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Zeta Potential of Tropical Soil in Presence of Polyvinyl Alcohol

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The focus of this research is to study the influence of polyvinyl alcohol (PVA) adsorption on the zeta potential properties of two tropical soil namely peat, and kaolinite. Two different PVA species were used, partially hydrolyzed (PVA-T) as well as fully hydrolyzed (PVA-F). As results, adding the PVA species into the suspended colloids led to an increase of zeta potential in their surfaces, contrary to measuring done in water. The zeta potential of the Peat soil varied from 25.26 to -95.1 mV, according to the PVA type and concentration. Kaolinite, however, showed zeta potential varied from +32.4 to -161.78. Negative charge in both peat and kaolinite soils were highly pH dependent and their surface charge dropped to zero at pH 3.1 to 3.4. Using PVA cause a significant reduction in isoelectric point (IEP) of soil samples to about 1.9 for peat and 2.1 for kaolinite. PVA-F enhanced the negative zeta potential than PVA-T. However, increasing of zeta potential in negative sign with increasing degree of polymerization can be observed in both PVA-T and PVA-F samples.

Keywords: Polyvinyl alcohol, zeta potential, isoelectric point, tropical soil

1. INTRODUCTION

Modification of surface charge characteristics of solids is a new method in several geoenvironmental engineering applications, including electrobioremediation [1], electrochemical remediation, ion injection [2], contaminant removal [3], and electroosmotic (EO) dewatering [4-8].

Zeta potential is a function of surface coverage by charged species at a specified pH, and it is theoretically determined by the activity of the species thorough the solution [7]. The zeta potentials of colloidal particles occurring in soils, such as iron oxide minerals and clay, directly affect the efficiency of the electroosmotic (EO) [5, 9-10]. The potential at the surface can be fixed or varied; the value of zeta potential changes with compression or expansion of the Electrical Double Layer, EDL, with adsorption of chemical reagents or polymers at the surface either non-uniformly or uniformly, thereby changing the composition of the EDL and hence the net charge of the 'kinetic' particle [9]. Until now, it has been reported that the effect of pH on the zeta potential is more important than electrolyte concentration in the EK process [2, 7, 10], however, soil pH is considerably affected by electrolytes concentration and type (i.e. polymers) [11].

Due to the fact that polymers show the great variety of stabilization mechanisms, they find a good practical application in many fields of human activity, e.g. environment protection, industry, and agriculture [12]. The stabilization processes are widely used in production of cosmetics, food processing, paints, pharmaceuticals, and coatings whereas the flocculation ones in water purification, flotation, separation of hazardous solids from chemical waste, oil recovery and mineral technology (purification of ceramic powders) [13]. A polymer such as polyvinyl alcohol (PVA) has excellent emulsifying, film forming, and adhesive properties. It is also resistant to grease, oil, and solvent. It is nontoxic and odorless. It has high flexibility and tensile strength, as well as high oxygen and aroma barrier properties [14]. However these characteristics are dependent on humidity, in other words, with higher humidity more water is absorbed. PVA is also a quick dissolver and fully degradable [12-17].

Although the characteristics of zeta potential system have been studied extensively [3, 10, 11, 18 - 19], few studies conducted in the relationship between the interaction of EDL in the mixture of PVA with aluminosilicates and oxides of tropical soil. The main objectives of this research is to investigate the difference of zeta potential between the single system of kaolinite and peat colloid and the binary systems containing PVA at different concentration, and to discuss the interaction between EDL of common tropical soil and PVA based on the change of zeta potential.

2. MATERIAL AND METHODS

Soil samples were collected and sealed to maintain soil moisture in accordance with the British Standard Institution [20] methods of test for soils (BS 1377-1:1990) from several locations of Serdang, Malaysia. The pH, water content, specific surface area, and zeta potential were measured in accordance with BS 1377-3:1990, BS 1377-2 1990, BET technique, ASTM D 4187 [21], respectively. The CEC of soils also was measured at pH 7 with ammonium acetate [22].

