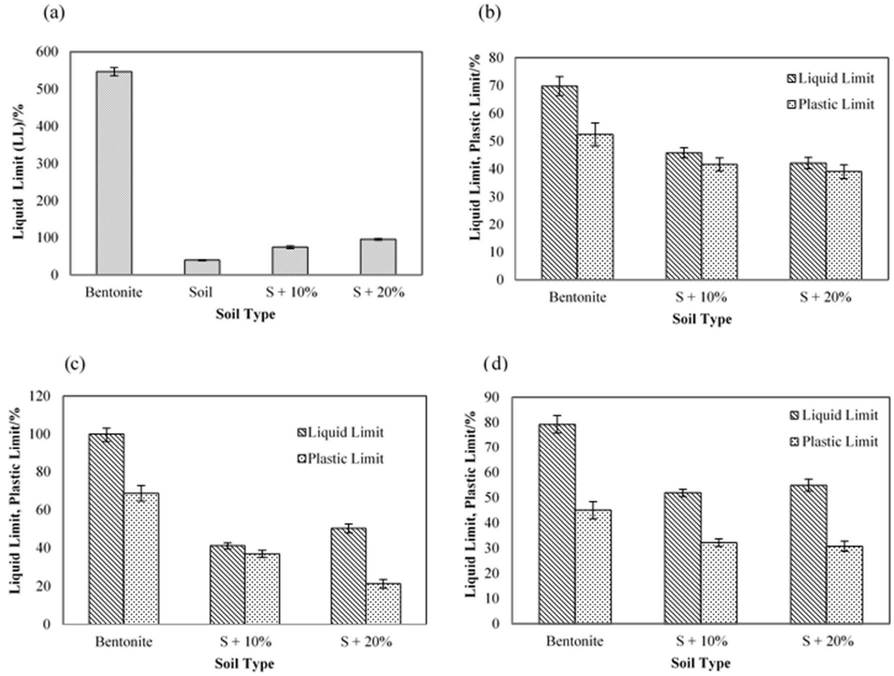

Fig. 1: (a) Liquid limit of Bentonite, Soil, S +10% and S+20% under permeation of water (b) Liquid limit, Plastic limit of Bentonite, Soil, S +10% and S+20% under permeation of $CaCl_2$ (c) Liquid limit, Plastic limit of Bentonite, Soil, S +10% and S+20% under permeation of $MgCl_2$ (d) Liquid limit, Plastic limit of Bentonite, Soil, S +10% and S+20% under permeation of NaCl.

