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Inter-Particle Pressure as influenced by Physicochemical Parameters on Microscale of Saturated Heavy Clay

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Abstract

Determination of inter-particle repulsive pressure in clays is essential in many geotechnical engineering fields, for example, in determination of compressibility and swellability behavior, hydraulic conductivity, shear strength, swelling pressure, and volume change because of changes in matric suction. With respect to heavy clay there are a lot of factors overlapping with each other that control its behavior. This behavior may be companion with great perils on the structures and as a result, complex damages and exorbitant costs are expected. Physicochemical factors such as ion type and concentration play a great role in the properties of such clays. Limited number of past research focused on the effect of ion type and its concentration on such type of soils. This study reports the influence of these two factors on the inter-particle pressure that developed upon saturation of laboratory-prepared heavy clays. A new approach of inter-particle pressure prediction using double layer theory was proposed by investigating the forces within the diffuse double layer of clay particle in saturated state. The experimental results of oedometer tests showed a consistent behavior with the results represented by the double layer theory using special cases of application on micro scale forces. It can be assumed that the forces at macro-scale may be an applicable concept for an assessment of swell characteristics of heavy clay that subjected to saturation.

Keywords: heavy clay, inter-particle pressure, double layer, ion type.

1 Introduction

Heavy clays usually possess a contradictory behavior (swell and shrink) in consequence of variation in its moisture content in the course of time due to environmental changes. The main reason of such behavior was attributed to the environmental conditions that escort its geological formation. In Iraq, heavy clays are spread in the south, middle and north of Iraq and causing sever damages to the structures. Numerous studies have been conducted to investigate the characteristics of heavy clays. However, Problem associated with high heavy clay soil is worldwide, having occurred in such countries as Africa, Australia, India, South America, Canada and the United States. In heave problem, the soil will undergo a volume change upon saturation causing a high inter-particle pressure which may reach high values if volume change is not allowed Al-Omari and Oraibi, 2000 [1] and heave of 1000% if volume change is allowed Jones and Jones, 1987 [2]. However, there are several experimental methods that may be used to determine the inter-particle pressure of clay that subjected to saturation. The time required for such test may extended from one day to several weeks depends on clay type. Furthermore, some methods need a continues sample monitoring in laboratory with exhausted works. The details of each test can be found elsewhere.

2 Ion Exchange Phenomenon and its Importance in Heavy Clay Behavior

Cation exchange is the interchange between a cation (a positive charge ion) in a solution and another cation on the surface-active material. A cation exchange reaction in clays occurs mainly near the surface of colloidal silicate-clay particles and does not affect the structure of the silica-alumina pocket. A clay particle usually carries thousands of negative charges. The presence of these negative charges may be attributed to broken bonds around the edge of the silica-alumina units, the isomorphous replacement of ions within the clay lattice, and the hydrogen of exposed hydroxyls. In montmorillonite and Vermiculite that are the main minerals that compose the heavy clays, broken bonds are responsible for a relatively small portion (20%) of cation exchange capacity, while substitution within the lattice cause about (80%) of the total cation exchange capacity. Thus, in heavy fat clays about (80%) of the exchangeable cations are on basal plane surface.

In fact, the presence of the negative charge on the clay particle was neutralized by the adsorbed cation such as Ca^{++}, Mg^{++}, H^{+}, K^{+}, NH_4^{+}, Na^{+} as shown in Figure (1). These cations are however attracted towards the negatively charged particles by electrostatic attraction and form a diffuse layer of positive charges surrounding the clay particles. Associated with this envelope of charge there will be a gradient of electrical potential (the work required to move a unit charge from infinity to the point in question and is negative for clay surface) whose intensity decreases with increasing distance from the surface of the particle. The negatively charged clay plate and the positively charged cation cloud are called the diffuse double layer. The double layer thickness is thus the distance from the surface required to neutralize the net charge on the particle, i.e., the distance over which there is an electrical potential.

Actually, ion exchange is of very great importance, because the mechanical behavior of soil system is frequently dependent to a large extent on the exchangeable cation present in the clay and the salt concentration of the solution with which it is in contact. The importance of these two factors arises from their influence on the effectiveness of the forces of repulsion between the negatively charged clay particles. Furthermore, the plastic properties of a clay will be very different depending on whether Na^+ or Ca^{++}, for example, is the exchangeable cation. With respect to the heavy clays subjected to saturation the separation of uite lyer will completely separate with Na-base cation, while it may partly separate with other cation such as Ca^{++} or H^+. On the other side, the high negative charge at the clay surface of soils with layer silicate systems, or high cation exchangeable clays, repels anions (co-ions of the surface as they carry a negative charge) to such an extent that anion exchange is of little importance in such soils (Grim, 1953 [3]; Bolt, 1955 [4] and 1956 [5]; Taylor, 1959 [6]; Lambe and Whitman, 1979 [7]; Baver and Gardner, 1984 [8]; Komine and Ogata, 1996 [9]; Mitchell and Soga, 2005 [10]; Rengasamy and Marchuk, 2011 [11]; Israelachvili, 2011 [12]; Thompson and Goyne, 2012 [13]).