2.1. Soil Sample Purification

The solid sample was dried for 24 h at 105°C, ground, and then sieved by a 75-µm sieve. For the electrophoretic mobility measurements, the suspensions were prepared in a polyethylene container by mixing 0.1 g of the soil sample with 100 ml of double-distilled water with a specific conductivity of 1.4µS/cm, after adding the desired concentration of PVA. Soil samples were treated before their use in the experiments. In order to obtain homogenized colloidal particles: The suspension containing particles (100 mg L^{-1}) were mechanically stirred for 24 h. After agitation for 30 min in a shaker, the suspension was allowed to stand for 25 min to let the larger particles settle. An aliquot taken out from the suspension was slowly poured into the electrophoresis cell. Molybdenum anode and platinum cathode electrodes were inserted into the cell. After a desired electric voltage applied between the electrodes, the movement of particles was observed via a microscope nodule. To minimize reading error, a minimum of 10 particles are tracked and their average time is calculated. The applied voltage during the measurements was in range of 40–100V. The pH was adjusted in the range from 2.5 to 11 before each measurement by drop wise addition of 10^{-3} N and 10^{-1} N of NaOH. The pH that was observed after the zeta potential measurement was recorded as the final pH.

2.2. Zeta Potential Measurements

To determine the zeta potential of peat and kaolinite, electrophoretic mobility measurements were conducted using a Zeta Meter Model 3.0^+ (Zeta Meter Inc., USA). The instrument, using Smoluchowski's equation, determines the electrophoretic mobility of the particles automatically and converts it to the zeta potential [23].

The zeta potential measurements were measured as a function of pH, and adsorbed amount of PVA on the surface of colloidal particles. After adsorption period, we measured the zeta potential of particles which were obtained by centrifuging the suspension. For the zeta potential measurements, a 50mg sample was transferred into aqueous solution and the soil particles mixed homogenously with a magnetic stirrer. All the measurements were carried out therefore at 100 mg/L solid concentration. The voltage 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 10 particles are tracked and their average time is calculated. The pH measurements were carried out using a Mettler–Toledo Model MP220 pH meter combined with a Mettler–Toledo Model InLab 413 pH electrode. Boiled and double-distilled water was used to prepare aqueous solutions for all experiments to measuring zeta potential.

3. RESULTS AND DISCUSSION

3.1. Baseline soil samples characteristics

Kaolinite showed a CEC of only 10meq. per 100 g, compared to other minerals such as montmorillonite with 80-100 meq. per 100 g or highly organic soil like peat between 60-120 meq. per 100 g, is not considerable. As expected CEC corresponding peat was 85 meq. per 100 g which is much higher than kaolinite specimen. Physicochemical characteristics of peat and kaolinite used in present research showed in Table 1.

Table 1. Physico-chemical properties of soil samples

Parameter	Peat	Kaolinite	Standard
Color	Brownish	Yellow	
Moisture content,%	158	24	BS 1377-2 1990
Soil pH	6.65	5.2	BS 1377-3:1990
Specific surface area, m²/g	46	15	BET technique
CEC, meq/100 g soil	85	10	After Chapman, 1965
Organic content %	78	2	BS 1377-3: 1990
Zeta potential, mV	-78	-28.4	ASTM D 4187

3.2. Zeta potential and pH in non PVA solution

The zeta potential of peat soil suspended in water varied from +25.1mV to -95.4 mV at pH 2.8 and 11.3, respectively. Also, zeta potential for peat was almost zero at pH 3.25 to 3.35 (Fig. 1). For peat, 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). Moreover, the variations in zeta potential with pH could be related to the nature of electrical energy field in peat as highly organic soil. The natural pH of peat was equal to 6.65 where zeta potential showed -78mV.

