Optimizing Stabilizers Enhanced Electrokinetic Environment to Improve Physicochemical Properties of Highly Organic Soil

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This paper present the findings of a study into physicochemical properties of highly organic soil mixing with varius binders. The study also investigate increasing unconfined compression strength achieved using founded stabilizers. The influence of concentration of stabilizers as well as dispersion of their pHs on physicochemical properties of organic soil were also investigated. The results proved that even a very low concentration of the reagents (0.001 mol/L) causes a remarkable change of the zeta potential (ζ) of organic colloids. The ζ of the pure organic soil varied from +25.1 to -95.4 mV at pH 2.8 and 11.3, respectively. However, according to the chemical reagents and electrolyte concentration, ζ varied from +189.22 at pH 4.43 for 0.1 M $CaCl_2$ to -88.2 at pH 9.69 in 0.001 M Na_2Co_3 solution. The pH of pure organic soils at iso-electric point (IEP) was 3.3. It is noteworthy that using reagents caused a significant shift in IEP to the alkaline part since pH at IEP for $Al_2(SO_4)_3$, H_3PO_4, Na_2Sio_2, and Na_2Co_3 increased as electrolyte concentration increased. Based on results of ζ, observed from using different cationic species, new binder systems were used. New binders showed a strength increasing to around 350% for soft organic soil. Those chemicals that caused thickening in diffuse double layer also used as secondary additives in order to find their effectiveness in soil strength properties. Optimizing stabilizers cause significant improvement as for electrokinetic (EK) injection method.

1. INTRODUCTION

The total tropical land which covered with highly organic matter soil in the world amounts to about 30 million hectares. In Malaysia approximately 3 million hectares of the country's land area are

covered with organic soil. These soils are extremely soft and from the geotechnical point of view known as problematic [1]. Stabilization of mechanical properties of highly organic soils is a key component in many new geo-construction methods due to their high compressibility and low hydraulic conductivity [2]. Although there are a huge number of stabilization methods, some are able to be applied for soils with low hydraulic conductivity. The main objective of such stabilization is to increasing shear strength, reducing compressibility as well as water content. Using different chemical species to modifying surface characteristics of colloidal particles has practical implications in environmental and industrial applications, Soil stabilization and decontamination processing being some of them. It is well understood, improving soil strength in large scale is happening based on interactions between destabilized colloidal particles through the soil structure [3-6]. The Stability of colloid particle into soil structure is determined by the charge that the colloid exerts on the diffuse layer as well as the distance that the ions of the diffuse layer are from the surface of the colloid. Stable colloids are much far away from each other than unstable colloids [6, 7]. Colloidal particles stability is affected by several factors; the surface area, the size, and the surface charge. Surface charge in turn is a function of pH and dissolved salts concentration [8] and is defined by ζ. ζ is known as the potential at the shear surface between the mobile part of the electrolyte surrounding a charged particle moving in an electric field and the stagnant part [9-11]. The value of ζ is less than the surface potential of particle and represents the value at the slip plane, which is located at a small unknown distance from the colloidal surface [12]. Based on above arguments, the ζ and the charge distribution in the fluid adjacent to the capillary wall play key roles in determining the electro-osmotic (EO) flow [13]. Several researchers have reported that the ζ of the soil particle correlates well with the EO flow [13-18]. ζ theory was developed by von Smoluchowski (1921) however has been later modified to fit more complex systems [19]. The value of ζ is depend on the concentration and type of ions in the medium, as well as the surface composition of the particle and varies with distance χ away from the surface of the particle [12]. Measuring ζ potential are mostly used for characterizing surface charge as well as adsorption onto, colloidal particles from solution as an small scale of solids [12, 20]. While, the composition and structure of soil humus is complex and incompletely known, it is well understood that clay and organic fractions strictly affect the electrical properties of organic soils [2, 5]. Hunter (1981) provided detailed description of experimental treatise to show the significance of pH on ζ of soils [12]. Vane and Zang (1997) showed that the pH and ζ vary depending upon the types of mineral for mineral soils [21]. In organic soil, ζ is dependent mainly on organic content, degree of humification, and mineral fraction [1]. Charge in highly organic soils is strongly pH dependent. Such negative charge of organic soil is believed to be due to the dissociation of H^+ from functional groups [2]. Also, colloidal samples from humified organic soils have a higher ζ potential with a negative mathematical sign and more charge than the undecomposed samples [5]. Decomposition involves the loss of organic matter either in gas or in solution forms, through the disappearance of physical structure and change in chemical state [22]. It is also clear that, the natural ζ ranges of the soils with high fibrous content are less than the natural ζ ranges of the soils with low fibrous content [5, 23-24]. The successful treatment of organic soils has been less documented and it is only including using cement materials whether combined with polymers or not [25]. Even if successfully treated, the stabilized highly organic soil is a new material that has not been investigated previously, thus little is known about the mechanisms

involved in its stabilization especially when cement did not used as main stabilizer. Many questions remain to be answered regarding the influence of reagents on ζ potential of humus and its mechanical behaviour in terms of shear strength, compressibility as well as EK properties. This study was conducted to (i) evaluate changes in physicochemical properties of highly organic soil according to using various designed stabilizers enhanced EO flow injection, (ii) interpret the findings with respect to implications for selecting the best binders for highly organic matter soil such as peat. EO can usually be applied to silts, clayey silts, and silty clays of soil texture [24]. EO treatment can cause various electro-chemical effects in soils: oxidation, electrolysis, ion diffusion, hydrolysis, formation of osmotic and pH gradients, desiccation by heat generation across the electrodes, precipitation of salts or secondary minerals, cation exchange, mineral decomposition, fabric changes, physical and chemical adsorption, and reduction [5, 9, 13, 24, 27, 28].

2. EXPERIMENTAL PROGRAM

The experimental program was included two phase. In phase I, fundamental ζ tests were performed on the organic soil mixed with several chemicals, $Al_2(SO_4)_3$, H_3PO_4, Na_2Sio_3, $CaCl_2$, $MgCl_2$, and Na_2Co_3, in various concentrations. Based on results achieved from Phase I, the organic soil was mixed with some new designed binders and cured for 14 days as for Phase II.

2.1. Organic soil

Organic soil was collected as hand specimen from a deposit in Mardi - Malaysia. It has a cation exchange capacity of only 54 meq. per 100 g, compared to electronegative minerals such as Kaolinite with 2-10 meq. per 100 g or peat between 60-120 meq. per 100 g, is considerable [2]. That means small amounts of Kaolinite particles or other minerals can act as stabilizing agent to organic soil suspension, by having a peptizing effect [28].