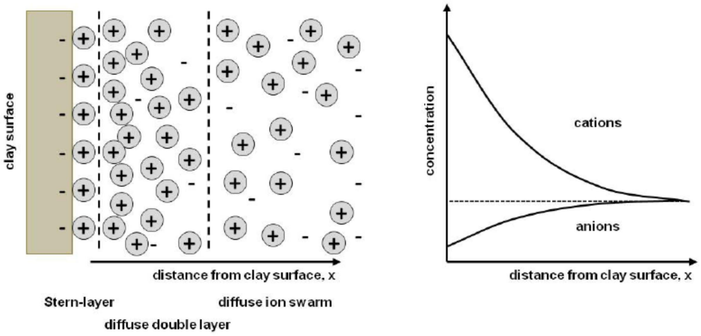

Figure 1. Scheme of the extended diffuse double layer (DDL) theory of Gouy-Chapman and distribution of ions adjacent to a clay surface (after Mitchell and Soga, 2005 [10]).

3 Diffuse Double Layer (DDL) Theory

Shang et al., 1994 [14] discussed various applications of the double layer theory in geotechnical and environmental practices. Tripathy et. al.,2004 [15] stated that very little information is available with regard to the use of the theory for determination of the inter-particle pressure of compacted clays. This wok presents and discusses the prediction of inter-particle pressure of compacted heavy clay from the consideration of the Gouy–Chapman diffuse double layer theory. According to the diffuse double layer theory, the inter-particle pressure is the difference between the osmotic pressure in the central plane between two clay plates and the osmotic pressure in the equilibrium solution (Bolt, 1956 [5]). In other words, the inter-particle pressure is the pressure required to keep the clay–water system at the required void ratio when it is permitted to absorb water. Thus, in oedometer compression test, for any given external vertical pressure, the equilibrium void ratio can be calculated from the double layer theory (Tripathy et. al., 2004 [15]).

Basically, the mechanism that controlling the volume changes behavior of heavy clays is the branching between the mineral unit layers due to developed forces between the adjacent plates resulting from the overlapping between the electrical domains of the particles. Generally, this mechanism depends on soil properties represented by Base Exchange capacity, surface area, and ion type and fluid properties represented by ion type and concentration, cation valency, dielectric constant, and temperature. The interaction between solid and liquid phase gives rise to the formation of a “surface phase”, situated between solid and liquid. The properties of this surface phase are the domain of surface chemistry (Mitchell, 1993 [16]). So, physicochemical interaction between clay particles and the surrounding liquid phase leads to the existence of a repulsive force between the particles which can extend over a considerable distance. Terzaghi and Peck, 1948 (cited by Bolt, 1956 [5]) discussed the existence of such a repulsive force between clay particles, which is attributed to “an increase in the thickness of the adsorbed layers, which separate the grains”. Komine and Ogata, 1996 [9] recognized that the repulsive and attractive forces between two layers cause the swelling pressure and swelling deformation of minerals. The repulsive pressure midway between two parallel clay plates, Fig. (1, a), can be given by the langmuir equation which plotted graphically in Fig. (2,b). (Wu, 1982 [17] and Das, 1983 [18]): -

$$P_d - P_\infty = 2n_\circ kT\,(\cosh \frac{\nu\varepsilon\psi_d}{kT} - 1) \quad \ldots(1)$$

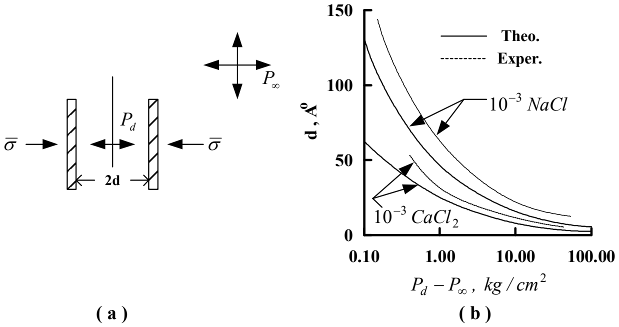

Figure 2. Repulsive pressure midway between two clay sheets (after Wu, 1982 [16])