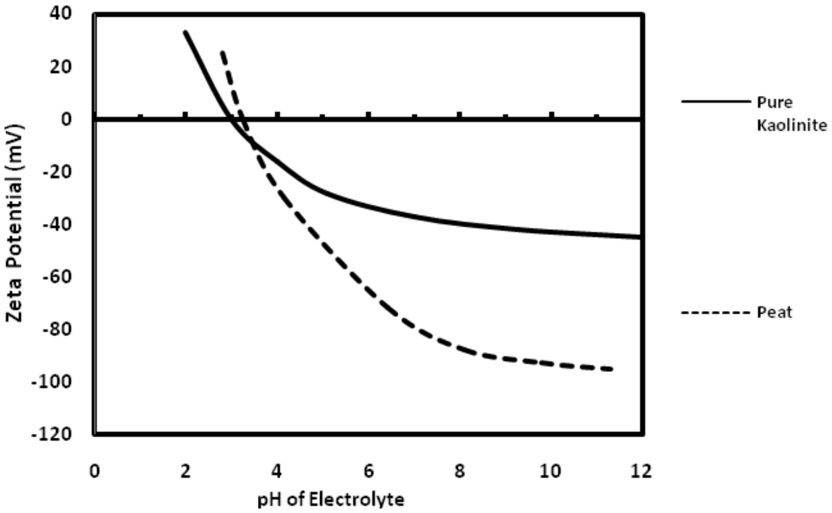


Figure 1. Zeta potential versus pH for peat and kaolinite

The zeta potential of kaolinite particles suspended in water varied from +32.9 mV at pH 2.03 to -41.9 mV at pH 12.2. The zeta potential was almost zero at pH 3.2 to 3.4 (Fig. 1). The natural pH of kaolinite soil was 5.2 where zeta potential showed -28.4mV. The sign of the natural zeta potential in kaolinite also was negative (Fig. 1). The source of negative electric charges on the broken edges of kaolinite is belived to arise from dissociation of a proton (H^+) from an exposed OH group. This is possible because oxygen atoms at the edges are in contact with one rather than two Si of Al atoms [9, 24-27]. The hydrogens of tetrahedral OHs (those related with Si) are presumed to be more likely to dissiciate than those of octahedral OH (those related with Al). As one might dissociation is strongly pH dependent [11, 24-25, 28]. In comparison with the pure kaolinite, peat soil has more negative charge and higher pH value at IEP. This is due to stronger pH-dependent behavioral of humic substance which behaves like polyprotonated weak acid [9]. However, peat soil suspention pH is significantly depend upon concentration of electrolyte type, and dissociation are also in a direct relation with chemical reagents. It means the charge is affected by pH [25, 29-30].

3.3. Effect of polyvinyl alcohol on interactions in electric double layer of tropical soil

Figure 2 and Figure 3 show the zeta potential of peat and kaolinite soil particles as a function of pH for various concentrations of PVA-T as well as PVA-F.