2.2. Purification of organic soil

Organic soil sample was treated before its use in the experiments to obtain homogenized organic colloidal particles. Also the suspension containing organic soil (0.1 g L^{-1}) was mechanically stirred for 24 h, and after waiting for 2 min the supernatant suspension was filtered through filter paper (Φ= 125 mm). Moreover, the solid sample was dried at 105°C for 24 h, ground, and then sieved by a 75μm sieve. The particles under 75 μm were used in further experiments.

2.3. Zeta Potential Measurements

The ζ of organic soil suspensions was measured using a Zeta Meter 3.0 (Zeta Meter Inc.). It was equipped with a microprocessor unit. The unit is able to calculates automatically the

electrophoretic mobility of the particles and also convert it to the ζ using the Smoluchowski's equation [29-30]. The Smoluchowski equation, for ζ gives a direct relation between electrophoretic mobility and ζ as showed in Eq. 1,

$$\text{Eq. 1,} \qquad \zeta = \frac{4\pi V_t}{D_t} \times EM,$$

Where D_t is dielectric constant, V_t is viscosity of the suspending liquid, EM, is electrophoretic mobility at actual temperature, π is a constant, and ζ is zeta potential [29]. The ζ measured as a function of adsorbed amount of cations on the surface of organic colloid and pH. After adsorption period, the ζ of particles which were obtained by centrifuging the suspension was measured. For the ζ measurements, a 50mg sample was transferred into aqueous solution and then soil particles mixed homogenously with a magnetic stirrer. All the measurements were conducted therefore at 100 mg/L of solid concentration. Also the voltage need to be applied is determined based on the specific conductivity of the solution. The particle movement is observed through a microscope and the time taken for a particle to travel a particular micrometer distance is measured. To minimize reading error, a minimum of 15 particles are tracked and their average time is calculated. The solution's pH was adjusted by a drop wise addition of NaOH and HCl solutions. The solution was stirred using a magnet stirrer until the pH of the solution became constant. The pH of the solution was measured before and after each measurement. If changes occurred in pH, the last one was recorded as the pH of the solution.

2.4. Preparation of the stabilized organic soil samples

The mixing procedure that used in this research project followed the EuroSoilStab project method (Design Guide: Soft Soil Stabilization) [26]. Such guideline, explain the different methods of stabilizing soft organic soils, the tests methods to determine the appropriate binder, the design approaches that are normally adopted, and the site equipment and installation procedure to be used. The soil was mixed initially in a laboratory dough-mixer for homogenization before mixing with binder in a dry state. Immediately after mixing, specimens for unconfined compression test were placed in plastic tubes 38 mm in diameter and 76 mm in height, kept immersed in water, and left to be cured under a pressure of 18 kPa, which represents normally about 1 m of fill laid out in the field.

2.5. Design of Binders

36 mixtures were made, mixing Na_2Sio_3, H_3PO_4, $CaCl_2$, and $Al_2(SO_4)_3$ in a various molarities (Table 1). Designing the mixtures was based on optimizing diffuse double layer thickness enhances EO flow injection and shear strength in treated soil as well. Binders are selected based on whether electro-ion migration (EM) or EO injection is required. EM is a crucial objective that can be achieved only when pH is at IEP. In this research Na_2Sio_3 and H_3PO_4 were selected as main stabilizer compounds while, $CaCl_2$ and $Al_2(SO_4)_3$ were used as secondary reagents to find suitable electro static

bonding between particles. Electrostatic phenomena arise from the forces that electric charges exert on each other [7]. The composition of the mixtures showed in Table 1.

Table 1. Chemical compound of the binders used

Chemicals	Used Synonyms	Molarity (mol/L)			
		Step-1	Step-2	Step-3	Step-4
Na_2SiO_3	N	0.1, 1, 3	0.1, 1, 3	0	0
H_3PO_4	H	0	0	0.1, 1, 3	0.1, 1, 3
$CaCl_2$	C	0, 0.1, 1	0, 0.1, 1	0, 0.1, 1	0, 0.1, 1
$Al_2(SO_4)_3$	A	0	0.1	0	0.1

3. RESULTS AND DISCUSSION

Physico-chemical properties of organic soil used in present research showed in Table 2. It is well understood, organic soil environments are suitable candidates for the EK phenomena due to presence of hydrated ions and a very wet soil mass [5 ,15]. Also, the organic soil sample used in this study showed very high specific surface area, WC and CEC (at the soil pH) (Table 2). Such properties increase with an increase in organic content [1, 5]. In EK remediation, it is important to know the exact CEC and ζ of the soil to estimate the presence of water sorption potential, water momentum, and positive charges on the soil surface [16].

Table 2. Physicochemical properties of the Organic soil

Parameter	
Color	Brownish
Moisture content,%	158
Particle density, Mg/m³	1.18
Von Post classification	H_4
Liquid limit, %	155.2
Soil pH	5.63
Specific surface area, m²/g	46
CEC, meq/100 g soil	54
Organic content %	57
Undrained shear strength, kPa	6.5
Zeta potential, mV	-78
Fiber content	41.94%

Humus as a most chemically active fraction of the organic soil colloids can help to justify the underlying reasons for the physicochemical effects on the tropical highly organic matter soils from the EK treatment [2]. The soils had a net negative charge because of the negative charges on the organic matter and on the layer silicates of the mineral portion (kaolinite), which was limited in amount when compared with humus.