Since P_∞ is the pressure in the water at infinite distance, where the field intensity is 0, it is the same as the hydrostatic pressure. Hence $P_d - P_\infty$ is the repulsive pressure that is in excess of the hydrostatic pressure. The clay plates therefore tend to be pushed apart by this repulsive pressure, if the plates are to be prevented from moving farther and farther apart, a pressure $P_d - P_\infty$ in excess of the hydrostatic pressure must be applied to the two particles. The effective stress in Fig. (1, a) becomes $\overline{\sigma} = P_d - P_\infty$ where the effective stress $\overline{\sigma}$ must be resisted by the repulsive pressure. Now, if the soil is subjected to external pressure, the distance between the particles will decrease and water will be squeezed out. As a result, the ion concentration between the particles will increase and the osmotic pressure P_d in turn increases, i.e., $P_d - P_\infty$ increases until it is equal to the new value of $\overline{\sigma}$. The reverse process involves the decrease of external pressure (if $\overline{\sigma}$ is reduced) and the suction of liquid by osmotic pressure between the particles to dilute the concentration of ions. The distance between the particles would increase, resulting in volume increase and a reduction of osmotic pressure, i.e., $P_d - P_\infty$ reduced. This process continues until a new equilibrium is established (Bolt, 1956 [5] and Wu, 1982 [16]).

3.1 Repulsive forces between two parallel layers

A repulsive force is generated between two parallel layers if the two layers approach each other's and their electrical potential distributions overlap (Komine and Ogata,1996 [9]).

The distribution of the electrical potential in the clay-water-elctrolyte system was represented by [Mitchel, 1993 [18]). In the diffuse double layer theory, the electrical potential distribution around the layer is given by:

$$\frac{d\phi}{dx} = -\sqrt{\frac{2nkT}{\varepsilon}}\, 2\sinh(\frac{\nu e'\phi}{2kT}) \qquad \ldots (2)$$

$$\phi_\circ = \frac{kTz}{\nu e'} \qquad \ldots \qquad (3)$$

for $$z = 2\sinh^{-1}(96.5 \times \frac{B}{S}\sqrt{\frac{1}{8\varepsilon nkT}}) \qquad \ldots (4)$$

where x is the position from the layer surface (m).

Solving (2) for conditions $\phi=\phi_\circ$ for $x = 0$, and $\phi=\phi_d$ for $x = d$, gives the

$$y_d = 4\tanh^{-1}\left[\exp(-Kd)\tanh(\frac{z}{4})\right] \quad \ldots \qquad (5)$$

for $K = \sqrt{\frac{2n\nu^2 e'^2}{\varepsilon kT}}$ (m^{-1}) $\quad \ldots \qquad (6)$

Therefore, the repulsive force per unit area between two parallel layers is given by

$$f_r = 2nkT(\cosh u - 1) \qquad (J/m^3)$$

$$= 2nkT(\cosh u - 1)\times 10^{-6}\ (\text{MPa}) \qquad \ldots(7)$$

for $$u = 2y_d = 8\tanh^{-1}\left[\exp(-Kd)\tanh(\frac{z}{4})\right] \qquad \ldots(8)$$

3.2 Attractive forces between two parallel layers

In addition to a repulsive force between the approaching clay particles, there is also a component of attractive force (f_a) between the two particles. This attractive force is the van der waals force, or secondary bonding force, which acts between all adjacent pieces of matter.

So, attractive forces act between two layers if they approach extremely close to each other [Komine and Ogata, 1996 [9]. However, one of the limitation of the Gouy-chapman double layer theory that attraction forces between the clay platelets were ignored (Sridharan and Jayadeva, 1982 [19]).

Komine and Ogata, 1996 [9] and Tripathy et. al.,2004 [15] give the attractive force per unit area as:

$$fa = \frac{A}{24\pi}\left[\frac{1}{d^3} + \frac{1}{(d+t)^3} - \frac{2}{(d+\frac{t}{2})^3}\right] \qquad (\text{J/m}^3)$$

$$= \frac{A}{24\pi}\left[\frac{1}{d^3} + \frac{1}{(d+t)^3} - \frac{2}{(d+\frac{t}{2})^3}\right]\times 10^{-6} \quad (\text{Mpa}) \qquad \ldots(9)$$

Therefore, the force P per unit area between two parallel layers can be obtained by

$$P = f_r - f_a \qquad \ldots(10)$$

4 Factors Influencing the Inter-Particle Pressure

The factors that involved in the double layer theory are:

- Soil type, i.e., base exchange capacity (B) and Surface area (S_A).
- Fluid properties, i.e., ion concentration (n), cation valence (ν), dielectric constant (ε) and temperature (T).