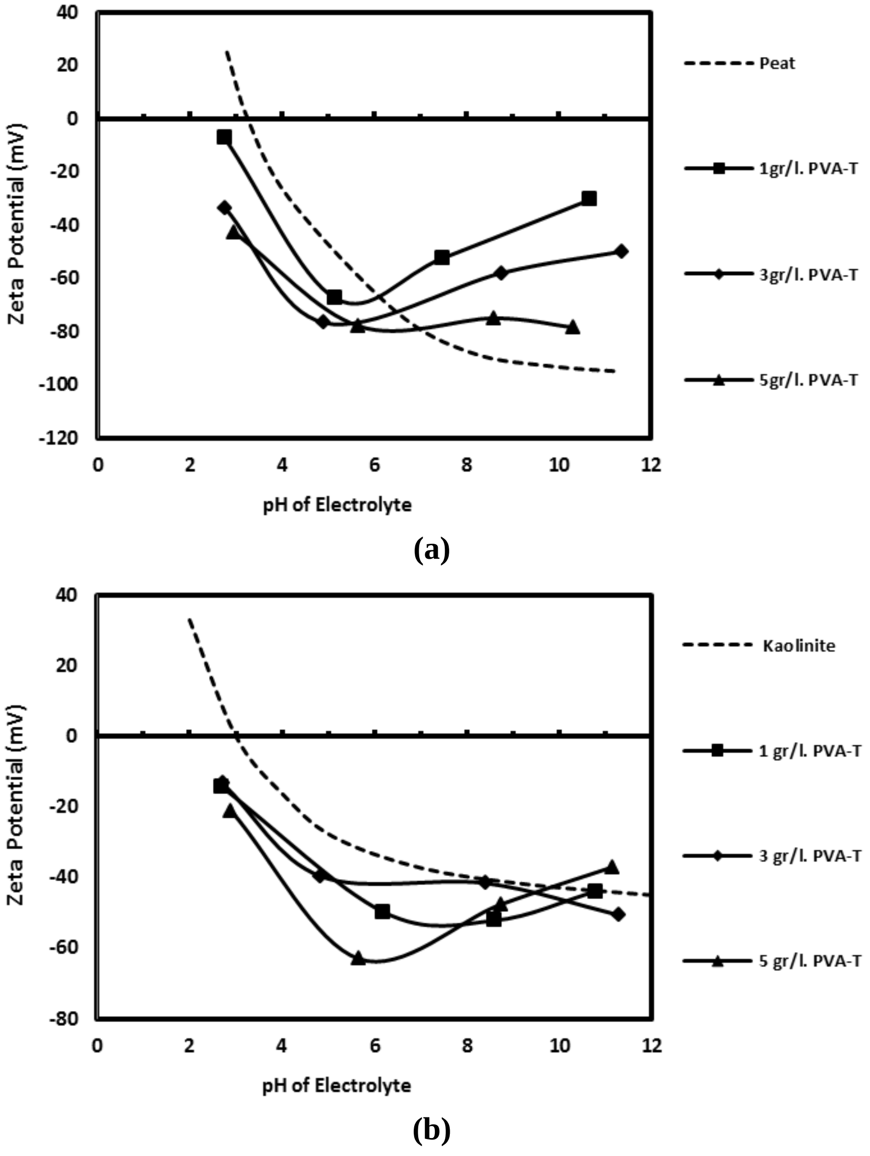
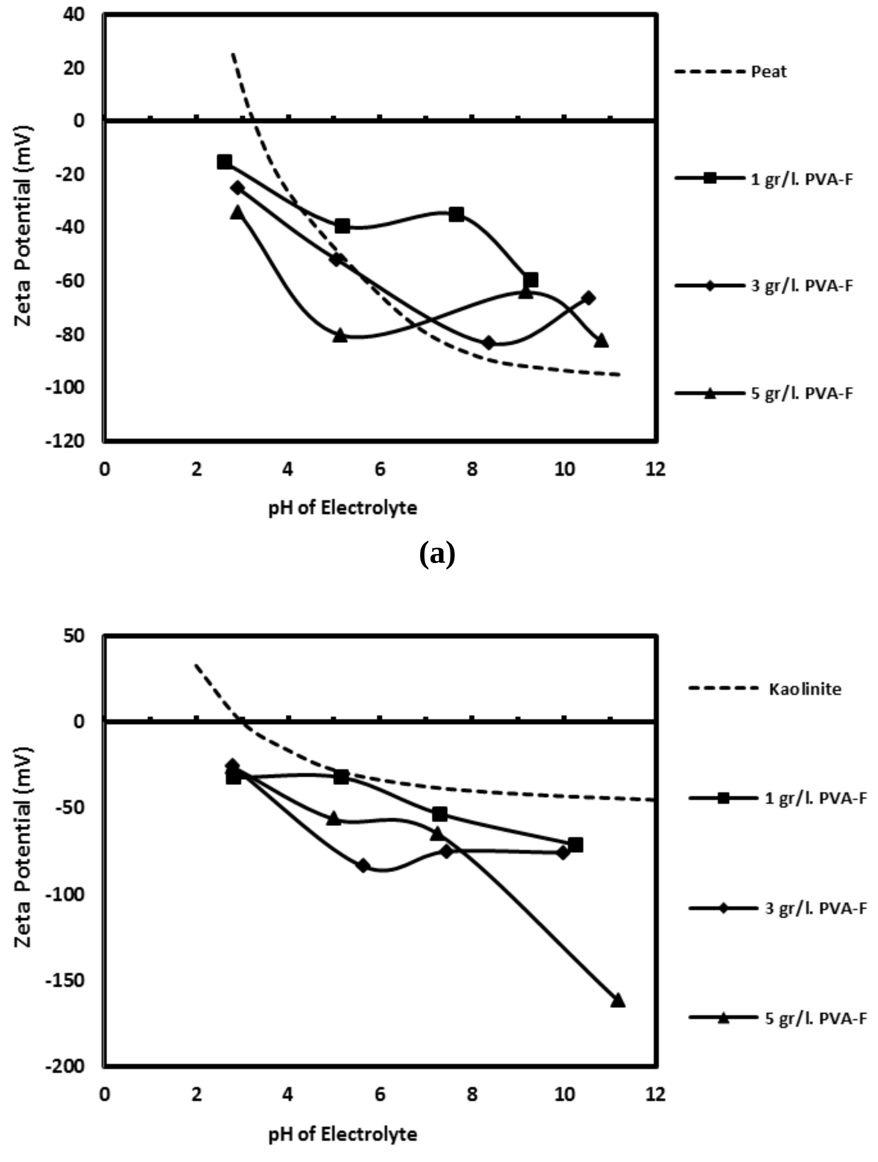