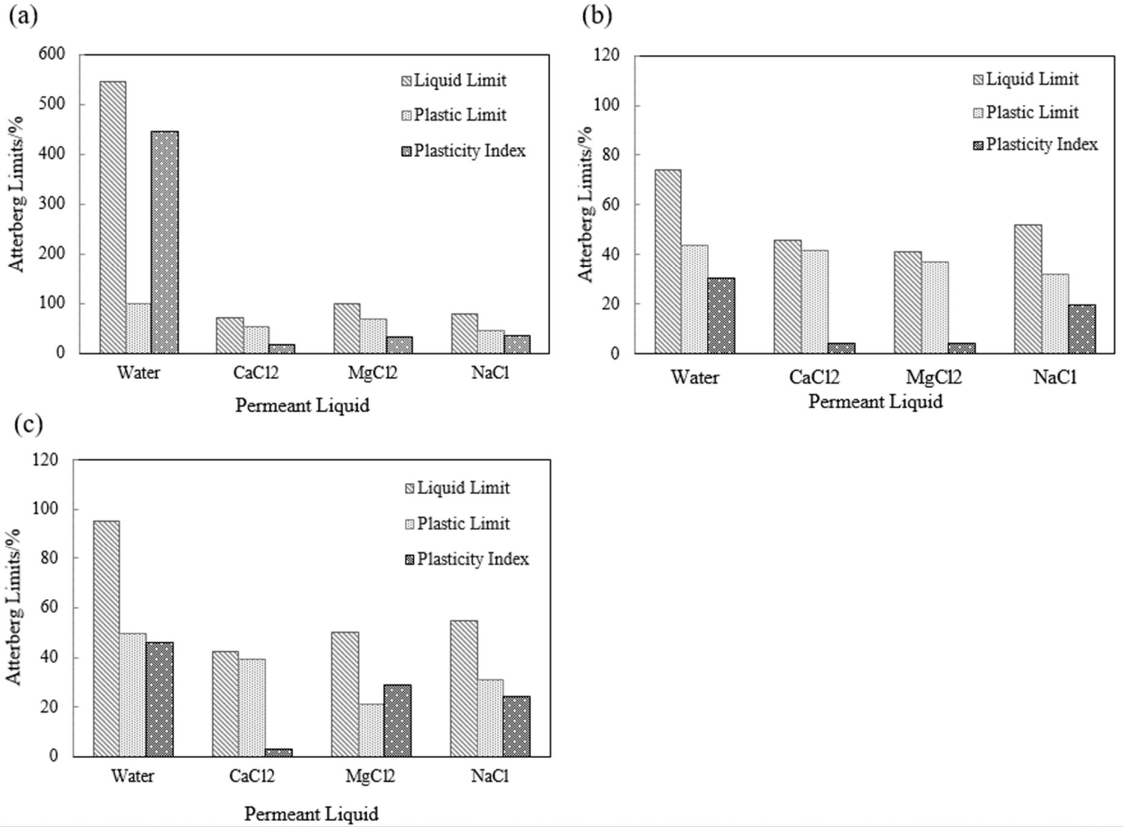

Fig. 2: (a) Variation of LL, PL, PI of bentonite with permeant liquid, (b) Variation of LL, PL, PI in S+10% with permeant liquid, (c) Variation of LL, PL, PI in S+20% with permeant liquid.

41%, and 52% under permeation of $CaCl_2$, $MgCl_2$, and NaCl, respectively. In S+20%, the highest liquid limit of 95% was observed under permeation of water, and it was decreased to 42%, 50%, and 55% under permeation of $CaCl_2$, $MgCl_2$, and NaCl, respectively. It has been shown that a higher lowering of the liquid limit happens when there is a high bentonite content because the cation valence of the permeating fluid rises. When compared to divalent cations, the impact of monovalent cations on the plasticity properties of both S+10% and S+20% mixtures was less pronounced. Because the double layer of existing fine fractions in the mixes is thinner, the repulsive forces between the clay particles are reduced, and the structure changes from dispersing to flocculating, which is why the plasticity characteristics are reduced with the cation valence (Nath et al. 2023).

The liquid limit and plastic limit values of S+10% and S+20% soaked in inorganic salt solutions for a period of 180 days are shown in Fig. 3. The liquid limit values of S+10% and S+20% immersed in $CaCl_2$ are 37.1% and 33%, respectively. A significant reduction of liquid limit was observed when comparing these values with freshly prepared S+10% and S+20% using $CaCl_2$. For S+10%, about 19% reduction in liquid limit was observed after immersing the samples in $CaCl_2$ for 180 days, while it was around 21% for S+20%. It should be noted that the liquid limit of soil without adding any bentonite was 39.4%, and the bentonite-amended samples exhibited an even lower liquid limit when immersed in $CaCl_2$. Because the diffuse double-layer thickness decreases when exposed to inorganic salts, bentonite is more prone to lose its ductility (Shariatmadari et al. 2017). So, when part of the soil in the clay liner is replaced with bentonite, due to the above phenomenon, lower liquid limit values compared to the unamended soil can be expected when the liner is subjected to long-term contact with inorganic salts. A similar reduction of liquid limit values was observed in samples soaked in NaCl for a long period. Based on these findings, it is unclear how cation valence affects the plasticity properties.

## Swelling Characteristics

Free swell index (FSI) test results of bentonite under water and 1M solutions of $CaCl_2$, $MgCl_2$ and NaCl are presented in Fig. 4.