However, in this research two factors (the ion type and concentration) were taken into account for discussion that they seem to be the more affecting factors in the double layer theory and play a major role in inter-particle pressure values.

4.1 Ion concentration

As ion concentration increases, the inter-particle pressure decreases because the double layer is compressed. Bolt, 1956 [5] layout that as the external pressure increase, the distance between particles will decrease. As a result, the ion concentration between the particles will increase due to squeeze out of water. The opposite is true. Komine and Ogata, 1996 [9] presented a new equation for evaluating the ion concentration of pore water as:

$$n = \frac{n_\circ \times N_A}{1 + \frac{\varepsilon_{SV}^{\;*}}{100}} \quad \ldots \qquad (11)$$

Bolt, 1956 [5] showed experimentally that the concentration n_o of ions in pore water for most clays is within 10 mol/m^3 and in most cases are of about 2.5-5 mol/m^3.However, other researchers like *Komine* and *Ogata*,1996 [9] used a range of 20-40 mol/m^3 to demonstrate the effect of ion concentration of a compacted bentonite. However, for the present work, a range from 2.5-10 mol/m^3 may be agreeable.

4.2 Ion Type and Cation Valence

Ion type has a large influence on the repulsive pressure between the two layers. Its effect results from its valence value and its relation with K in equation (6). The cation valence value may vary directly with K and hence reversely with ($1/K$) which represent the thickness of the double layer (Wu, 1982 [16]; Das, 1983 [18]). However, some researchers used a value of 1.5 for cations valence to represent the Na^{+1} and Ca^{+2} cations that present in the soil. However, in the present application this value may be convincing for all samples. One of the limitations of the double layer theory that it may not give accurate predictions for bivalent electrolytes, though the theory can give accurate prediction for monovalent electrolytes.

5 Experimental Study

The soil used in this research is a heavy clay of Na-base, Table (1) illustrate the physical properties of the soil. It should be recalled here that all the tests mentioned in this work are conducted according to the procedures described in Head, 1984 [20].

In order to illustrate the ion type and concentration effects, a white powder of calcium chloride (to provide Ca^{++} cations) of purity range from 94-97 % is dissolved in a distilled water (to provide a calcium fluid solution) and mixed with soil. A 2, 4 and 8% of the total weight of oven dry mass of the soil is selected to represent a range of ion concentrations.

Table.1 Soil Properties of the soil used

Property	Value
Liquid limit, L.L, %	91
Plastic limit, P.L,%	28
Specific gravity, G.S	2.76
Swell potential	High
% Sand	5
% silt	11
% Clay	84
Max. dry density, kN/m^3	14.22
Optimum moisture content, %	25

Taylor, 1959 [6] illustrated that the material associated with the ion exchange phenomena is governed by two principle consideration. It should not contain a cation which is likely to be already present in the soil itself, and it should be the salt of a cation which is strongly adsorbed by the soil so that it will be efficient in displacing the ions already present. Thus, heavy clay with Na-base was used.

The volume change characteristics of compacted soils are predicted on samples compacted using static compaction. These samples are compacted at some chosen moisture contents to achieve the same dry densities as those determined by the dynamic compaction method. Where all samples are statically compacted to the required dry density in the oedometer ring of diameter 75-mm and a height of 14-mm (instead of 20-mm to ensure full confinement during volume change upon saturation). The entire assembly is monitored in the consolidation cell and positioned in the loading frame with a nominal surcharge of 7 kPa. In the first series of tests all the samples were compacted to a value corresponding to the dry density of 1.655 g/cm^3 at water content 20.5%. In the second series of tests the soil samples is compacted at its own maximum dry density and optimum moisture content.

6 Method of Predicting the Inter-Particle Pressure of Heavy Clay

In this research, the following new procedure for evaluating the inter-particle pressure has been proposed. For orientation purpose, the structure of a unit layer mineral can be modeled as shown in Figure (2). The Unit (two opposite layers) shown in Fig. (2,a) may undergo a volume change throughout the soaking process starting from stage I to II till reaching steady state at III. Keeping in mind that the distance between the two layers in the later stage depends on the placement condition (initial moisture content and dry density) in stage I.

Since the soil at stage III has no tendency for further volume change it may represent the datum in loading stages for determination of the inter-particle pressure, Fig. (3,b). Theoretically, this may seem not true. Since for the condition in stage III there is a pressure value which can be calculated by the double layer theory equations. Thus the load ($\Delta P_{\circ}$) in Fig. (3,b) which corresponds to the volume change ($\Delta\, d_{\circ}$) may be calculated as the difference between the theoretical pressure value for stage IV and III. The same calculation may be done with respect to V and VI. Therefore, the inter-particle pressure may be defined as the summation of the theoretical pressure, which required to reach initial height, i.e., ($P_S = \Delta P_{\circ} + \Delta P_1 + \Delta P_2 + ... + \Delta P_n$).