Figure 2. Effect of PVA-T concentration on zeta potential (a) Peat, (b) kaolinite

Observed results clearly show that PVA concentration has significant influence in the increasing zeta potential value in acidic part and decreasing in alkaline part. Immersing kaolinite particles inboth PVA types caused increasing in EDL (Fig. 2.a and Fig. 3.a). On the other hand, zeta potential for peat soil suspended in 1, 3, and 5 gr/L of PVA-T at pH~7 were equal to -60.3, -72.1, and -79.35mV. It means the more concentration of PVA led to longer EDL length. The rest of specimens also showed a similar trend (Fig. 2 and Fig 3).



(a)

(b)

Figure 3. Effect of PVA-F concentration on zeta potential (a) Peat, (b) kaolinite

For tropical soil used in this research, resulted surface charge from 1 and 3 gr/L PVA solution were almost similar in both alkaline and acidic parts, however, using more than 3 gr/L showed similar EDL thickness. Theories on EO flow suggest that decreasing the EDL decreases EO [8, 9, 25, 31]. It means to optimizing the EO flow injection, using 3 gr/L of any PVA type could be consider as a suitable option. The concentration however could change in real EK treatment. It is also important for soil to be adjusted using zeta potential measurement, later doing EK and finally field operation.

Figure 4 showed suspended peat in both PVA-T and PVA-F electrolyte with different electrolyte concentrations of 1, 3, and 5 gr/L. zeta potential is equal to -52.5, -31.21, and -63.06mV respectively at their natural pH at 7.47 and 8.76, and 5.66 respectively, compare with pure kaolinite which were -49.7, -41.6, and -47mV at natural pH of 6.19, 8.14, and 8.73(Fig. 4). On the other hand, using similar concentration for PVA-F solution results a thicker EDL. The peat colloids which suspended in PVA-T solution showed zeta potential equal to –39.3, -83.26, and -80.18 mV, respectively, in their natural pH at 7.66 and 8.37, and 9.18 respectively, compare with kaolinite which the measured zeta potential was -32.08, -61.43, and -56.23mV in natural pH of 5.18, 7.44, and 4.99 (Fig. 4).

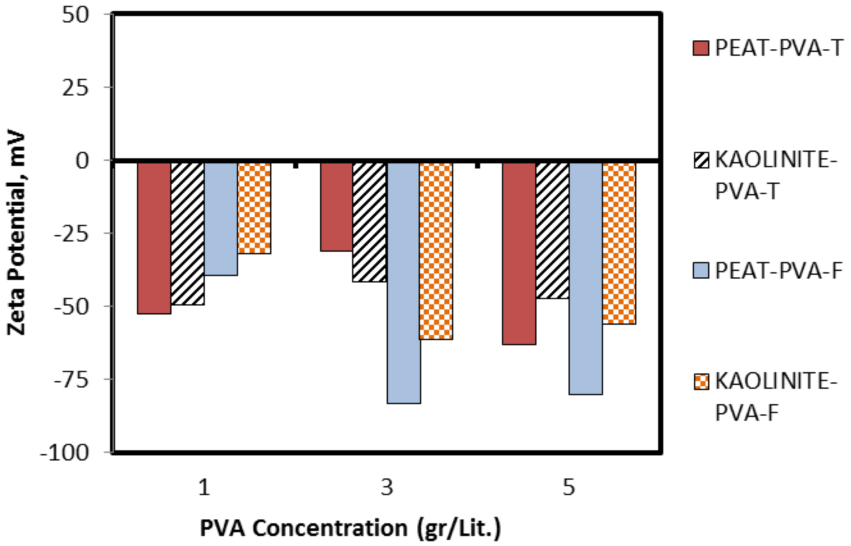


Figure 4. Effect of PVA type and its concentration on the zeta potential of variable charge soil in their natural pH