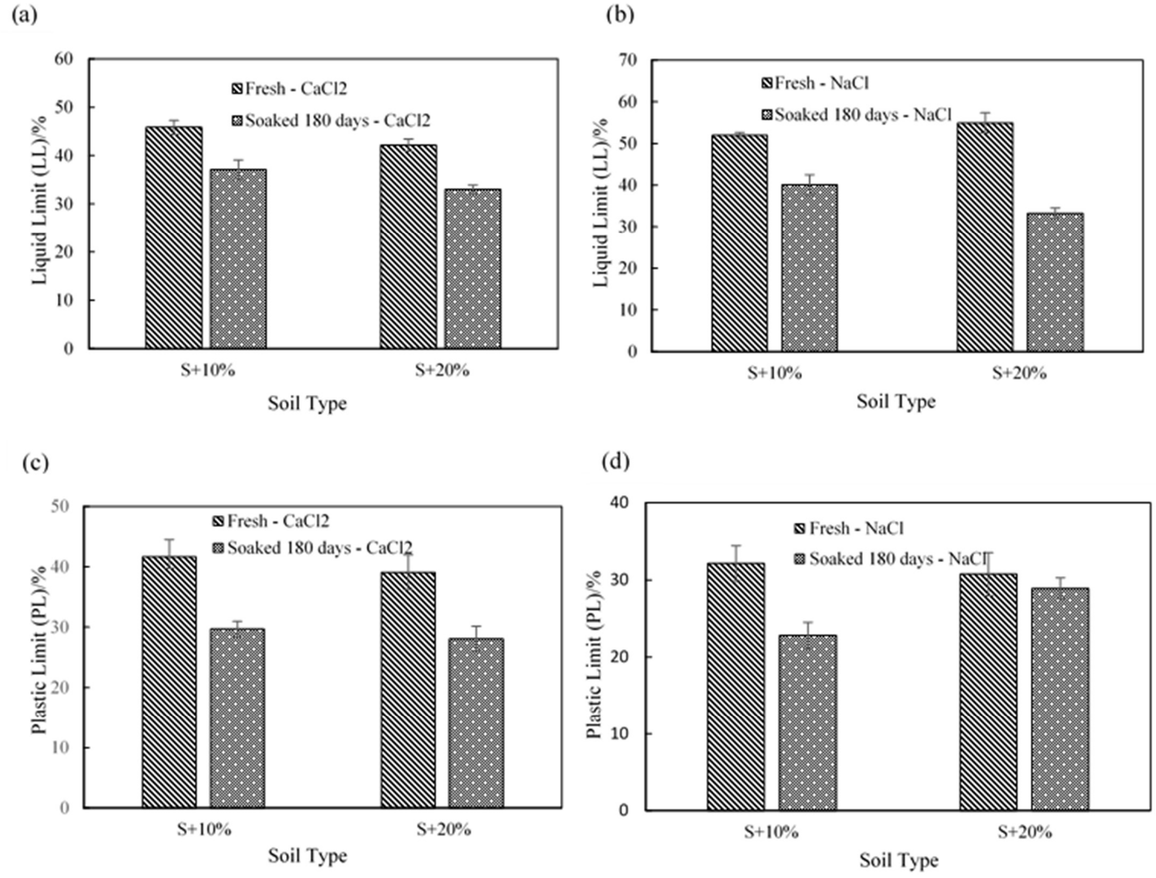

Fig. 3: Comparison of the liquid limit of S+10% and S+20% with (a) $CaCl_2$ as the pore liquid, (b) NaCl as the pore liquid, and the plastic limit with (c) $CaCl_2$ as the pore liquid, (d) NaCl as the pore liquid.

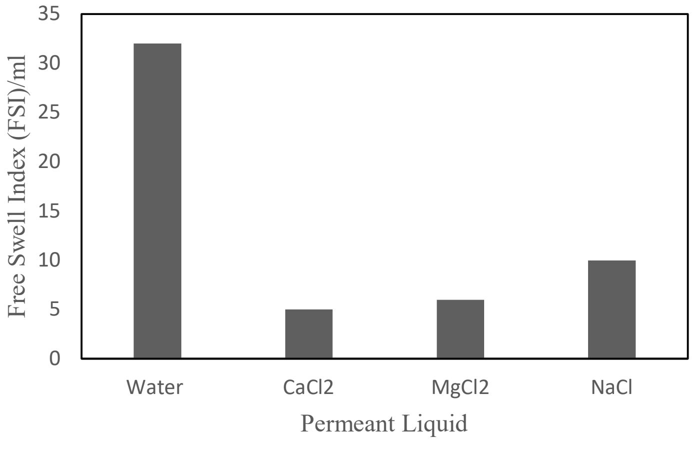

Fig. 4: Variation of free swell index of bentonite under water, 1M $CaCl_2$, $MgCl_2$, NaCl.

According to the results, the free swell index of bentonite under water as permeant liquid is 32 mL, and it reduced significantly to 5 mL, 6 mL and 10 mL when the permeant liquids are $CaCl_2$, $MgCl_2$ and NaCl, respectively. The results indicate that swelling characteristics are significantly affected by these inorganic compounds present in the leachate. At higher salt concentrations, the ions may compress the electrical double layer around the bentonite particles (known as "ion exchange"), further reducing the swelling potential (Ören & Akar 2017). The FSI can decrease significantly in this scenario because the clay particles become more tightly bound due to the increased ion concentration, thus preventing expansion.