3.1. Diffuse double layer interactions of treated organic soil

The ζ of pure organic soils varied from +25.1mV to -95.4 mV at pH 2.8 and 11.3, respectively. Also, ζ for pure organic soil was almost zero at pH 3.25 to 3.35 (Fig. 1).
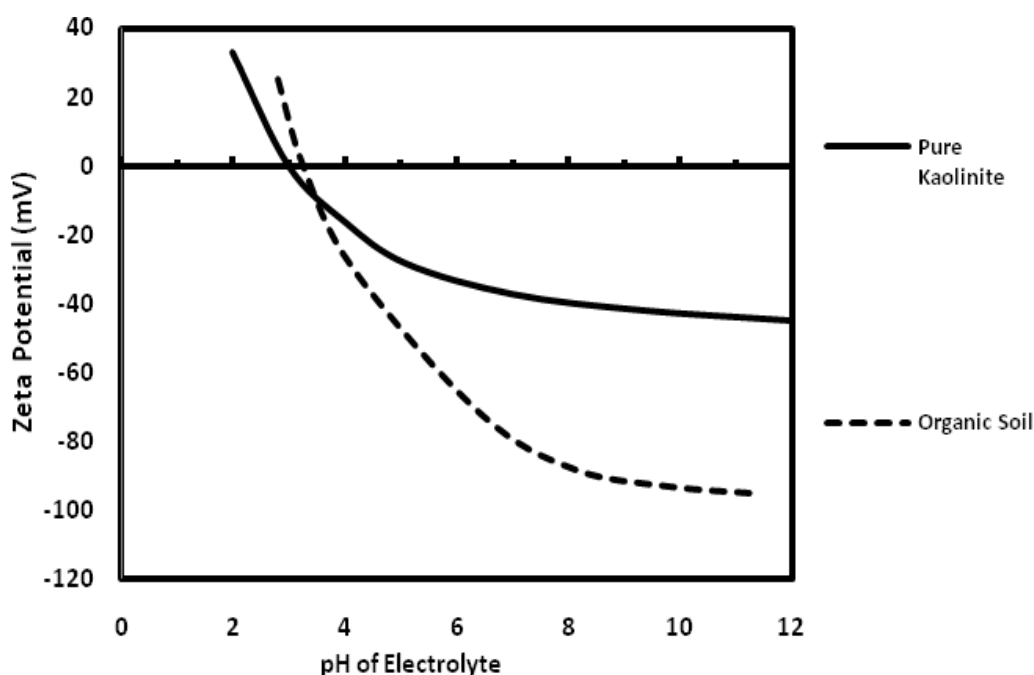
Figure 1. The variation of ζ for pure organic soil and kaolinite

Organic soil suspension pH is significantly depend upon concentration of electrolyte type, and dissociation are also in a direct relation with chemical reagents. This means the charge is affected by pH. For organic soil, as the pH went up, the net negative charge was produced and as the pH dropped, there was less and less negative charge (Fig. 1), however, ζ showed different trend with pH when organic soil immersed in cationic electrolytes. Moreover, the variations in ζ with pH could be related to the nature of electrical energy field in organic soil. The natural pH of used organic soil was equal to 5.63 where ζ showed -78mV. The sign of the natural ζ in organic soils is negative (Fig. 1). As mentioned the negative charge of humus is generally believed to be due to the dissociation of H^+ from carboxylic and phenolic functional groups [1, 2].
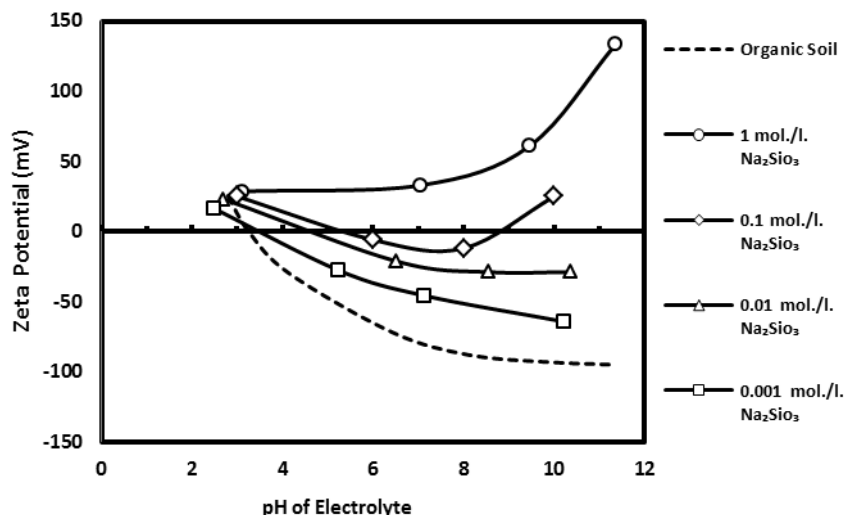
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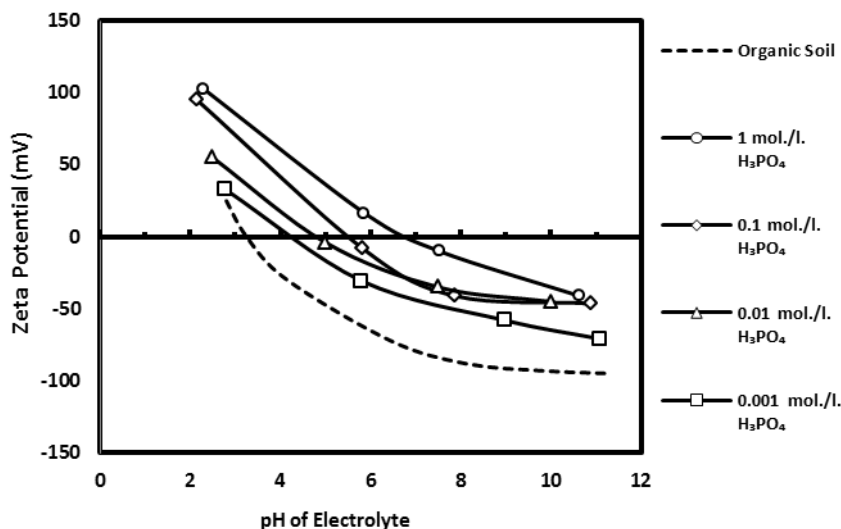
(b)

Figure 2. The variation of ζ with electrolyte concentration of organic soil: (a) Na_2Sio_3, (b) H_3PO_4

In comparison with the pure kaolinite, organic soil has more negative charge and higher pH value at IEP. That is because all charge on humus is strongly pH-dependent, with humic substance behaving like polyprotonated weak acid [31]. However, as for kaolinite as well as other clays, broken edges is belived to arise from dissociation of a proton (H^+) from an exposed OH group. Such dissociation is possible because oxygen atoms at the broken edges are in contact with one rather than two Si of Al atoms [2, 24]. The hydrogens of tetrahedral OHs (those associated with Si) are presumed to be more likely to dissiciate than those of octahedral OH (those associated with Al) [31]. As one might dissociation is strongly pH dependent [24].

The ζ of organic soil in the presence of Na_2Sio_2, H_3PO_4 electrolytes at different electrolyte concentration showed in Fig. 2. As for both Na_2Sio_3 and H_3PO_4, ζ sign did not change and remained negative except for 1 mol/L electrolyte concentration that ζ significantly increased to the +133.18mV at pH equal to 11.35 (Fig. 2).

