Contract Report No. 3-101 THE STRENGTH OF CLAYS AT LOW EFFECTIVE STRESS

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Research in Earth Physics Phase Report No. 8

by

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January 1967

#### Sponsored by

U.S. Army Materiel Command Project No. 1-V-0-14501-B-52A-01

under

Contract No. DA-22-079-eng-457 with

U.S. Army Engineer Waterways Experiment Station CORPS OF ENGINEERS Vicksburg, Mississippi

> Soil Mechanics Division Department of Civil Engineering Massachusetts Institute of Technology

#### Research Report R67-4

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#### **II. INTERPARTICLE FORCES**

Knowledge of clay mineralogy is a prerequisite for the study of the physico-chemical or interparticle forces that act among clay particles. The mineralogical composition of clays has been discussed extensively in the literature. Grim (1953) and (1962), and Lambe and Martin (1953-1957) are some of many comprehensive references in this discipline. It is not the intent of this report to repeat what has already been written in this field. For the sake of continuity, however, a brief summary of clay mineralogy is contained herein.

In general most clay minerals may be viewed as producing particles having a plate shape. The principal building blocks of these minerals are two dimensional arrays of siliconoxygen tetrahedra and two dimensional structures of aluminum or magnesium-oxygen-hydroxyl octahedra. For example, the clay mineral kaolinite is a two layer silicate composed of one tetrahedral sheet and one aluminum octahedral sheet, gibbsite. Illite and montmorillonite are both three layer minerals composed of two silica sheets and one octahedral sheet.

Isomorphous substitution, the proxying of one atom for another, occurs in most clay minerals to varying degrees and usually results in a net negative charge on the unit crystal. This charge is manifested on the broad faces of the crystals. Electrical neutrality must be achieved by these unit layers. In fulfilling this requirement, diffuse double layers composed of exchangeable cations form around the particle faces. These double layers have been observed to exert a profound influence on soil behavior. For example, Bailey (1965) presents a discussion showing the significant

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influence which double layer forces have on the consolidation behavior of the clay minerals montmorillonite and illite. Figure 3 illustrates the relative particle sizes of three clay minerals, montmorillonite, illite, and kaolinite. Additional mineralogical information is shown in this figure. The approximate double layer size is also shown assuming univalent ions are present.

Electrical charges also exist on the edges of clay particles due to unsatisfied or broken bonds resulting from termination of the crystal structure. Bailey (1965) summarizes data which show that this edge charge has a very significant effect on the consolidation behavior of kaolinite. The edge charge is usually positive but can become neutral, at slightly above pH = 7.0, or negative in a more basic environment. See Schofield and Samson (1953, 1954) for a detailed discussion of this topic. As shown in Figure 3, double layers also form at the edges of the particles. Anions or cations, as appropriate, make up the exchangeable ions in these double layers.

In summary, it is imperative that the inherent electrical nature of the clay minerals be considered during any study devoted to understanding the fundamental behavior of clay soils.

#### A. Physico-Chemical Effective Stress Equation

In this study we are concerned with investigating the physico-chemical forces that produce cohesion in soils. The physico-chemical effective stress equation is a convenient tool with which to introduce and discuss these phenomena.

Lambe (1958a) provided stimulus to the concept of

viewing effective stress in clays as a phenomenon derived from the electrical properties of the clay-water system. The physico-chemical effective stress equation resulting from this idea is:

$$\overline{\sigma} = \left(\overline{\overline{\sigma}}_{r} - \overline{\overline{\sigma}}_{a}\right) a_{m} + (\overline{R} - \overline{A})$$
(3)

where:

- o = normal effective stress acting between clay
  particles
- $\overline{\overline{\sigma}}_{r}$  = average contact repulsive stress
- $\bar{\bar{\sigma}}_{s}$  = average contact attractive stress
- $\overline{R}$  = long range double layer repulsive stress
- $\overline{A}$  = long range attractive stress
- a<sub>m</sub> = ratio of the area between the two clay particles where their interparticle spacing is less than 20 Å to the total area between the particles

The original form of this equation was proposed by Lambe (1958b) and later modified by Lambe (1960) and Ladd (1961). A discussion of the evolution of this equation is presented by Bailey (1965). As implied in the definition of the terms in equation 3, two clay particles are considered to be in contact where their interparticle spacing is equal to or less than 20 Å. This distance, which was chosen to divide the contact or short range forces from the long range forces, was proposed by Ladd (1961). The choice of this value is discussed by Bailey (1965).

