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ABSTRACT

Mechanical behaviour of high-plastic clay soils is strongly influenced by physicochemical interactions between clay particles and pore water. Tests have measured osmotic suctions and strengths in unsaturated dense sand-bentonite 'buffer' prepared at constant densities but with different NaCl concentrations. Osmotic suctions and matric suctions produce opposing changes in strength.

RÉSUMÉ

Le compartement mécanique des sols d'argile de grande plasticité est fortement influencé par les interactions physicochimiques entre les particules d'argile et l'eau interstitielle. Des tests ont mesuré les succions osmotiques initiales et les résistances en sable-bentonite denses et non-saturés préparés à des densités constantes mais avec différentes concentrations en NaCl. Les succions osmotiques et matricielles agissent en directions opposées.

INTRODUCTION

Negative potentials (soil suctions) strongly influence the mechanical behaviour of unsaturated soils. Osmotic suction is closely related to the salt content in the pore-water, whereas matric suction is mainly associated with air-water interfaces and adsorption forces. In projects involving unsaturated silts and low-plastic clays, matric suction changes can be substituted for total suction changes, and *vice versa* (Fredlund and Rahardjo 1993). Wan *et al.* (1995) show that compared with matric suctions, osmotic suctions vary only slowly with water content in high plastic clays.

However osmotic suctions can be expected to change with ground water chemistry or contamination. Knowledge of the influence of osmotic suction is therefore needed.

Relatively little attention has been paid to how osmotic suction influences mechanical and hydraulic properties of unsaturated soils. With increasing osmotic suction, strengths have been reported as increasing (Ho and Pufahl 1987), remaining essentially constant (Blight 1983), and decreasing (Yong and Warkentin 1975). However, conditions were not identical in the three sets of tests.

In its proposal for safe underground disposal of nuclear fuel waste, Atomic Energy of Canada Limited (AECL) has adopted a dense compacted mixture of unsaturated sand and bentonite as one of the seals in a multi-barrier approach. The effects of changes in osmotic suction on the behaviour of expansive soils such as sand-bentonite are relatively unknown. In such soils, suctions are generally large. Testing requires total suctions, not the matric and osmotic components, to be controlled or measured using vapour equilibrium techniques or psychrometers (Delage *et al.* 1995, Wan *et al.* 1995). Total suctions can only be interpreted easily when one of the two components is dominant. When both components contribute significantly, interpreting the results requires a good understanding of controlling mechanisms and processes. For example, when interpreting mechanical behaviour in terms of total suction, it is important to understand the changes in osmotic suction that occur when soil chemistry is altered. Conceptual models such as the diffuse double layer theory and the osmotic pressure concept are commonly used to explain the influence of osmotic suction, at least in an approximate way (Barbour and Yang 1993).

SPECIMEN PREPARATION

Specimens of AECL's sand-bentonite mixture, known as 'buffer', were made from equal masses of oven dry Na-bentonite and quartz sand (Graham *et al.* 1992). Two different pore fluids were used (1) distilled deionized water, and (2) sodium chloride solutions with concentrations of 0.1 M, 0.5 M, and 1.0 M NaCl. After mixing, the material was cured for 3 days to allow moisture equalization. Specimens for suction measurement were 40 mm high × 50 mm diameter. Specimens used for strength testing were 100 mm high × 50 mm diameter. Most specimens were statically compacted to 1.67 Mg/m^3 (Graham *et al.* 1995) using the amount of water or NaCl solution needed for degrees of saturation of 65% - 98%. For comparison, some specimens were prepared at about 1.4 Mg/m^3. Strengths and stiffnesses were measured in quick undrained triaxial tests.

EXPERIMENTAL METHOD

Suction measurement. Tests measured total suctions in saturated specimens, and total and matric suctions in unsaturated specimens at various water contents. Depending on the pore fluid, water content, and degree of saturation, the tests used either the filter paper technique or thermocouple psychrometers in conjunction with a Campbell Scientific CR-7 measuring and control system.