Although, in kaolinite soils, higher PVA-F electrolyte contents had more influence over increasing the natural zeta potential, consequently EDL thickness, the effect of PVA-T electrolyte concentration was almost negligible in changing the EDL. This could probably due to available silicate minerals in kaolinite composition. The silica surface charge is negative practically in the whole range of studied pH and its absolute value increases when solution pH is more alkaline. It means that pH increase induces stronger electrostatic repulsion between the negatively charged surface and dissociated acetate groups in PVA macromolecules. Despite this fact, the adsorption of PVA on the silica surface takes place, which is testified by obtained changes of the solid surface density in the presence of polymer. Therefore, different non-electrostatic forces have to be responsible for PVA adsorption process on the SiO_2 surface.

Zeta potential of peat suspension also showed a same trend with pH (Fig. 4). Thus, the relationship between electrolyte concentration and zeta potential are not only under influence of electrolyte concentration contents, but also under influence of electrolyte and soil types. This can be attributed to the increasing molecular attraction between the peat and/or kaolinite particles and PVA-F with increasing degree of saponification due to the formation of hydrogen bonds. PVA-T shows inferior compressive and flexural strength to PVA-F due to the presence of ester group which interrupts the formation of hydrogen bond. Also, this could probably due to specific behavior of tropical area soil (i.e. variable charge soils) to adsorb ions. Variable-charge soils generally carry both positive and negative charges, and soil can adsorb cations as well as anions [24, 28, 32]. The adsorption can be electrostatic and/or specific in nature. One important consequence of specific adsorption of anions is that the adsorption may result in a decrease in positive surface charge and/or an increase in negative surface charge of the soil [7, 17, 25, 33]. This kind of effect has been found to induce changes in EK properties of these soils when they specifically adsorb inorganic anions [7, 34-38].

3.4. Effect of polyvinyl alcohol on isoelectric point and electroosmosis in Tropical soils

The EO flow can virtually be eliminated at the IEP point. Therefore, elimination of EO flow in the kaolinite and peat soils suspended in water can be expected at pH 3 to 3.3 however adding PVA caused a shift in pH at IEP towards to the acidic part at pH~1.8 to 2.1 (Fig. 2 and Fig. 3). On the other hand, negative surface charge of particles (negative zeta potential) causes EO to occur from anode to cathode, while positive surface charge causes EO to occur from cathode to anode [25, 32, 35]. When the net charge is zero, soil particles in soil water suspension will not repel each other but will tend to aggregate and form larger particles [7, 10]. This effect in turn will contribute to an increase in soil permeability through the soils. In contrast, negatively charged soil particles repel each other, resulting in dispersion and decrease in soil permeability [28, 31, 39-40]. Figure 5 shows the values of IEP of some minerals in comparison with peat and kaolinite soils immersed in PVA from this study. Since all charge in humus is strongly pH-dependent, the sensitivity of the highly organic soils such as peat to pH changes in EK phenomena is more than mineral soils like kaolinite. Despite this high sensitivity, the IEP point of Peat and kaolinite soils immersed in PVA as depicted in Figure 5 was less than amorphous iron, gibbsite, as well as pure peat and kaolinite. Thus, the consistency of flow direction in tropical soil immersed in PVA may be more probable than that in mineral soils (Fig. 5).