We shall now consider the various known interparticle forces which combine to form the terms of equation 3. These are listed in Table 1. As with clay mineralogy, several of these physico-chemical forces have been discussed extensively

in the literature. Consequently, where appropriate, references will be cited and a minimal discussion presented. The purpose of the following discussion is two fold: 1) to present principles upon which to later base a hypothesis for cohesion in clays, 2) to point out factors which can alter the magnitude of these interparticle forces.

#### B. Long Range Stresses

#### 1. Double Layer Osmotic Repulsion

Osmotic repulsion resulting from the interaction of similarily charged double layers is believed to be the only significant repulsive force acting over an interparticle spacing of greater than about 20 Å. Verwey and Overbeek (1948), Bolt and Peech (1953), Ladd (1961), van Olphen (1963), and others have discussed the double layer. Bailey (1965) has summarized much of this work. He lists the equations used in computing  $P_y$ , the double layer repulsion existing at the centerline between two parallel flat plates of like charge and constant surface charge density. He also presents plots of the results of  $P_y$  computations for certain idealized soil-water systems.

From these equations and Bailey's computed solutions, it is readily seen, for a given interparticle spacing, that P<sub>y</sub> will increase with any of the following: decrease in pore fluid salt concentration, replacement of the double layer's cations by cations of lower valency, and increase in the pore fluid's dielectric constant. The double layers surrounding each particle will expand following any of these changes in environment. Therefore, P<sub>y</sub> will increase for a fixed spacing since the similarily charged double layers between the particles will interact more strongly as they

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try to expand. Conversely, for a given spacing,  $P_y$ , and hence  $\overline{R}$ , will decrease if opposite changes in environment are made. Obviously, adjustments in interparticle spacings will accompany changes in  $P_y$  if the particles are not maintained at a fixed spacing by some external force. For example, two parallel particles will move apart until a new equilibrium position is reached if sodium ions replace calcium ions in the face double layers.

General trends in double layer behavior are thought to be independent of particle orientation. This statement is generally valid with respect to long range stresses since the interparticle spacings under consideration are large enough to permit reasonably complete double layer development. Meaningful computations for double layer repulsion are not possible for non-parallel particles. Nevertheless, for the same environmental change, we should expect variations in  $\overline{R}$  to be similar to those for parallel particles.

#### 2. Long Range Attractive Stresses

Bailey (1965) and Martin (1958) present discussions of the three components of the van der Waals attractive forces. Of these three components, orientation, induction, and dispersion, only the dispersion effect is considered to be of sufficient magnitude to warrant consideration in a clay-fluid system. Van der Waals forces are not considered to vary with changes in environment, but because they are dependent upon interparticle spacing they are stress dependent. The equation for the attractive pressure,  $P_A$ , between two parallel infinite flat plates at a spacing of 2d is:

$$P_{A} = \frac{A'}{48\pi} \left[ \frac{1}{d^{3}} + \frac{1}{(d+\delta)^{3}} - \frac{2}{(d+\delta)^{3}} \right]$$
(4)

where  $\delta$  is the thickness of the plates, and A' is a constant of unknown magnitude although a value of  $10^{-12}$  ergs is commonly quoted. Bolt (1956), Ladd (1961), and Bailey (1965) present graphs of P<sub>A</sub> versus interparticle spacing for parallel infinite flat plates. Ladd (1961) also presents equations for the van der Waals attraction between two equal diameter spheres and between a sphere and a half space. In a soil system having a random fabric, no equations are applicable and we are forced to make qualitative remarks with respect to how  $\overline{A}$  varies among its particles as the void ratio changes. Nevertheless, from the existing equations for van der Waals attraction, we can predict that  $\overline{A}$  will increase as the interparticle spacing decreases. Conversely, as two particles move apart, their long range attractive stresses will decrease.