Strength and compression. Quick undrained triaxial compression tests were performed with both water and NaCl solutions as pore fluid. Methodology generally followed procedures described by

Graham *et al.* (1995). Total suctions before shearing were measured using psychrometers installed in identical specimens. After shearing, suctions were measured in the specimens after drilling a 5.0 mm diameter hole in the surface of the specimen (Tang *et al.* 1997a).

RESULTS AND INTERPRETATION

Initial osmotic suctions Figure 1 shows the measured relationship between water content, matric suction and total suction for the buffer. Detailed procedures were reported by Tang *et al.* (1997b). The difference between the total suction and matric suction lines in Fig.1 is almost constant, and assumed to be the osmotic suction. The average difference between total suction and matric suction (the osmotic suction) is 2.5 MPa over the range of water contents used in the tests. The figure also shows the relationship between suctions measured using filter papers and psychrometers. There is fairly good agreement between the two methods. When the water content is 22.7% or higher, specimens approach saturation, the matric suction goes to zero, and total and osmotic suctions are equal. The data in Fig.1 for saturated specimens with water contents ranging from 22.7 % to 50.0% show osmotic suctions decreasing slowly with increasing water content. Such reductions have been calculated using the van't Hoff equation (Mitchell 1976). However, the equation assumes widely separated particles in a suspension, and the agreement was obtained using cations only, without considering anions. In unsaturated specimens with water contents less than 22.7%, total suctions increase rapidly with decreasing water content. The initial osmotic suction varies only slowly when other conditions such as soil water chemistry and temperature remain unchanged. Thus, for the dry density used in these tests, increases in total suctions arise mainly from changes in matric suction with saturation, or more precisely, with water content.

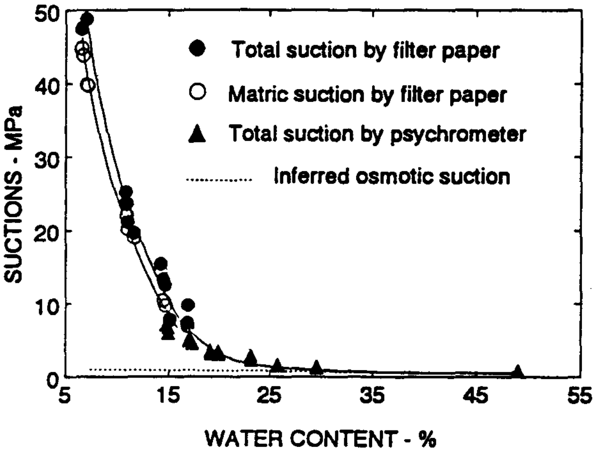

Figure 1. Suctions versus water content.

Elevation of total suctions by osmotic solutions Since osmotic suctions are controlled mainly by exchangeable cations in the mineral particles, they do not change greatly with water content when only deionized water is used. However, they do change when salt solutions are introduced to the pore fluid. This produces corresponding changes in total suctions (Fig.2). Specimens were examined at three saturations, namely 65%, 85%, and 98%; and NaCl concentra-

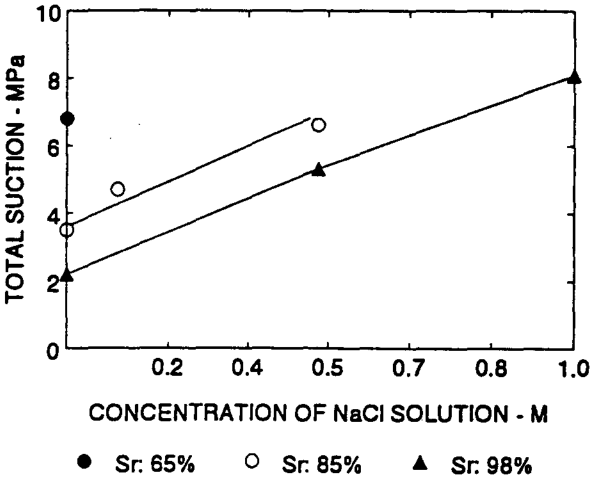

Figure 2. Total suction versus concentration.

tions C = 0.0 M, 0.1 M, 0.5 M, and 1.0 M. The data at 0.0 M concentration were made with deionized water. However, exchangeable cations available in the dry bentonite meant that the pore fluid became an electrolyte (Dixon *et al.* 1996). Increases in total suction introduced by NaCl solutions are not simply additive: measured total suctions are greater than the sum of the suction in a specimen made with deionized water and the osmotic suction of the solution itself. For example, 1 M NaCl solution has osmotic suction of 4.6 MPa. However, its use as pore fluid raises total suction in a saturated specimen by 5.9 MPa from 2.2 to 8.1 MPa (Fig. 2). At particulate level, increasing the pore fluid concentration, increases potentials near the particle surface, but decreases the thickness of the diffuse double layers (Mitchell 1976, Yong and Warkentin 1975).