The ζ of organic particles as a function of pH for various electrolytes namely; Na_2Co_3, $CaCl_2$, $MgCl_2$, and $Al_2(SO_4)_3$ in four electrolyte concentrations, 0.001, 0.1, and 0.1mol/L showed in Figure 3. ζ of organic soil changed from +189.22 at pH 4.43 according to 0.1 M $CaCl_2$ (Fig. 3.a) to -88.2 at pH 9.69 in 0.001 M according to Na_2Co_3 (Fig. 3.c). It should be mentioned that ζ for some of solutions such as $CaCl_2$ and $MgCl_2$ showed higher than expected.
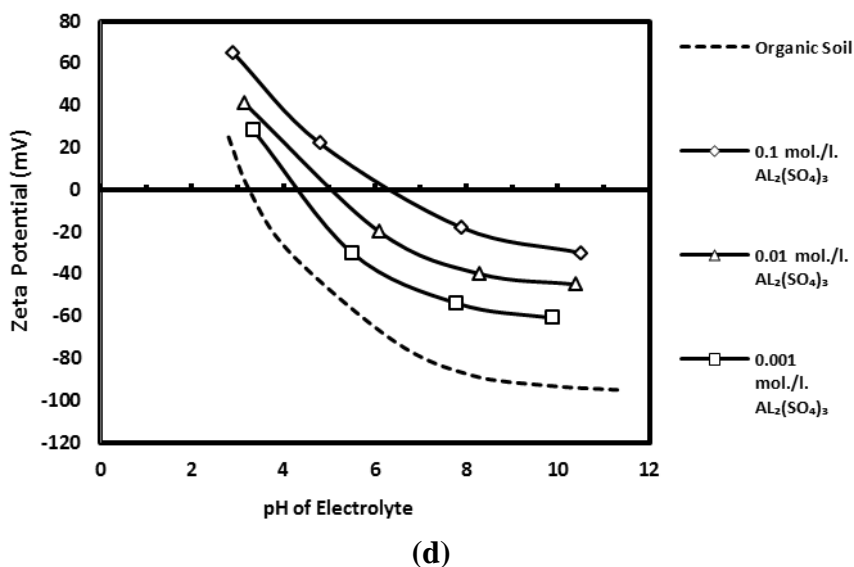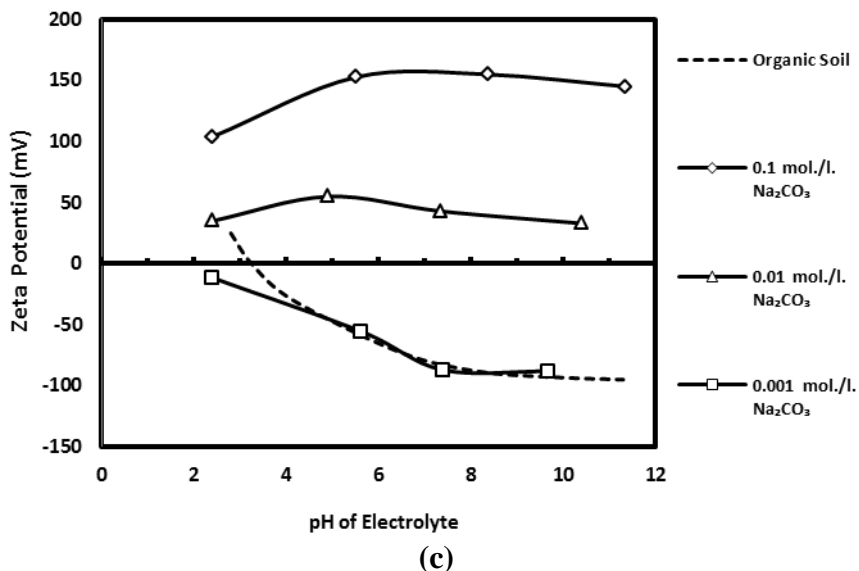

(a)

(b)

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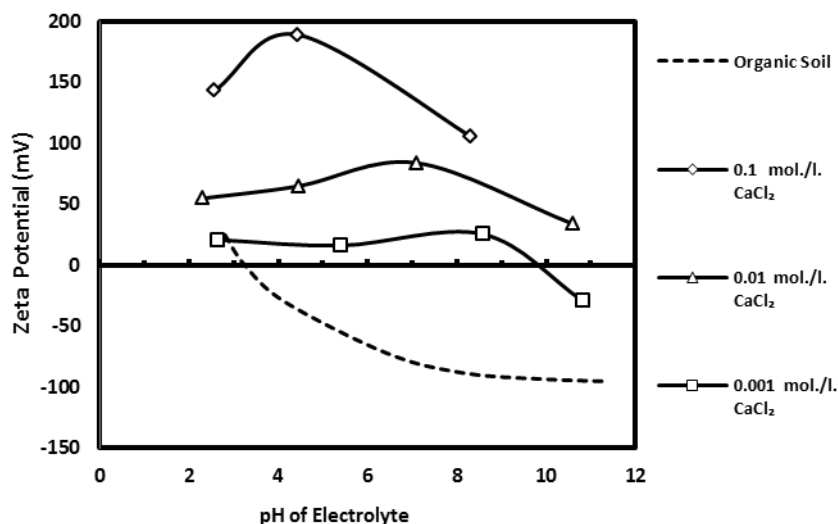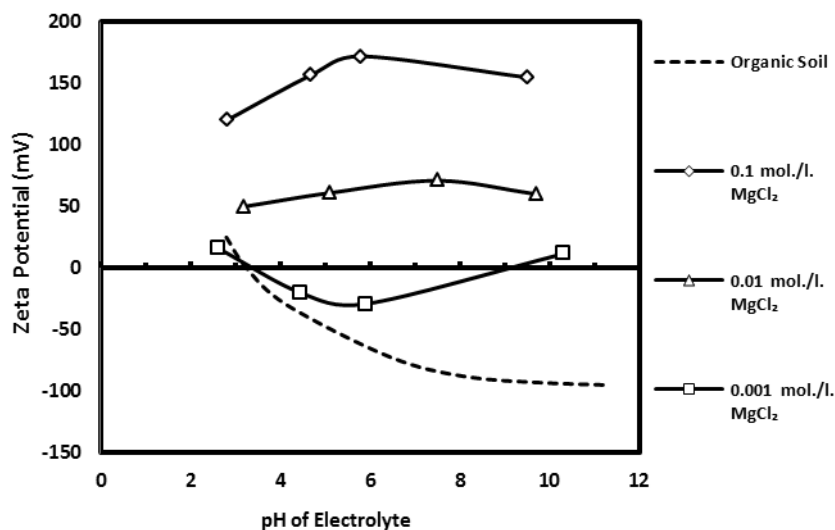
Figure 3. The ζ variation of organic vs. pH in the presence of various electrolytes, (a) $CaCl_2$, (b) $MgCl_2$, (c) Na_2CO_3, (d) $Al_2(SO_4)_3$,

Due to high ion concentration in diffuse double layer thickness of colloids immersed in these reagents, ζ could not be calculated using Smoluchowski equation. The higher ζ lead to more electric conductivity as well as suitable environment regarding EO flow injection [13].