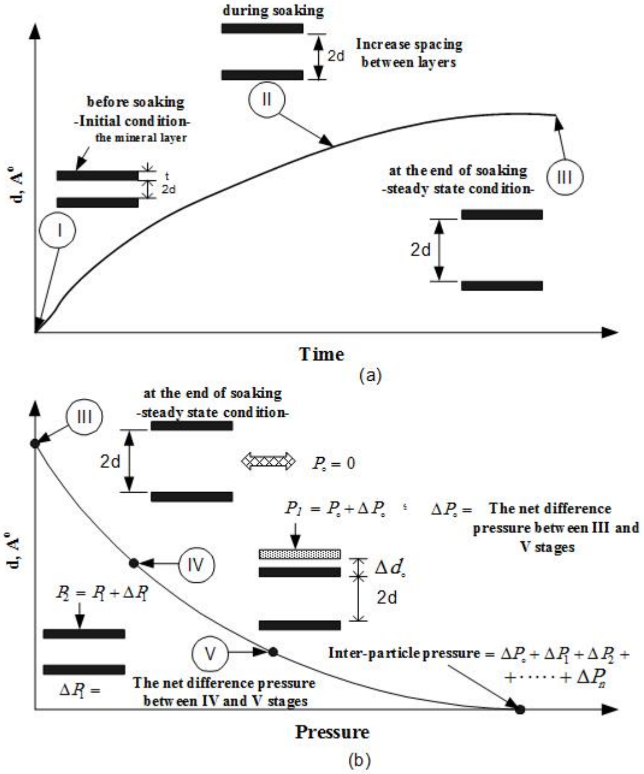

Figure 3. Proposed concept of unit layer deformation upon saturation (a) deformation with time during soaking, (b) Consolidation stage.

For simplicity, the inter-particle pressure may be obtained as the difference of the calculated pressure between the initial and final conditions, and given as: -

$$Inter - particlep\ pressure = P_i - P_f \qquad .....(12)$$

where

P_i = the calculated pressure at stage I, Fig. (3,a), using double layer theory equations.

P_f = the calculated pressure at stage III, Fig. (3,a), using double layer theory equations.

It is interesting to mention that the final condition stage III in Fig. (3,a) can be achieved using the hyperbolic equation which represent the relation between the unit layer distance with time after heavy clay saturation. Thus, the deformation and inter-particle pressure can be calculated theoretically after two days of simple laboratory testing (to extract equation constants) rather than several days (may reach 30 days in high heavy clay) of exhausting laboratory testing.

7 Applicability of the Proposed Prediction Method

The applicability of the given prediction method is investigated by comparing the predicted results with laboratory tests results on the unit layer deformation and inter-particle pressures of compacted heavy clay. In the experimental work, the conventional consolidation method was adopted to predict the inter-particle pressure.

The parameters used in theoretical analysis include the specific surface S = 119.94 m^2/g which is calculated from the correlation with liquid limit measured value. the cation exchange capacity $B = 0.525$, which is also a measured value. The ionic valence ($\nu = 1.5$) for all samples is assumed since it is difficult to obtain the value of ionic valance of available cations in the double layer and 1.5 is an equivalent expected value for the available ions. Furthermore, the Boltzmann constant k (1.38×10^{-23} J/K), Hamaker constant A_h (2.2×10^{20} J), the electric charge $\acute{e}$ (1.602×10^{19} C), the static permittivity of pore water ε ($80 \times 8.8542 \times 10^{-12}$ $C^2.J^{-1}.m^{-1}$), the thickness of unit layer t (9.60×10^{-10} m), and Avogadro's number (6.023×10^{23}) are quoted from (Sridharan and Jayadeva, 1982 [19];

Komine and Ogata, 1996 [9]; Tripathy et. al., 2004[15]).

Figures 4 to 10 show the relationship between predicted inter-particle pressure calculated by the proposed prediction method and deformation of unit layer represented by spacing between the adjacent plates (2d). Also, it illustrates the curves that obtained from the experimental work. It is clear that the curves obtained experimentally are within the range of the proposed prediction method. However, in order to investigate the generalization of this method, the same analysis described above were done on the same soil with different placement conditions, figures 8 to 10 show that the same trend may be obtained. In general, this reflects that this method of prediction is basically influenced by both, the physicochemical factors represented by ion type and concentration, and mechanical properties represented by initial dry density and moisture content.