#### C. Short Range Contact Stresses

Ladd (1961) and Bailey (1965) devote considerable effort to study of the short range forces listed in Table 1. The reader is referred to these references for a comprehensive treatment of these factors.

#### 1. Repulsive Stresses

If two particles are in actual mineral to mineral contact, a repulsive stress results from Born repulsion. This repulsion exists simply because two objects can not occupy the same space at the same time.

Osmotic repulsion can also exist at interparticle spacings of less than 20 Å. About thirty angstroms is generally considered to be the minimum distance required for the complete development of the classical Gouy-Chapman double layer. For

interparticle spacings of 10 - 20 Å, the Donnan double layer theory has been proposed. Even though the accuracy of the equation for this theory has been questioned (Bailey 1965), this short range osmotic repulsion should be a function of the valency of the ions between the particles, the bulk fluid salt concentration, and the medium's dielectric constant. Therefore, it is reasonable to believe that changes of long and short range osmotic repulsion will at least be in the same direction for a given environmental change.

The exact mechanisms by which surface hydration occurs and the nature of the resulting adsorbed water are not yet clearly defined, Martin (1960) and Ladd (1961). Present information suggests that surface hydration results from hydrogen bonding, van der Waals forces and electrostatic attraction between the clay particle and hydrated cations and water molecules. Data exist which suggest that hundreds to thousands of atmospheres of pressure are required to force out adsorbed water from between parallel particles of a moist clay. For a random soil fabric where edges of particles are pushed up against the faces of others, the repulsive stresses exerted by the adsorbed water to prevent mineral to mineral contact are undoubtedly changed. Unfortunately, the direction and magnitude of this change are unknown. For the present, therefore, we can only use existing data and make rough estimates of this phenomenon's magnitude. Surface hydration is probably independent of salt concentration.

As with surface hydration, many questions remain unanswered concerning ion hydration and the extent of its contribution to the repulsive stresses between clay particles. Until further light is shed on this factor we can only

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acknowledge its existence and estimate its magnitude.

Repulsive stresses due to orientation of water molecules in an electric field need not be considered at the centerline between two parallel clay particles of the same charge because the electric field strength at that point is zero. However, an electric field will exist between the positive edge and negative face of two particles which are in a flocculated array such as illustrated by Figure 4. Ladd (1961) presents three equations which might be used to estimate the repulsive stress between the particles due to the orientation of water molecules in this area. One equation uses the electric field strength, E, existing between a pair of ions. The second can be used if pairs of parallel infinite flat plates of opposite charge are considered. The third considers the E existing in the neighborhood of an electrically charged colloidal particle. All three equations have the form:

Repulsive pressure = 
$$P_{ER} = \frac{bE^2}{v_o}$$
 (5)

where b is a function of the pore fluid and temperature, and  $\overline{v}_{o}$  is the partial molar volume of the pore fluid.

Regardless of the equation chosen for computation, E will decrease for an increase in the pore fluid dielectric constant,  $\varepsilon$ . It is not clear which of the aforementioned equations is most applicable to the area between two flocculated particles. It should be noted that any significant change in the  $\frac{b}{\overline{v}}$  term of equation (5) can be expected to have an effect  $\overline{v}$  on the magnitude of  $P_{ER}$ .

2. Attractive Stresses

The van der Waals dispersion component discussed

previously for long range stresses is still operative at shorter distances. This factor will not be considered further. . .

Electrostatic attraction between the negative face and positive edge of two clay particles can occur if they are in a flocculated orientation. This electrostatic attraction is obviously dependent on the existence of oppositely charged surfaces. Consequently, any factor changing the potential between the surfaces will also alter the magnitude of this attraction. The magnitude of this interparticle force is inversely proportional to the dielectric constant of the medium surrounding the particles. Furthermore, the attractive stresses caused by this force should increase for decreased particle spacing and vice versa.