A difference in the IEP between different samples having the same chemical formula has been often attributed to the differences in their crystallographic structure and degree of hydration [42-47]. In general, multivalent ions, polymers, and surfactants tend to adsorb specifically and shift the IEP. The magnitude of the shift depends on the solid to liquid ratio. The shift in the IEP is commonly negligible when the amount of the solute in the system is small compared with the proton charge [44-45]. This explains the discussed above difference in the IEP obtained by means of electrophoresis on the one hand and electroacoustics on the other, namely, in electrophoresis (small solid to liquid ratio) traces of the impurities in solution, e.g. silicates leached out of the glassware, may induce a substantial surface coverage and thus a shift in the IEP [24, 41]. In other words, the pristine IEP obtained in the absence of specific adsorption are also valid for sufficiently low concentration (compared with the proton charge) of specifically adsorbing ions. When the concentration of the solute is high enough, its specific adsorption may (but not necessarily does) induce a shift in the IEP. Also, specific adsorption of anions induces a shift in the IEP to low pH for materials having a high pristine IEP (e.g. iron and aluminum oxides and hydroxides) [5, 46, 48]. Specific adsorption of cations changes the EK curves of materials having a low pristine IEP [5, 49-51].

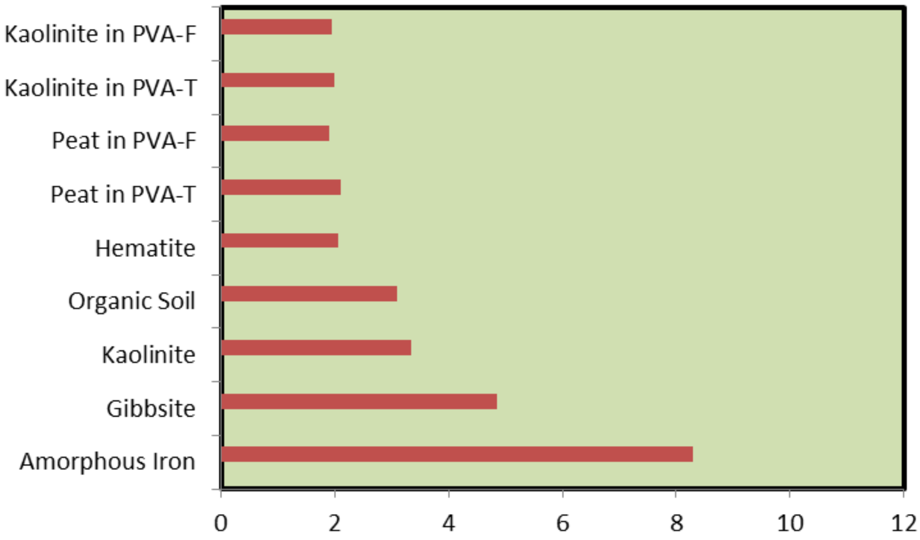


Figure 5. pH at iso-electric point of variable charge soil by comparison with the some minerals

3.5. Effect of electrolytes on specific conductivity

The concept of specific conductivity is typically applied to the case of homogeneous electrolytes [25, 52]. Inside the charged pores, the ionic concentration and the potential vary from the charged surface to the bulk solution [7, 10, 53-55]. It is noteworthy the specific conductivity titration curves were not very similar. It is also found, PVA-T is more conductive than the PVA-F indicating differences in the response of the cations to the tropical soil surface. Suspended peat colloids in PVA-T solution at different concentrations of 1, 3, and 5 gr/L showed Specific conductivity equal to 21, 22.7, and 82.8µS/cm respectively at their natural pH at 7.47, 8.76, and 5.66 respectively, compare with peat soil immersed in PVA-F which were 11.3, 13.4, and 20.1 µS/cm respectively in their natural pH of 7.66, 8.37, and 9.18 (Fig. 7).

On the other hand, suspended kaolinite particles in PVA-T solution at different concentrations of 1, 3, and 5 gr/L also showed the same trend with peat having specific conductivity equal to 31.2, 51.1, and 83.7 µS/cm respectively at their natural pH at 6.19, 8.4, and 8.73 respectively, compare with kaolinite immersed in PVA-F which were 9.3, 12.2, and 17.2 µS/cm respectively in their natural pH of 5.18, 7.44, and 4.99 (Fig. 7). Based on Helmholtz Smoluchowski equation more specific conductivity causes to increasing in zeta potential values which finally increase EO flow injection (Fig. 6) [24, 39, 56-59]. This also indicates the significant effect of PVA-T on the EO flow and electro ion migration through the soils.