In general, there are only limited application procedures based on the diffuse double layer theory. Komine and Ogata, 1996 [9] reported that the double layer theory is not applicable when ion concentration is very high and when the compacted soil is very dense. For instance, the prediction method proposed by Komine and Ogata, 1996 [9] and Tripathy et. al., 2004 [15] are difficult and time consuming to predict the inter-particle pressure. Also, an experiment for calculation of surface area and montmorillonite content for a given soil may be needed in their calculations, which seem to be complex and costly. Naturally, these problems may be overcome with the present proposed procedure, since the surface area may be calculated approximately as stated earlier and there is no need for montmorillonite content prediction. Also, the final conditions of soil [represented by stage III in Fig. (3,a)] are taken into consideration, this condition appears to have a great effect on the eventual inter-particle pressure values.

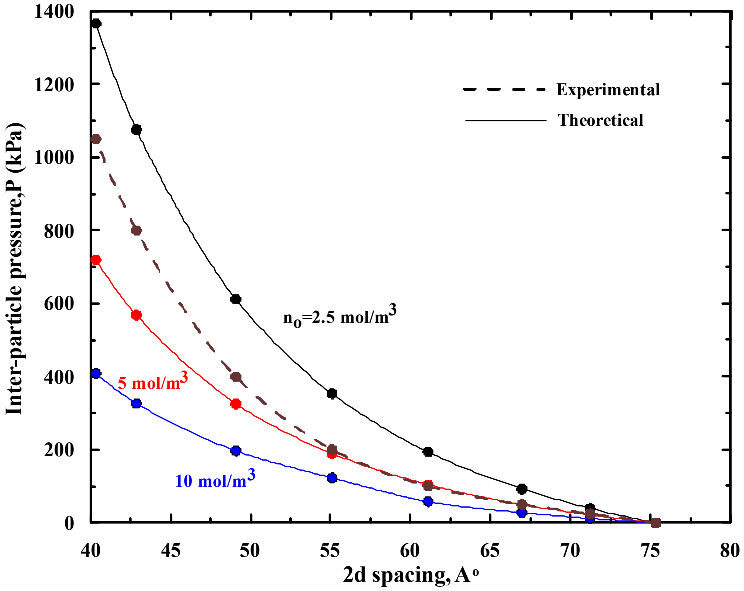

Figure 4. Inter-particle pressure relationships for different ion concentrations (initial w_c = 20.5% and γ_d = 16.23 kN/m^3)

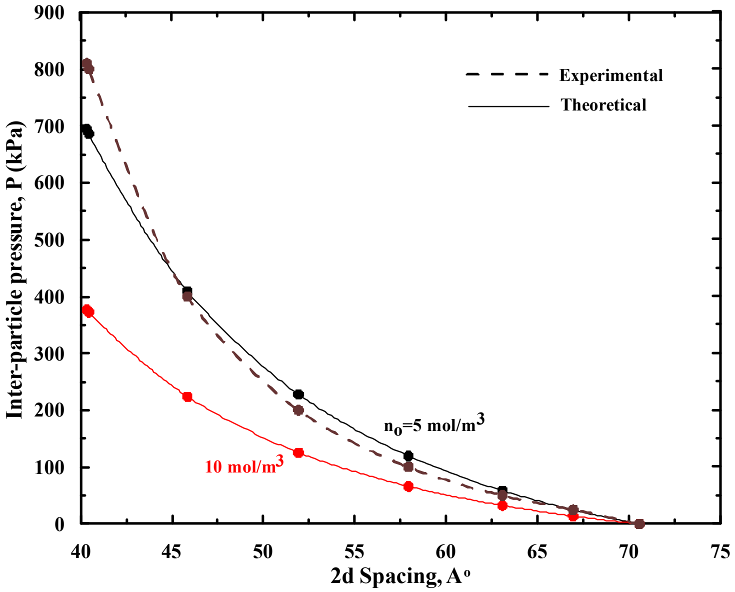

Figure 5. Inter-particle pressure relationships for different ion concentrations for soil immersed with 2% of Calcium solution (initial wc = 20.5% and γd = 16.23 kN/m^3)