Primary valence bonding will be very strong if it occurs between clay particles. It is probably akin to the primary valence bonding occurring at points of contact in metals and the crystalline growth hypothesized by the Terzaghi-Bowden-Tabor Theory of Friction.

Ionic lattice attraction is a form of "cation linkage" in which the cations between parallel clay particles are hypothesized to form a rectangular crystalline arrangement with each cation remaining in its particular cell. Ladd (1961) presents equations for this phenomenon. Since ionic lattice attraction only occurs for parallel clay particles at a spacing of less than about 10 Å, its incidence in a typical soil-water system of interest to the engineer is not significant enough to warrant further consideration.

The last short range stress to be discussed is cementation. The partial cementing of some natural clays due to carbonates, iron oxides, and possibly organic matter is well

known. Such cementing should not be a significant factor if the strength of remolded clay is being studied. Cementation of this type will not be considered further since remolded clays were employed in this research program. Cementation resulting from the slight solubility of clay particles is possible in most soil-water systems and will be considered when the results of this research program are discussed.

#### D. Conclusion

The discussion of interparticle forces was necessarily very brief. No computations were made since accurate calculations for most of these physico-chemical forces are only possible under idealized or highly restricted conditions. Therefore, these interparticle forces will be considered in a qualitative manner when the hypothesis for cohesion is formulated and discussed.

#### III. <u>MECHANISMS OF STRENGTH GENERATION AND</u> HYPOTHESIS FOR COHESION IN CLAYS

#### Strength Generation in Clays: General

Ladd (1961) discusses the possible mechanisms for shearing resistance in clays. These may be summarized as follows:

- Sliding between surfaces that on a molecular scale are considered smooth. Possible sources of resistance resulting from these interparticle movements are:
  - a. Adhesion due to primary valence bonding -Terzaghi-Bosden-Tabor friction\*
  - b. Some sort of water structure that may exist within the thin "film" of adsorbed water near clay particle surfaces.
  - c. Natural cementation
- Reorientation of particles during shear. This reorientation generally results in an increased "closest" particle spacing and breaking of contacts and requires that energy be applied to the soil system.
- 3. Sliding between rough surfaces. The faces of clay particles are frequently not perfectly flat but possess raised plateaus called terraces. These

Bromwell (1966) discusses this theory in detail.

terraces can be considered as leading to rough surfaces.

4. Resistance due to interaction of the double layers of the particles involved.

Items 1, 2, and 3 are illustrated in Figure 5.

The frictional character of item la is obvious. Terzaghi-Bowden-Tabor friction will certainly make a significant contribution to shear strength if it occurs among a large percentage of a soil's particles.

From the previous section it will be recalled that the exact nature of adsorbed water is still unknown. Two views exist, at the present time, with respect to its nature, (Martin, 1960). One group of researchers sees this water as having a more ordered structure than that of normal bulk water. This ordered "water" is believed to have a viscosity high enough to resist applied shear stresses. Thus, a particle moving through it, such as the top particle in Figure 5 (a), will encounter a "frictional" resistance much higher than the normal viscous resistance of bulk water. The second group views adsorbed water as having two degrees of freedom on the surface of the clay particle. In this case, the water can be pushed aside easily as one particle slides across the surface of another. Hence, this type of adsorbed water will not offer any frictional resistance to shear.

If cementation should be operative, the contacts of the soil will exhibit an indigenous strength. This contact strength will contribute to shear strength since energy must be applied to break the cementation. In addition to natural cementation, a contact will sometimes exhibit a strength which is independent of any friction resulting from

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applied effective stresses. The energy necessary to overcome this contact strength can be considered a measure of the contact's cohesive strength.

Rotation of particles as illustrated in Figure 5(b)(1) is frictional in that during rotation, adjacent particles bump into and slide past each other. This resistance might be considered analogous to the interference which develops among the grains of a sand when sheared. This could also be generated by a combination of the items illustrated in Figures 5(a) and 5(b)(2).