The non-load swelling curves of S+10% with water and 1M solutions of $CaCl_2$, $MgCl_2$, and NaCl are shown in Fig.5. Research indicates that observed swelling occurs only after the voids, filled with non-swelling particles, are occupied by the swollen clay particles. The magnitude of swelling within the voids, referred to as intervoid swelling, is more significant when the size and percentage of the non-swelling coarse fraction are larger. The swelling observed after intervoid swelling is termed primary swelling, and it follows a rectangular hyperbolic relationship with time, accounting for about 80% of the total swelling. Secondary swelling, which continues over a long period, exhibits a straight-line relationship with logarithmic time. Primary swelling happens immediately in mixtures with smaller non-swelling fractions but takes longer for mixtures with larger non-swelling fractions (Abbey et al. 2020).

It is clear from comparing the swelling behaviour of S+10% under various pore fluids that these inorganic salt solutions considerably lower the bentonite-amended soil's swelling capability, which is consistent with the FSI findings. A substantial chemical consolidation of the bentonite after infiltration by various inorganic solutions is shown by the swelling strain of samples saturated with $CaCl_2$, $MgCl_2$, and NaCl solutions being less than that of the sample saturated with distilled water. Due to the significant drop in electrical repulsion forces that occurs when salt diffuses from the reservoir salt solution, the space between clay particles and the clay specimen's void ratio both decrease, resulting in chemical consolidation stresses. There are several ways to explain this phenomenon. The compacted bentonite expands as a result of matrix suction dissipating throughout the saturation phase. At the same time, the infiltrating salt solution weakens the electrostatic repulsion between montmorillonite crystals, resulting in a thinner dispersed double layer of bentonite. These physicochemical interactions, therefore, decrease the compacted bentonite's expansion potential.

It is commonly acknowledged that cation exchange plays a significant role in the clay-water interaction. The kind, valence, concentration, and size of the cations involved are the main factors influencing this process. The ability of a cation to replace another cation increases with its valence (Zeng et al. 2021). The replacement capacity for cations of the same valence rises with cation size. A typical order of cation exchange capacity is $Na^+ < K^+ < Mg^{2+} < Ca^{2+}$.

## Consolidation and Hydraulic Conductivity

The coefficient of consolidation ($C_v$) of S+10% and S+20% samples submerged in 1M $CaCl_2$ and 1M NaCl solutions is shown in Fig. 6. In general, both the S+10% and S+20% samples submerged in $CaCl_2$ solutions indicate an increasing trend of $C_v$ values with the increase of effective consolidation pressure. Those $C_v$ values of S+10% range from $4.5 \times 10^{-9}$ $m^2.s^{-1}$ to $1.2 \times 10^{-8}$ $m^2.s^{-1}$. For S+20%, those values increase from $3.3 \times 10^{-9}$ $m^2.s^{-1}$ to $1.2 \times 10^{-8}$ $m^2.s^{-1}$ with the increase of consolidation pressure. However, irrespective of the bentonite content, significant differences in their $C_v$ values were not observed.

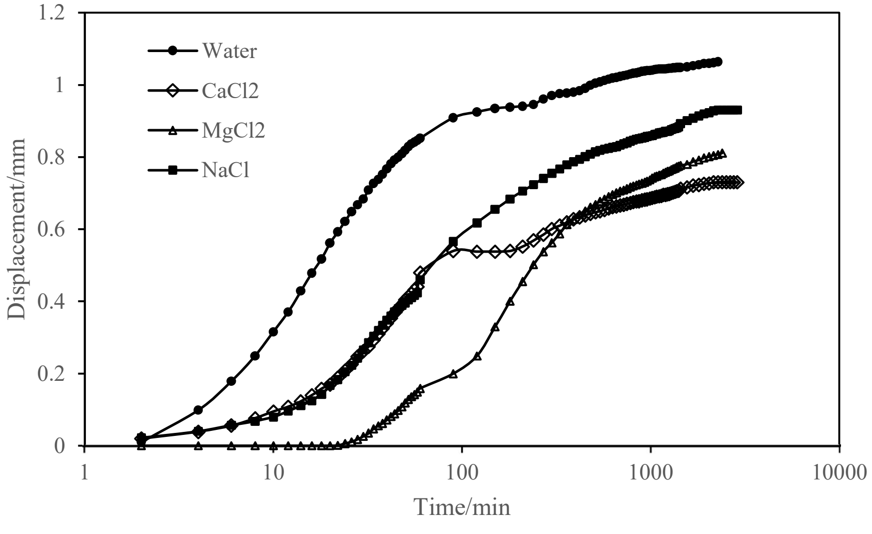

Fig. 5: Non-load free swell index results of S+10%.