The negative sign of ζ changed to the positive area moving towards the alkaline part in pure organic soil (Fig. 3). It is well understood in contact with water or any chemical solution, organic soil particles are surrounded by the diffuse double layer. Also, as in EK when an electric potential is applied across a wet soil mass, anions are attracted to the anode, while cations are attracted to the cathode. Mobile water and Cations migrate toward the cathode in the same direction, and this flow is called electro-osmosis (EO) [31]. Reverse in ζ sign from negative in pure organic soil to positive in $CaCl_2$ solution lead to change in the EO flow through the EK cell. As for organic soil, ζ behavior of

CaCl$_2$ solution showed a peak value of 189.22, 84 and -29.4 mV at electrolyte concentration of 0.1, 0.01 and 0.001, respectively. PHs at Peak values of ζ for CaCl$_2$ was 4.43, 7.1 and 10.83 at 0.1, 0.01 and 0.001 mol/L electrolyte concentration, respectively (Fig. 3.a). The same behavior has seen regarding MgCl$_2$ and Na$_2$CO$_3$ (Fig. 3.b&c). The ζ of organic soil immersed in Al$_2$(SO$_4$)$_3$ at electrolyte concentrations of 0.1, 0.01 and 0.001 mol/L were 65.3, 41.6 and 28.1 at pH respectively, in their natural pHs which were 2.9, 3.15, and 3.35 respectively (Fig. 3.d). Observed results clearly show that electrolyte concentration has significant change in the increasing ζ value in acidic part and decreasing in alkaline part (Fig. 3).
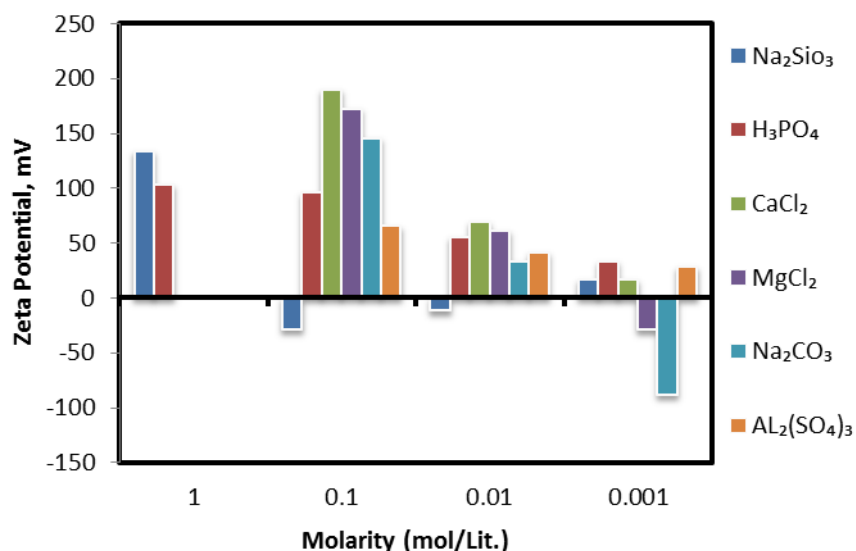

Fgiure 4. Effect of electrolyte concentration on ζ of organicsoil with different additives in their natural pH

Regardless the ζ sign, the natural ζ of the organic soils immersed in cationic and saline solutions increased with increasing electrolyte concentration (Fig.4). As mentioned, the thickness of diffuse double layer in some cases like MgCl$_2$, CaCl$_2$, Na$_2$CO$_3$ and Al$_2$(SO$_4$)$_3$ in 1 M was more than that can be calculated by the von Smoluchowski theory (1921) [26]. There was no compression in double layer thickness regarding immersing organic soil particles in electrolytes at various concentrations. For instance, Na$_2$CO$_3$ (Fig. 3.c) and H$_3$PO$_4$ (Fig. 2.b) at electrolyte concentration of 0.001 mol/L showed ζ equal to -88.2mV and 33.11mV at their natural pH which were 9.69 and 2.77, respectively.

Theories on EO flow suggest that decreasing the diffuse double layer decreases EO [14, 24]. Results proved that in the organic soils, higher electrolyte contents had more influence over increase of the natural ζ in comparison with reagents having less electrolyte content (Fig. 3). Thus, the relationship between electrolyte concentration and ζ are not only under influence of electrolyte concentration contents, but also under influence of electrolyte and soil types.

3.2. Effect of electrolytes on specific conductivity

The specific conductivity concept is typically applied to the case of homogeneous electrolytes. Inside the charged pores, the ionic concentration and the potential vary from the charged surface to the bulk solution [23, 29].
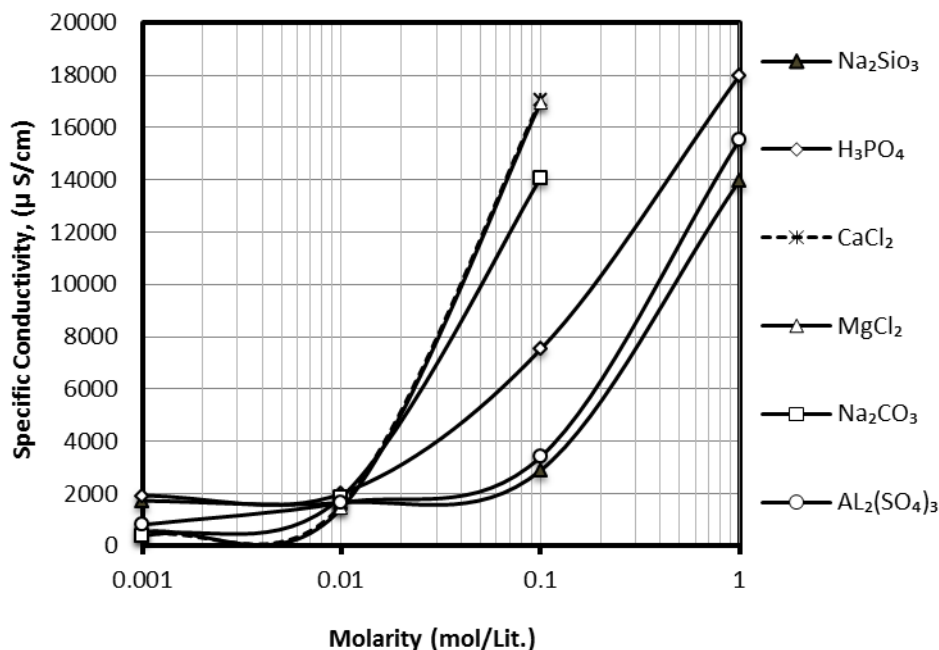

Figure 5. Effect of electrolyte molarity in suspended organic soil on specific conductivity

The specific conductivity titration curves were not very similar (Fig. 5). $CaCl_2$ and $MgCl_2$ were exactly similar and also found to be most conductive reagents. Moreover, the rest of chemical species, Na_2Co_3, H_3PO_4, and $Al_2(SO_4)_3$ were almost similar with a lower value in specified molarities indicating that $CaCl_2$ and $MgCl_2$ are more susceptible to work as secondary additive. Also, based on Helmholtz Smoluchowski equation more specific conductivity cause to increasing in ζ values (Fig. 5). This also indicates the significant effect of reagents on the EO flow and EM through the soils.