The behavior shown in Figure 5(c) is similar to interference in sands. Thus, it does not involve new interparticle forces since it actually magnifies the effects of Terzaghi-Bowden-Tabor friction.

Even though the shear strength of a soil is considered to be primarily dependent on contact forces, the importance of long range double layer forces, item 4, must not be under estimated. The double layer plays a key role in determining the ease with which contacts are remade during shear and influences the amount of effective stress that can be carried by the contacts. If expanded double layers exist in the soil, then it will be difficult for particles to remake contacts. This condition is illustrated in Figure 6(a). Conversely, in Figure 6(b), the clay particles surrounded by depressed double layers are seen to be more able to remake contacts. Thus, the potential magnitude of items one through three is seen to be dependent on double layer characteristics. By considering equation (3), it is seen that for constant effective stress, an increase in the long range stress term,  $(\overline{R} - \overline{A})$ , leads to a reduction in the contact stresses, and vice versa.

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The development of excess pore pressures and volume changes is seen to be dependent on contact behavior and double layer characteristics. In the great majority of normally consolidated, N.C., clays the ratio

$$(\overline{\overline{\sigma}_r} - \overline{\overline{\sigma}_a})a_m$$

is believed to be close to unity initially (Ladd, 1961 Bailey, 1965). During shear, net contact breakage occurs, its extent being dependent on double layer characteristics. Very little, if any, of the effective stress lost at the contacts can be taken up by the  $(\overline{R} - \overline{A})$  term of equation (3), since the double layer forces are too small. Hence, positive excess pore pressures develop to resist the applied total stresses. Likewise, volume decreases will occur during drained tests on N.C. clays, since interparticle spacings must decrease in order for enough contacts to be remade so that equilibrium with the applied stresses is maintained.

The development of negative excess pore pressures in heavily overconsolidated clays can be visualized by considering the requirement for no volume change during undrained shear. Initially, the  $(\overline{\sigma}_r - \overline{\sigma}_a)a_m$  term of equation (3) might be negative and the long range stress term positive. As contacts are broken during shear, the double layer stresses will try to dilate the sample. Negative excess pore pressures will develop to counteract this tendency. On the other hand, if drainage is permitted dilation occurs. Alternatively, the pore pressure-volume change behavior might be explained simply in terms of geometric interference as is done for sands.

### B. The Nature of Cohesion

Let us now look at the factors which can lead to the development of cohesion in clays. In this discussion, natural cementation will be neglected because this item only augments the behavior to be considered. For simplicity, the following discussion considers only two clay particles. The principles discussed for them can then be applied to a clay soil as a whole.

Cohesion is hypothesized to exist between two particles if when the applied effective stresses are zero, energy must be expended in order to break their contact. This energy must be applied to overcome net internal attractive stresses acting between them wherein:

$$(\overline{\overline{\sigma}}_{a})a_{m} + \overline{A} > (\overline{\overline{\sigma}}_{r})a_{m} + \overline{R}$$
 (6)

It is important to note that equation (6), of necessity, considers both long and short range stresses. As an analogy, consider the attractive stresses between a magnet and a piece of steel held initially at some oblique angle with a 2d spacing less than 20 Å. Obviously, the magnetic attraction exerted on the steel extends beyond a 20 Å distance. Similarly, the long range stresses  $\overline{R}$  and  $\overline{A}$ , even if of small magnitude, should not be neglected when considering the cohesion developed between two skewed clay particles.

The definition for cohesion between two clay particles is general and not limited to a condition where the particles are at some oblique angle. For example, ionic lattice attraction and van der Waals forces can cause cohesion between two parallel clay particles at a small 2d spacing, say 3 - 6 Å.

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The formation of domains in montmorillonite is one example, Ladd (1961). For this condition  $\overline{A}$  and  $\overline{R}$  of equation (6) are zero and, initially at least, we are only left with short range interparticle forces.