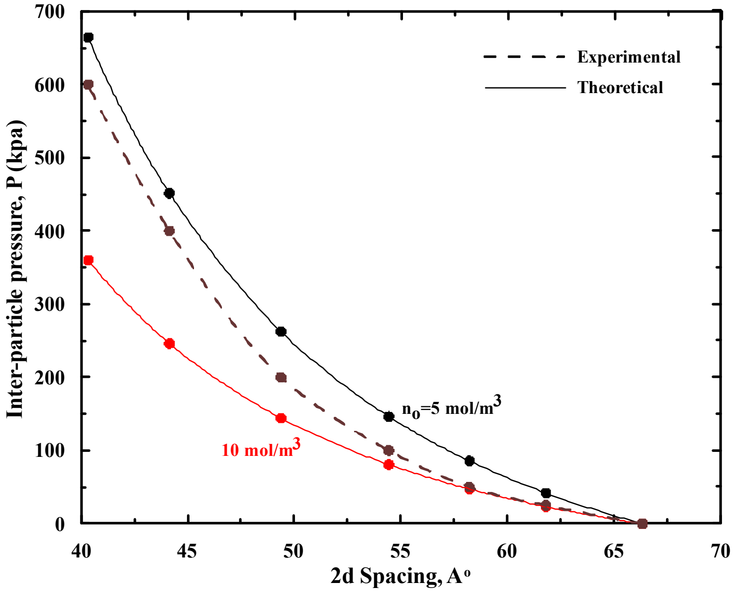

Figure 6. Inter-particle pressure relationships for different ion concentrations for soil immersed with 4% of Calcium solution (initial wc = 20.5% and γd = 16.23 kN/m^3)

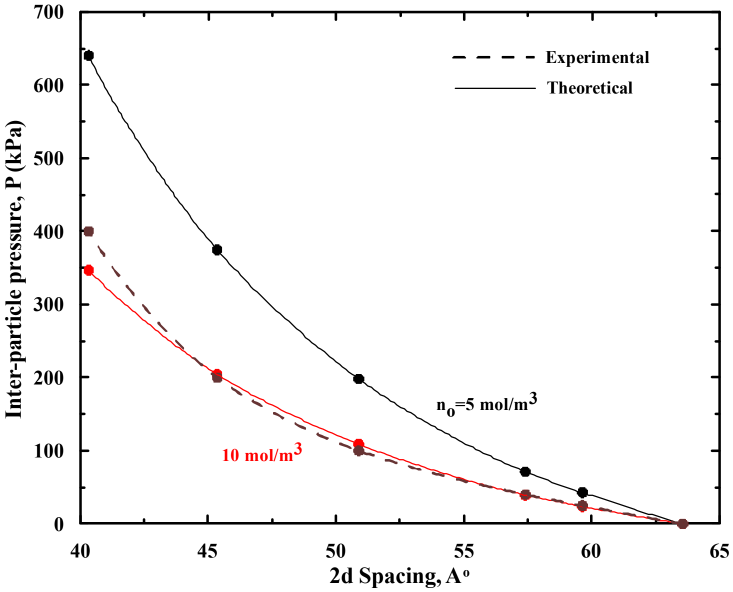

Figure 7. Inter-particle pressure relationships for different ion concentrations for soil immersed with 8% of Calcium solution (initial wc = 20.5% and γd = 16.23 kN/m^3)

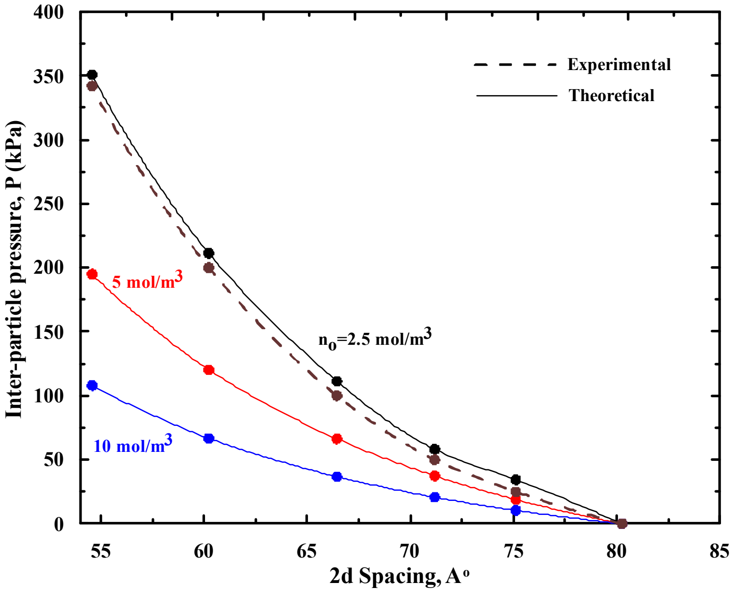

Figure 8. Inter-particle pressure relationships for different ion concentrations (initial wc = 25.1% and γd = 14.22 kN/m3)