3.3. Unconfined Compressive Strength Tests

As for Phase II, the 14 days undrained shear strength (C_u) on treated organic soil recorded in unconfined compression tests as for both Na_2Sio_3, and H_3PO_4. Tests were conducted in Phase II by mixing soil samples with 36 different binders (Table 1). As mentioned, the mixing procedure and curing system used in this study project was that adopted in the EuroSoilStab project (Design Guide: Soft Soil Stabilization). The stabilized organic soil specimens showed significant increase in unconfined compressive strength for all stabilizers when compared with the shear strength of the natural organic soil sample, which amounts to 5-8 kPa. The strength increased with an increase in the molarities of Na_2Sio_3 however there was no considerable increase in case of H_3PO_4 even in high amendment concentration (Fig 6). High strengths were achieved with the higher Na_2Sio_3 stabilizer.
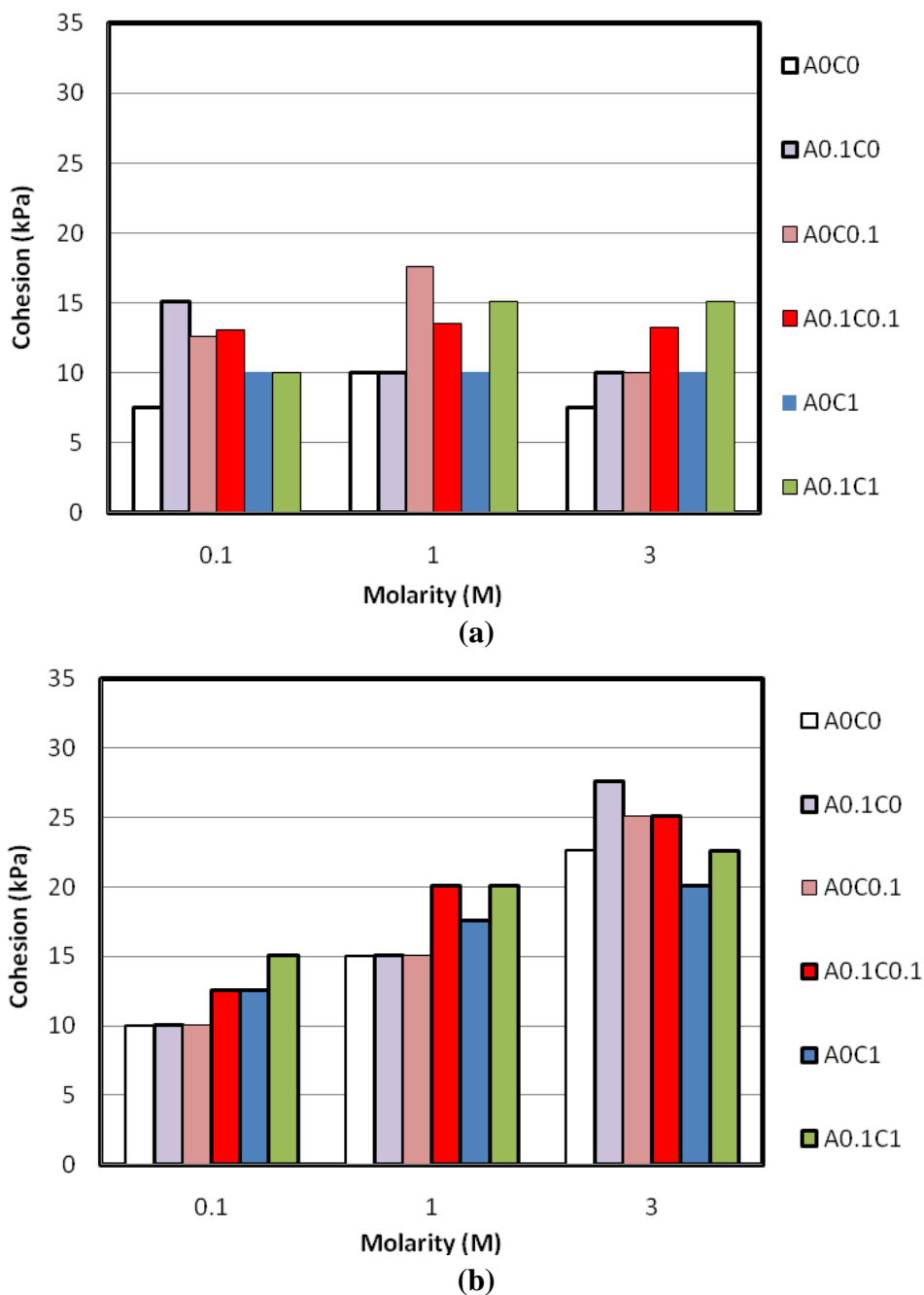
(a)

(b)

Figure 6. Phase II, the 14 days undrained shear strength (Cu) for treated organic Soil, (a) H_3PO_4 (b) Na_2SiO_3

3.4. Effect of $CaCl_2$ and $Al_2(SO_4)_3$ on treated organic soil

It is well understood, $CaCl_2$, as a saline solution of calcium and chlorine behaves as a typical ionic halide. As for EO with injection, in addition to the EO effects, the increase of cation in the soil due to the injection of $CaCl_2$ will result in an increase of electric conductivity and hydration of cation, which causes more absorbed water, along with the cation, to migrate towards the cathode. The EO