Osmotic suction and strength Figure 3 shows how the maximum deviator stress (undrained shear strength) *vs.* degree of saturation changes with matric suction. (The tests were done with a low confining pressure of 0.2 MPa.) As expected, unsaturated specimens are stronger than saturated specimens due to the normal increase in matric suction (Fig.1) with decreasing saturation (Graham *et al.* 1995). However, according to Figs.3 and 4, increasing osmotic suctions produced by saline pore water, produce slightly lower strengths. That is, strength seems to increase with matric suction (inferred from Fig. 3), but decreases with osmotic suction (Fig. 4). These comparisons are from specimens made with the same dry density, water content, and saturation. If the same compaction effort is used, specimens with NaCl pore fluid will compress more, and be stronger (Mitchell 1976). As mentioned earlier, total suctions were measured before and after triaxial shearing using psychrometers. Total suctions seemed to be unchanged by shearing. Similar results were noted by Wan *et al.* (1995). This suggests that total suctions in buffer are largely reversible even after large non-reversible deformations (Tang *et al.* 1997a).

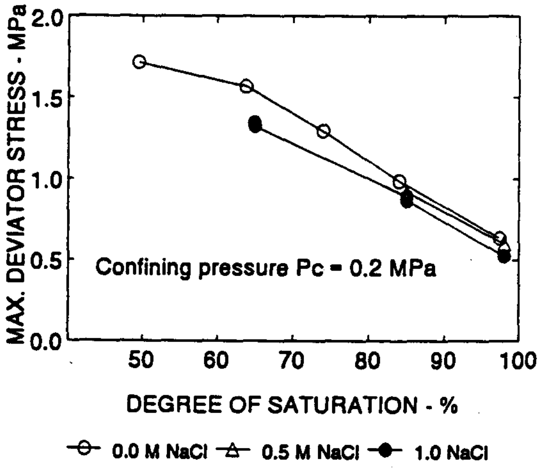

Figure 3. Max. deviator stress *vs.* saturation.

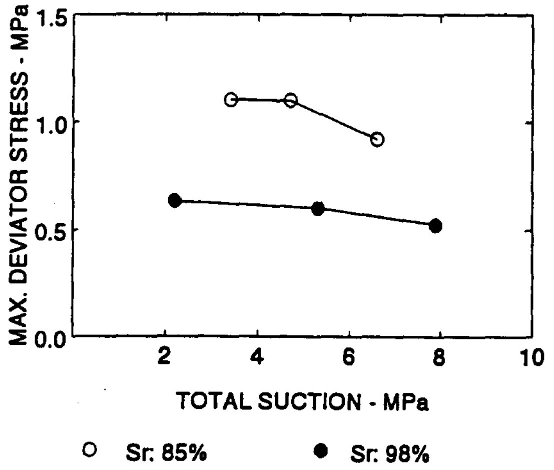

Figure 4. Max. deviator stress vs. total suction.

Osmotic suction and stiffness In a similar way, changes in osmotic suction affect the stiffness of specimens, here expressed as E_{50}, the secant modulus between the beginning of shearing and 50% of the maximum deviator stress. Figure 5 shows that the E_{50}-modulus for unsaturated specimens increases with decreasing saturation. Introducing 1.0 M NaCl increases the osmotic suction but decreases the stiffness. It should again be remembered that specimens at a given water content and saturation were all made with the same initial dry density.