Figure 7 presents a hypothetical relationship of how the energy might vary with spacing between two clay particles at some oblique orientation. The particles are assumed to be weightless so that gravity forces need not be considered. For purposes of this figure the applied effective stresses are zero. Thus, the relationship shown represents the energy which exists between the particles at various spacings, solely as a result of the several physico-chemical forces. Between points (1) and (4) a net attraction exists between the particles and energy must be applied to break their contact. To the right of point (4) a net repulsive energy exists between them, and work must be put into the system to form a contact.

To more fully understand the concepts of interparticle energy states, consider the idealized cases shown in Figure 8. Figure 8a illustrates the energy-distance relationship which might exist between two ideally dispersed weightless particles. For simplicity, it is assumed in this figure that cation linkage, van der Waals forces, and primary valency bonding do not generate a net attractive energy. Under these assumed conditions, if the two particles originally at distance (1) are pushed closer together, a net repulsion develops between them and continues to increase as the 2d spacing decreases. Finally, at a 2d spacing of 0 Å, infinite repulsion is caused by Born repulsion. If the external effective stress required to push the particles to spacing (2) were withdrawn, the repulsive energy at this point would cause the particles to move away from each

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other until equilibrium position (1) was reached. At this point they would no longer be considered in contact. The positive slope of the curve in Figure 8a shows that the rate of increase of repulsive stresses is greater than that of the attractive stresses for a given reduction in 2d spacing.

Figure 8b represents an idealized flocculated system for which gravity forces are neglected and all repulsive forces, exclusive of Born repulsion, are assumed to be so small that a net attraction always exists between states (1) and (2). As before, the energy relationship shown is assumed to be independent of external effective stresses. If the two particles originally at equilibrium position (1) were jarred or nudged in some manner so that their interparticle spacing decreased by an infinitesimal amount, then the net attractive energy existing at all points between (1) and (2) would cause a spontaneous reduction in interparticle spacings until state (2) was reached. At (2) Born repulsion could develop due to the impenetrability of matter. Point (2) illustrates the phenomenon of a potential well, which in this case represents a minimum energy state. We have seen, that of their own accord, the particles will spontaneously move from any state (3) between (1) and (2) to state (2). Likewise, if the external energy necessary to bring the particles to a state (4), at any 2d spacing to the left of (2) were released, the particles would automatically move to the potential well (2). The vertical distance (a) for distance (3) represents the energy which must be applied to the soil system at that distance, to overcome the net attractive energy and permanently break the contact between the two particles. The negative slope between states (1) and (2) corresponds to the fact that the rate of increase in attractive stresses is greater than that of the repulsive

stresses for a given reduction in 2d spacing. The positive slope to the left of state (2) reflects the preponderant Born repulsive stress.

Two potential wells are shown in Figure 7, distances (1) and (3). It has been shown that meaningful calculations for most interparticle forces are only possible under idealized conditions. Hence, this figure is highly speculative and mainly serves to illustrate what might occur. Therefore, the inclusion of two energy wells in this figure does not preclude the possibility of a larger or smaller number of them. The well at distance (3) may not actually exist. Since Born repulsion will eventually come into play when the particles reach small enough spacings, it is reasonable to expect that a well corresponding to distance (1) should exist.

This figure shows the approximate ranges over which the various known interparticle forces might act. For a 2d spacing greater than 20 Å only long range forces exist with the double layer repulsion predominating at low salt concentrations. In zone A osmotic repulsion is the ascendant force. Donnan repulsion exists here for the reasons discussed in Section II. Plus to minus electrostatic attraction might be most influential in zone B. In zone C repulsive forces might increase at a faster rate than the attractive stresses because of the physical interaction of hydrated ions and adsorbed water associated with the two particles. The amount of cohesion, therefore, decreases. Once some of the hydrated ions and adsorbed water are pushed aside, attractive forces might take over and inter-particle Primary valence and van der Waals cohesion increases. forces might be the prevailing influences for interparticle spacings in zone D. At a distance of less than

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1 A or so, Born repulsion will develop to tremendous values if the particles are forced together by external forces.