effect using $CaCl_2$ is certainly enhanced, compared with the EO only. Previous studies also proved that the EO injection tests have less loss of voltage, more current, and larger amounts of drained water than those using EO only. On the other hand, saline solutions which were mixed into the soil as part of binders will increase electrochemical effects such as an increase of cation exchange and precipitation on the surface of humus.
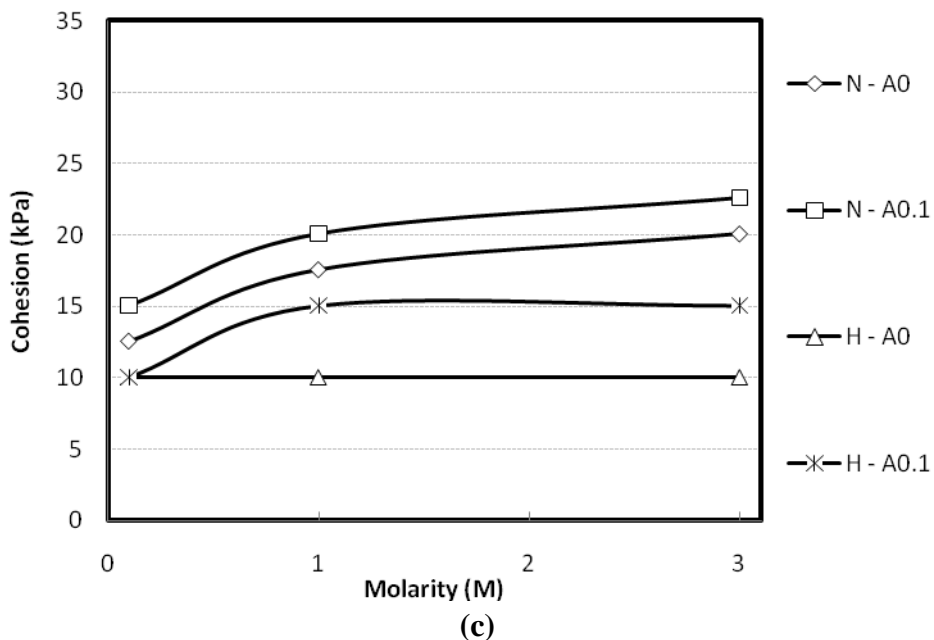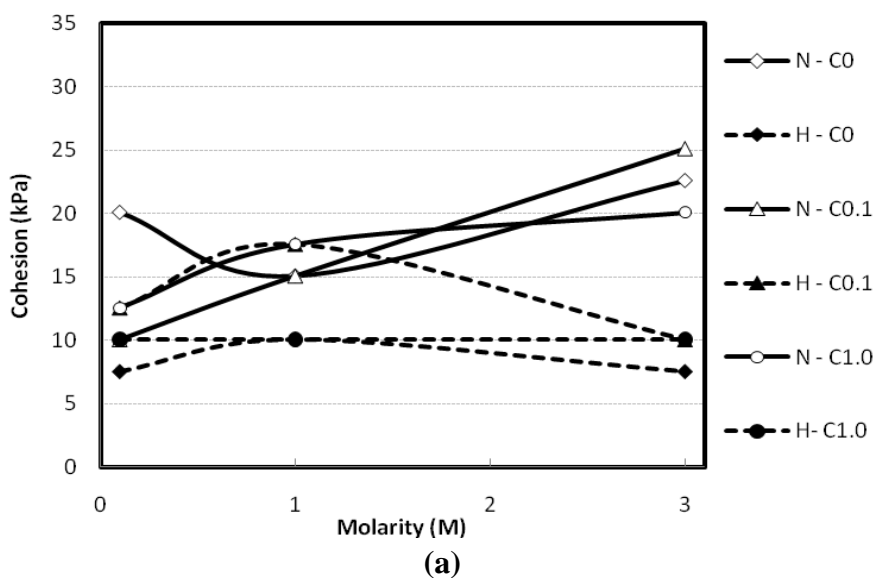

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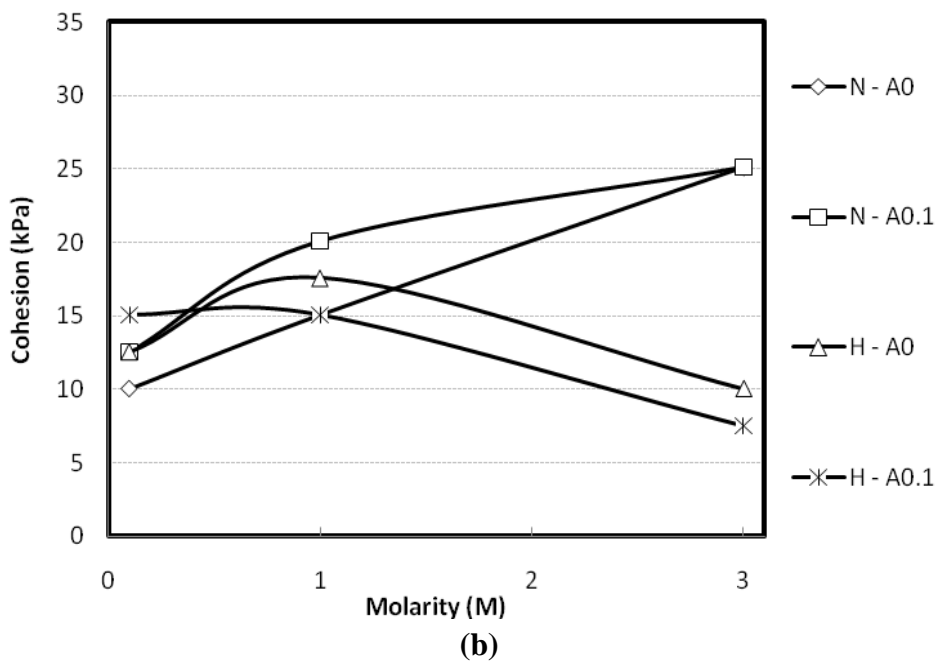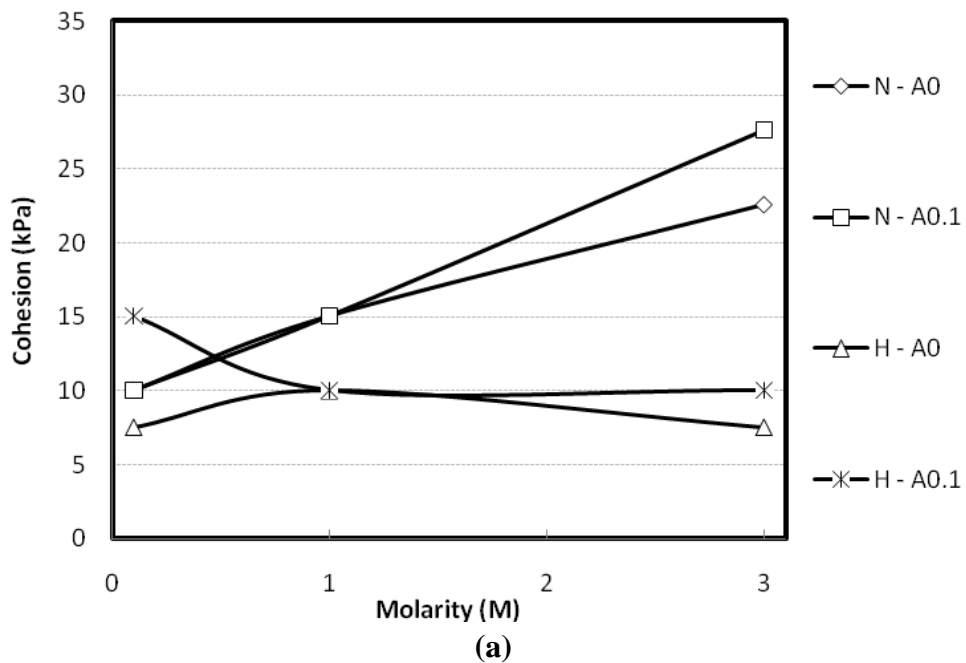
Fgiure 7. Effect of electrolyte concentration on undrained shear strength of organic soil, (a) Without $CaCl_2$, (b) 0.1 M $CaCl_2$, and (c) 1M $CaCl_2$