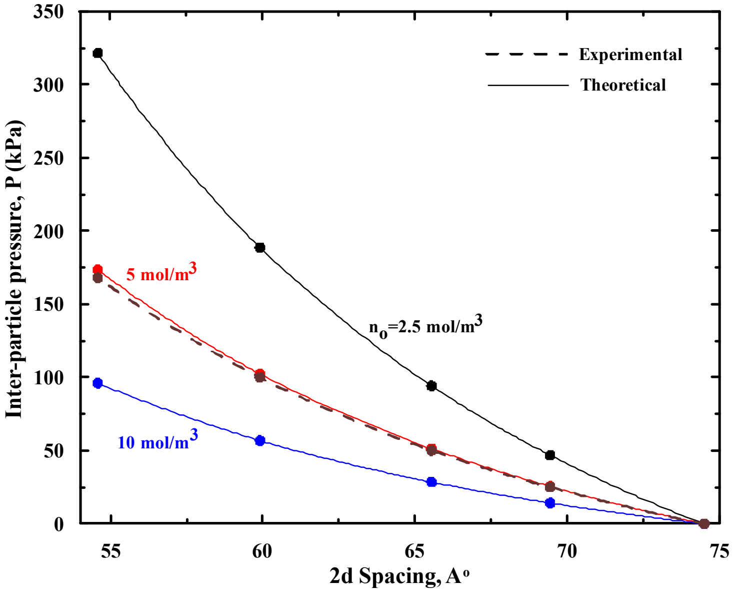

Figure 9. Inter-particle pressure relationships for different ion concentrations for soil immersed with 4% of Calcium solution (initial wc = 25.1% and γd = 14.22 kN/m3)

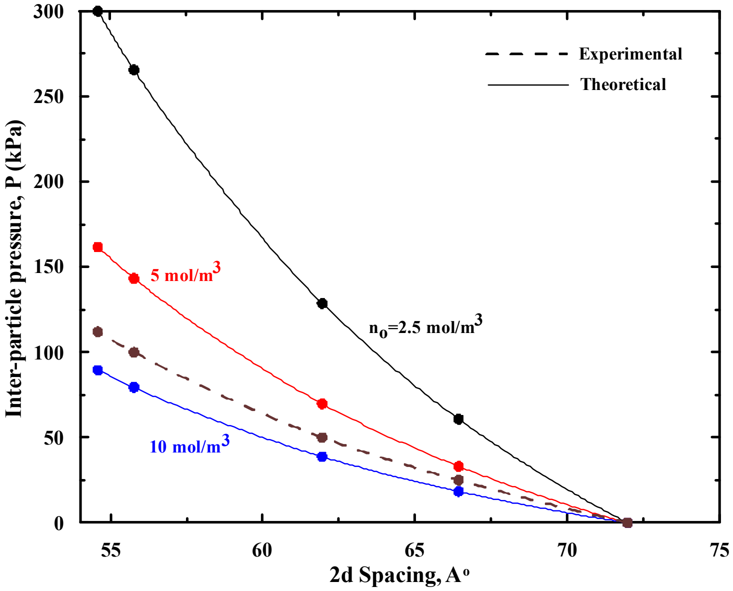

Figure 10. Inter-particle pressure relationships for different ion concentrations for soil immersed with 8% of Calcium solution (initial wc = 25.1% and γd = 14.22 kN/m3)

8 Conclusions

Experimental inter-particle pressures of heavy clay were compared with the theoretical inter-particle pressures derived from the Gouy–Chapman diffuse double layer theory. The study revealed that, in general, the experimental inter-particle pressures are consistent with their theoretical counterparts at different compaction dry densities. New procedure is proposed to predict the inter-particle pressures that taking in account the placement condition of soil represented by initial density and moisture content. The relationships between the unit layer distance and inter-particle pressure is introduced and simplified by reducing the number of required parameters of prediction that followed by other researchers. However, this new procedure overcomes the high differences between the experimental and theoretical pressures computed by double layer theory that most researchers found in their comparisons. The proposed method was verified with two different placement condition and different ion type and concentration within the water of double layer. The agreement between the inter-prticle pressures calculated using the suggested new procedure which includes new equation of the net inter-particle calculation at different stage of loading and the reported experimental data was very good. The use of the equations is based on weighted average valency with a range of 2.5 to 10 mol/m^3 of ion concentration within double layer of heavy clay, since the valency of the exchangeable cations and ion concentration present in heavy clays has a significant influence on the inter-particle pressure values. It is worth mentioning that the maximum clay deformation can be achieved using the hyperbolic equation which represent the relation between the unit layer distance with time after saturation. Thus, the deformation and inter-particle pressures can be calculated theoretically in a considerably shorter time in comparison to conventional methods.

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