The net attractive energy for the three states (1), (2), and (3) is represented by the distances (a), (b), and (c) respectively. Consider now two particles represented by distance (1). If we could apply and release energy at will, it would be possible, with an input of energy equal to a - b, to bring them to distance (2). The particles are in a metastable position at this point. Thus, while they might stay at (2), they could also "slip back" to state (1) or fall into the potential well at state (3). Thus, after the input of energy three possible values for cohesion between the particles exist, making it difficult to determine the original value for state (1).

From the discussion of energy states two conclusions can be drawn. Firstly, there is no such thing as "the true cohesion" between particles, since cohesion can vary with interparticle spacing. Secondly, if an accurate determination of cohesion is desired, the energy input required to permanently overcome the predominant attractive stresses causing this phenomenon must be measured.

#### C. Factors Effecting Cohesion

In the previous section, a hypothesis for cohesion in clays was presented. We saw how cohesion arises from the various physico-chemical forces existing between adjacent clay particles, and how the magnitude of cohesion will vary with interparticle spacing due to competition among these forces. We can thus visualize how the cohesion of a soil might be stress dependent. Increasing the consolidation pressure will cause interparticle movements, a decreased water content, reduced interparticle spacings, most

importantly an increase in the number of contacts and probably an increase in cohesion.

In Section II, the environmental factors which can affect the magnitudes of several of the known interparticle forces were considered. These factors, salt concentration of the bulk solution, valency of double layer ions, pore fluid dielectric constant, and pH will now be discussed with respect to how the cohesion of a soil can vary. Those interparticle forces affected by environment are listed in Table 2.

The cohesion of a clay can be decreased by increasing the interparticle repulsive stresses. From the principles of Section II, it follows that at a given interparticle spacing, the osmotic repulsion may be increased by decreasing the salt concentration of the bulk solution, by replacing the double layer ions with ions of lower valency, or by increasing the dielectric constant of the pore fluid. As the double layers expand for two particles such as in Figure 4 (a), their similarly charged double layers interact more strongly and the repulsion between them increases. If not prohibited by the application of an external force, this increased repulsion could lead to a breakage of their contact, increased spacing as shown in Figure 4 (b), and a loss of cohesion. Since Donnan repulsion follows double layer behavior, the short range osmotic repulsion will increase for the same environmental changes. Obviously, a transfer of Donnan repulsion into long range double layer repulsion occurs if the contact between two particles is broken and their spacing increases. With respect to osmotic forces, the cohesion of a clay can develop or increase following a reversal of these environmental factors. At a given interparticle spacing the attractive stresses will be constant. Hence, a decrease in the osmotic repulsive stresses

can lead to or augment the inequality expressed in equation (6).

The cohesive strength of a soil such as kaolinite, where the edge effects predominate, should decrease for an increase in pore fluid pH since the electrostatic attraction decreases. This follows from the fact that the edge charge should become neutral in a slightly alkaline environment and negative as the pore fluid becomes highly basic. Thus, the positive charge necessary for this attraction is eliminated or reduced in magnitude. The electrostatic attraction varies accordingly, and a reduction in cohesion should follow. Conversely, an increase in cohesive strength is predicted from a decrease in pore fluid pH.

It should also be possible to vary the electrostatic attraction and thus, cohesion, for a soil such as kaolinite, by the addition to the pore fluid of so called dispersants and salts. A dispersant's negatively charged radicals and a salt's anions should migrate to the particle edges and at least partially neutralize their positive charges. The edge to face attraction and hence cohesion should then drop. If the valency of the anions in the pore fluid was increased, the cohesion of kaolinite might drop further due to a more effective neutralization of the particle edge charge.

In Section II the electrostatic attraction between flocculated particles was stated to be inversely proportional to the dielectric constant of the pore fluid. Therefore, the cohesive strength of a soil where edge effects predominate, might be expected to decrease for an increase in this property. Conversely, in a soil of this type, cohesion might be expected to increase following a decrease in  $\varepsilon$ , since  $(\overline{\sigma}_a)a_m$  should

1,

increase.