As in many investigators' studies on clay, Ca^{2+} can exchange Na^+ on the surface of clay, which makes the double layer of the clay thinner, resulting in denser organic soil particles and thus increasing shear strength [32-35]. An increase in shear strength of soil can also be related to precipitation in the soil [36-40].

In the absence of $Al_2(SO_4)_3$, the Cu for 1mol/L of Na_2Sio_3 at 0, 0.1, and 1 mol/L $CaCl_2$ were almost same and equal to 15.07 kPa (Fig. 7). However having 0.1 mol/L of $Al_2(SO_4)_3$, the C_u for same molarities of Na_2Sio_3 at 0, 0.1, and 1 mol/L $CaCl_2$ were 15.07, 20.09, and 20.09, respectively, which means using $Al_2(SO_4)_3$ increase 25% unconfined compression results (Fig. 8).
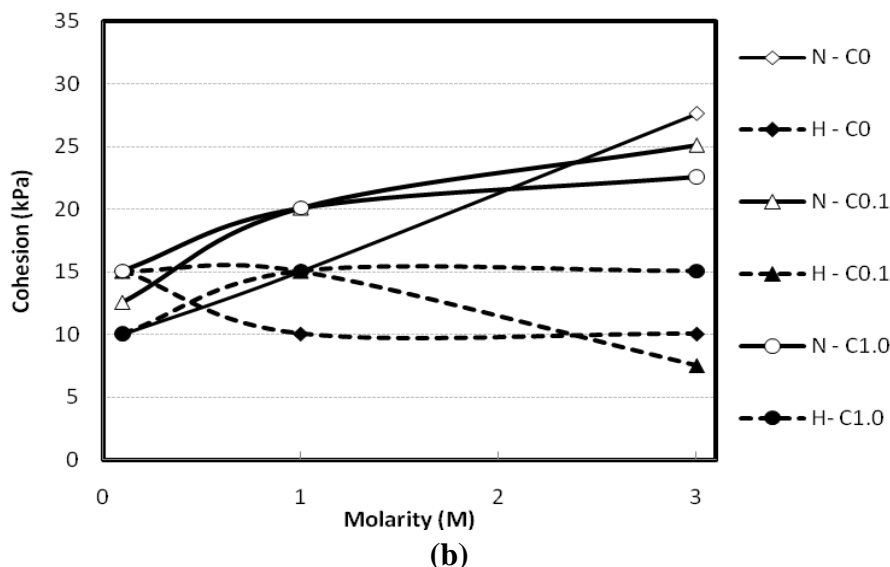

(a)

Figure 8. Effect of electrolyte concentration on undrained shear strength of organic soil, (a) Without $Al_2So_4^3$, and (b) 0.1 M $Al_2(SO_4)_3$

Although, $CaCl_2$ enhaced the EO flow injection process, it will reduce the strength properties of organic soil. Also, adding $Al_2(SO_4)_3$ in Na_2Sio_3 treated soil increase the undrained compression strength significantly. As for 3mol/L Na_2Sio_3 and in the presence of 0.1 mol/L of $Al_2(SO_4)_3$, the C_u at 0, 0.1, and 1 mol/L $CaCl_2$ were 27.62, 25.11, and 22.60. There was no considerable difference between 1 and 3mol/L Na_2Sio_3 according to presence of 0.1mol/L $Al_2(SO_4)_3$ (Fig. 7. c).

4. CONCLUSIONS

This paper describes effect of several new binders in order to stabilizing organic soils. Process has started from finding the effect of each electrolyte on diffuse double layer of organic colloidal. Then optimizing the ζ results led to choosing several amendments, as susceptible option regarding stabilizing. Also a comprehensive series of unconfined compression tests presented to give a good understanding of the behaviour of the stabilized organic soil with new suggested binders.

Using ζ results could be an effective in situ ground improvement tool in soft organic soils, particularly where ground movements need to be controlled. Then batch tests were done to find the effect of different binders on undrained shear strength of organic colloids. Based on the results of this study, the following conclusions may be drawn:

- The results show that in the batch tests, shear strength increases of up to 350% of the natural organic soil's baseline strength can be achieved by adding the Sodium silicate, while little strength increase resulted from the phosphoric acid addition.

- $CaCl_2$ at 0, 0.1, and 1 mol/L and $Al_2(SO_4)_3$ at 0, 0.1mol/L were mixed with Na_2Sio_3 and H_3PO_4 at 0.1, 1, and 3mol/L as binders for stabilizing organic soil. Although, higher molarities for Na_2Sio_3 as binders led to higher C_u, using $Al_2(SO_4)_3$ improved significantly the C_u even for lower

Na_2Sio_3 molarities. This could be due to better interaction between humus in organic soil as initial negative charge part and Na_2Sio_3 in presence of $Al_2(SO_4)_3$.

- In the presence of $CaCl_2$ at 0.1mol/L and absense of $Al_2(SO_4)_3$, effect of using high Na_2Sio_3 molarities as for achieving to high C_u reduced. This effect in turn will contribute to an increase in soil hydraulic conductivity through the soils. In contrast, negatively charged soil particles repel each other, resulting in dispersion and decrease in soil hydraulic conductivity.

- Although in the absence of $Al_2(SO_4)_3$ good bonding obtained according to using $CaCl_2$ at 1mol/L mixed with H_3PO_4 at 1mol/L, there was no considerable increase in C_u for H_3PO_4 as for stabilizer. This means H_3PO_4 cannot be a suitable option acting as a good stabilizer regarding organic soil stabilization.

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