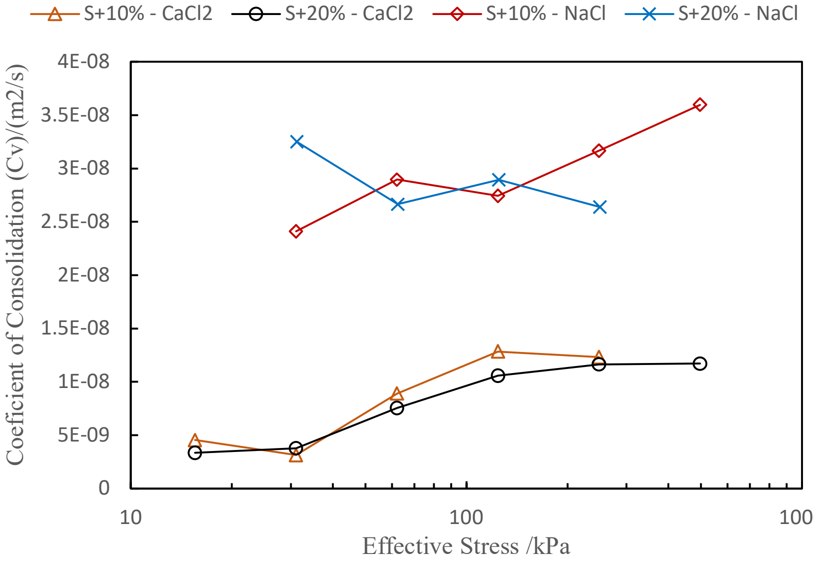

Fig. 6: Coefficient of consolidation versus effective consolidation pressure for clay-bentonite mixes submerged in 1M $CaCl_2$ and 1M NaCl salt solutions.

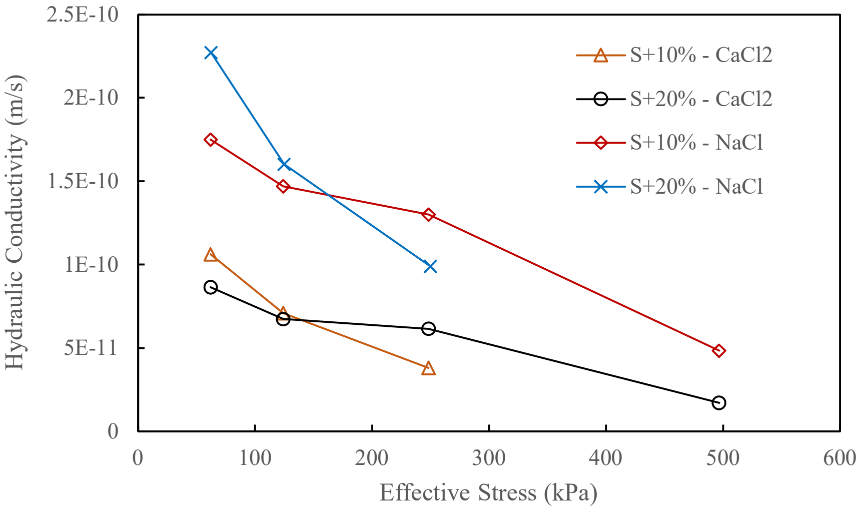

Fig. 7: Hydraulic conductivity versus effective consolidation pressure for sand-bentonite mixes submerged in 1M $CaCl_2$ and 1M NaCl salt solutions.

In S+10% and S+20% with 1M $CaCl_2$, after a steady increment in Cv, the settlement increased significantly from $1.17 \times 10^{-8}$ to $3.36 \times 10^{-8}$ $m^2.s^{-1}$. The Cv value for clay-bentonite mixes submerged in 1M NaCl containing 10% bentonite has decreased slightly in the initial consolidation process up to $2.7 \times 10^{-8}$. When increasing the applied stress up to 500 kPa, Cv has been increased gradually up to $3.7 \times 10^{-8}$ $m^2.s^{-1}$. The Cv value of the sample containing 20% bentonite, submerged in 1M NaCl, has decreased gradually up to 2.6 × 10-8 from 3.2 × 10-8 $m^2.s^{-1}$, when the applied stress is increased up to 1000kPa.