However, the situation is not necessarily this simple. One way to vary the dielectric constant is to replace the water in the soil system with some other fluid.\* While  $\varepsilon$ may change the electrostatic attraction as predicted, other properties of the new pore fluid must now be considered. For example, if the  $\frac{b}{b}$  term of equation (5) changes, then  $\overline{v}_{o}$  the molecules of the new fluid in the orientation of an electric field might vary and lead to different repulsive stresses between the particles. The cohesion would in turn be effected. Furthermore, from Section II it is recalled that a change in  $\varepsilon$  can also effect the magnitude of the electric field strength term of equation (5). As noted previously no equation now exists with which to calculate the electric field strength between two flocculated particles. In view of these complications, it is difficult to make predictions with respect to how a change in dielectric constant, which accompanies a different pore fluid, might effect the cohesive strength of a clay.

Bulk fluid salt concentration and ion valency have been listed in Table 2 as factors possibly effecting the repulsive force arising from orientation of molecules in an electric field. It is interesting to note that if these factors do effect this repulsive stress, their effect on orientation repulsion is similar to that with osmotic repulsion. For example, increased ion valency leads to decreased osmotic and orientation repulsion, thus, increasing cohesion.

A hypothesis for cohesion in clays has now been formulated. Predictions have been made with respect to the

The dielectric constant of the pore fluid could be changed by varying the temperature, although this environmental factor is not considered herein.

dependence of cohesion on environment. We have also seen that the cohesion of a soil should be dependent upon stress history. The validity of these predictions and the hypothesis for cohesion in clays must be verified by an experimental program.

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#### TABLE I

#### Types of Interparticle Forces

- I. Long Range (Interparticle spacing of greater than 20  $\mathring{A}$ )
  - A. Repulsive (Double layer osmotic repulsion)
  - B. Attractive (Van der Waals dispersion component)
- - A. Repulsive
    - 1. Osmotic or Donnan repulsion
    - 2. Surface hydration
    - 3. Ion hydration
    - 4. Orientation of water in an electric field
    - 5. Born repulsion (if actual mineral to mineral contact occurs)

#### B. Attractive

- 1. Van der Waals dispersion component
- 2. Edge to face electrostatic attraction
- 3. Primary valence bonding (if actual mineral to mineral contact occurs)
- Ionic lattice attraction, cation linkage (only for parallel particles)
- 5. Cementation

#### TABLE 2

#### Interparticle Forces as Influenced by Environment

Osmotic Repulsion (Long and Short Range)

Bulk Fluid Salt Concentration Dielectric Constant Valency of Double Layer Ions

Edge to Face Electrostatic Attraction

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Bulk Fluid Salt Concentration Valency of Double Layer Ions Dielectric Constant pH

Orientation of Molecules in an Electric Field

Dielectric Constant Bulk Fluid Salt Concentration? Valency of Double Layer Ions?

## SIZE, SHAPE, AND EXTENT OF DOUBLE LAYER FOR THREE CLAY MINERALS

#### I. Na MONTMORILLONITE





2. NO ILLITE



#### 3. NO KAOLINITE



AFTER LADD(1961) FIGURE

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 $2d_b$   $2d_b > 2d_a$ 

(b)

# HYPOTHETICAL CONTACT BEHAVIOR

DURING SHEAR

.



(a) SLIDING AT CONTACTS



(b) REORIENTATION OF PARTICLES AND BREAKING OF CONTACTS

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FIGURE 5

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(C) SLIDING BETWEEN ROUGH SURFACES

## INFLUENCE OF THE DOUBLE LAYER ON SHEARING RESISTANCE IN CLAYS



(a) EXPANDED DOUBLE LAYERS - DIFFICULT TO REMAKE CONTACTS



(b) DEPRESSED DOUBLE LAYERS - CONTACTS REMADE EASILY

FIGURE 6 . '.





FIGURE 8