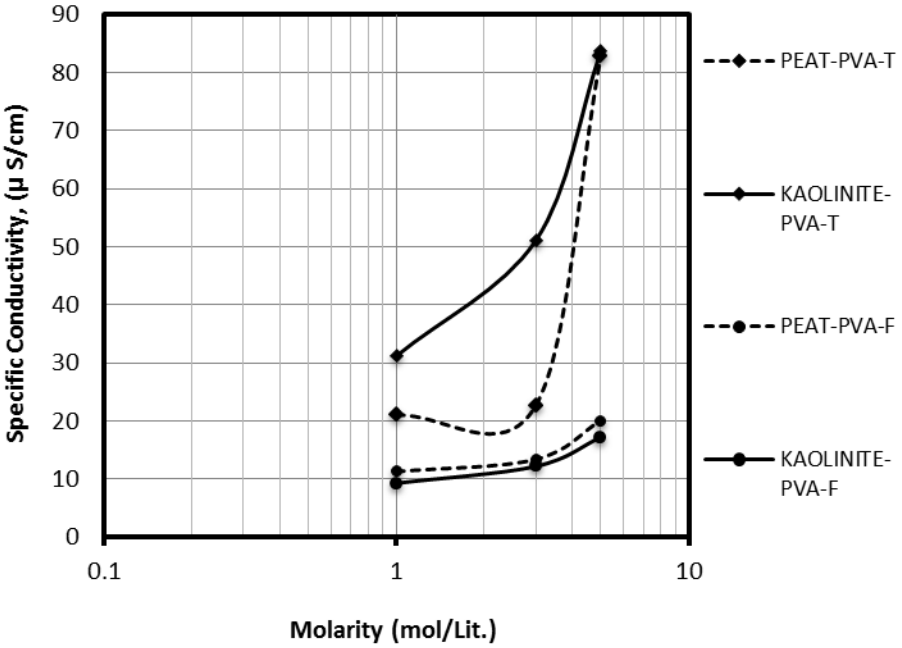


Figure 6. Effect of PVA type and its concentration on the specific conductivity of variable charge soil in their natural pH

4. CONCLUSIONS

In this study we optimized electrophoretic mobility (i.e. the velocity per unit electric field) which gives a measure of the net charge on the colloidal solid particle in order to optimize polymeric stabilizer flow injecting through soils with low permeability. The zeta potential values were obtained to determine the effect of various PVA reagents on the EK properties of two common tropical soils. Such method was performed to assess the effectiveness of injection treatment of PVA and is called electrophoresis technique. Based on the results of this study, the following conclusions may be drawn:

- Among the two types of PVA used in this study, fully hydrolyzed PVA (PVA-F) show the higher improvement in increasing the zeta potential than partially hydrolyzed PVA (PVA-T). However, improvement in EDL properties with increasing degree of polymerization can be observed in both PVA-T and PVA-F samples which can be due to increasing molecular entanglements, which in turn increases the coherence between the tropical soil and polymer. It is also found that for both tropical soils used, PVA-T solution in acidic part tent to increase the EDL thickness (e.g. increasing zeta potential), however, in basic portion caused decreasing the EDL thickness (accordingly decreasing zeta potential).

- Both peat and kaolinite particles surface immersed in water has a net negative surface charge at their natural pH and the IEP of them is at about pH 3.1 to 3.3. However, the IEP of them when they suspend through PVA solution significantly decrease to the values about1.8 to 2.1 (Fig. 5). It means flow direction in such soil immersed in PVA will consist more than those in non PVA. Also, shift in IEP towards the lower pH is often attributed to the differences in their degree of hydration and crystallographic structure.
- Maximizing ion migration in EK could be much achieved if interaction between PVA type and its concentration considered in their IEP. In other words, based on the soil pH and electric field applied (i.e. whether it is anode or cathode) PVA electrolyte is a suitable option to be selected achieving IEP in low pH
- This study shows that the method has the potential to be implemented in the batch scale or even field one, and produce improved soil mechanical properties. However, we may not find the similar trend applying same optimized concentration in an EK cell, because of (i) different soil particles environment, and (ii) different pH conditions across the cell which implement various zeta potential.

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