Except for S+20% with NaCl, all other samples indicate an increase in Cv with the consolidation pressure. Similar behaviour is reported for sand-bentonite mixtures, while the opposite trend is reported for pure bentonite (Francisca & Glatstein 2010). The mechanism governing the samples' compressibility behavior is responsible for these variations. While pure bentonite's compressibility behavior is determined by long-range attractive and repulsive forces produced by physicochemical factors, the compressibility of sand-bentonite mixtures is primarily controlled by mechanical factors that increase with increasing pressure (Sobti & Singh 2017, Petrov & Rowe 1997).

According to the 1-D odometer consolidation test results, it can be seen that the clay samples submerged in the $CaCl_2$ salt solutions show the lowest Cv values when compared to clay samples submerged in the NaCl salt solutions. According to these observations, samples submerged in the $CaCl_2$ have lower hydraulic conductivity than clay samples submerged in the NaCl solution.

As shown in Fig. 7, it can be clearly seen that hydraulic conductivity decreases with increasing applied stress. A drastic decrease in the hydraulic conductivity(k) was observed from $1.7 \times 10^{-10}$ to $4.8 \times 10^{-11}$ $m.s^{-1}$ for the sample submerged in 1M NaCl containing 10% bentonite-enhanced clay when the applied stress was increased up to 500kPa. For the sample submerged in the same salt solution containing 20% bentonite, as the consolidation process continued, it was found that the hydraulic conductivity values gradually decreased from $2.2 \times 10^{-10}$ to $9.9 \times 10^{-11}$ $m.s^{-1}$. A gradual decrease in hydraulic conductivity was observed for the sample submerged in 1M $CaCl_2$ containing 10% bentonite from $8.2 \times 10^{-11}$ to $3.4 \times 10^{-11}$ $m.s^{-1}$ when increasing the applied stress. The k value for sand-bentonite mixes submerged in 1M $CaCl_2$ containing 20% bentonite has significantly decreased from $8.6 \times 10^{-11}$ to $1.7 \times 10^{-11}$ $m.s^{-1}$ when continuing the consolidation process by increasing the applied stress up to 500 kPa.

The samples immersed in the $CaCl_2$ solution exhibit lower hydraulic conductivity than the samples immersed in the NaCl solution, according to an evaluation of the hydraulic conductivity values of the four bentonite-amended clay samples.

## CONCLUSIONS

The purpose of this investigation was to investigate how inorganic salts affected the swelling properties of clay liners that had been bentonite-amended. The inclusion of $CaCl_2$, $MgCl_2$, and NaCl salt solutions reduced the liquid limit and plastic limit of the bentonite-amended clay, according to the results. The decrease in the thickness of the diffuse double layer is the reason for the drop in these parameters when these salt solutions are present. The liquid limit and plastic limit of the samples soaked in $CaCl_2$ and NaCl for a period of 180 days are significantly lower than those of the fresh samples prepared using respective salt solutions. So, the inorganic salts present in the leachate significantly affect the plasticity of bentonite-amended clay liners. Furthermore, a comparison of the effects of different salts on plasticity characteristics indicates that bentonite-amended clay liners are more susceptible to attack by calcium ($Ca^{2+}$) ions present in leachate than by magnesium ($Mg^{2+}$) or sodium ($Na^+$) ions, resulting in a greater loss of plasticity. According to the 1-D odometer consolidation test results, the clay samples submerged in the $CaCl_2$ salt solutions showed the lowest coefficient of consolidation (Cv) values when compared to clay samples submerged in the NaCl salt solutions. Except for S+20% soaked in NaCl, other samples soaked in NaCl and $CaCl_2$ show an increasing trend of Cv values. To use as a bottom liner, compacted clay liners should exhibit an exceptionally low k value, a minimum of 1 x $10^{-9}$ $m.s^{-1}$. When considering the k values of the bentonite amended soil samples after submerging in salt solutions for 180 days, all the samples have reached k values lower than 1 x $10^{-9}$ $m.s^{-1}$. The evaluated k values of all the clay samples show that all the samples that were submerged in salt solutions for six months have maintained a hydraulic conductivity within the accepted range. However, the expected improvement of k with the addition of a higher amount of bentonite is not significant when submerged in different salt solutions. It can be concluded that samples with higher bentonite content are more vulnerable to attack by inorganic salts. Based on our results, liners containing lower amounts of bentonite (less than 10%) are recommended for landfill sites expected to generate leachates with high calcium ($Ca^{2+}$) concentrations.

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