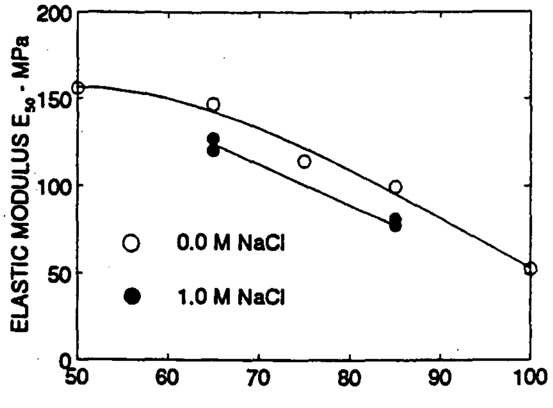

Figure 5. Elastic modulus *vs.* saturation.

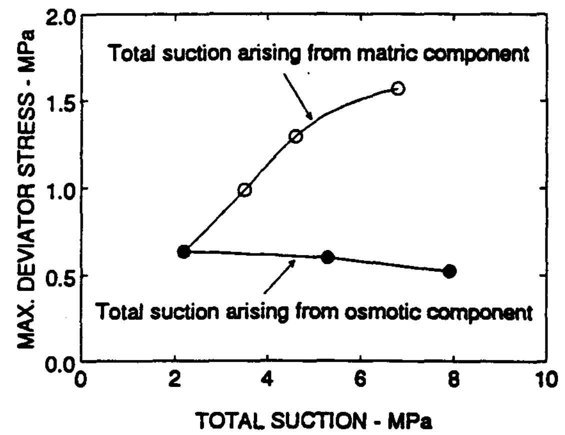

Figure 6. Effects of matric suction and osmotic suction on strength.

Influences of osmotic suction and matric suction Total suctions can arise from various combinations of matric suction and osmotic suction. For example, from Fig.2, a total suction of 7 MPa can be reached by (i) $C = 0.0$ M, $S_r = 65\%$; (ii) $C = 0.6$ M, $S_r = 85\%$; or (iii) $C = 0.8$ M, $S_r = 98\%$. Here, total suctions are largely controlled by the water content at the time of initial hydration of the dried solids (Wan *et al.* 1995).

These effects are examined in Fig.6 which shows maximum deviator stress plotted against total suction. The suctions were reached by either changing matric suction through controlling the water content and saturation, or changing osmotic suction through the pore fluid concentration. The specimens again had the same initial dry density. Figure 6 indicates the relative importance of osmotic suction compared with matric suction. Increases in osmotic suction decrease strengths and stiffnesses by small amounts. Increases in matric suction cause larger increases in strength and stiffness. This appears to negate the possibility proposed by Chattopadhyay (1972) that total suction, the algebraic sum of osmotic and matric suction, can be considered an independent stress state variable. Osmotic suctions and matric suctions may contribute separately as state variables, particularly when dealing with the entire range of soil suction.

DISCUSSION

The reason shear strength decreases with osmotic suction (at constant dry density) is clarified by considering the thickness of diffuse double layers round clay particles with saline pore fluid. In clays, long range attractive and repulsive forces develop between particles (Mitchell 1976) over distances of the order of 3 Å to perhaps 100 Å or more from particle surfaces. The attractive forces are primarily due to London van der Waals forces and decrease rapidly with distance from the particle surface. Adsorption of cations by clays and the formation of double layers are responsible for long range repulsive forces between particles. These develop by electrostatic repulsion between adjacent clay particles and associated overlapping diffuse double layers. For densities that are encountered in most soils engineering, electrostatic repulsion is the dominant long range force. This concept was incorporated by Graham *et al.* (1992) into an understanding of the stress state controlling the behaviour of dense saturated buffer.

The water surrounding bentonite particles in buffer at its reference dry density is only about 2 to 3 molecules thick (Graham *et al.* 1992). Nevertheless, for the following discussion, it will be assumed that diffuse double layer theory is applicable. Clay particle systems are frequently conceptualized as a series of parallel clay particles. The Poisson-Boltzmann equation (Mitchell 1976) for a single particle can be integrated to obtain the mid-plane electrolyte concentration and potential between two clay particles. An approximate indication of the influences of particle spacing and pore fluid chemistry can be seen in terms of the "thickness" of the double layer as given by:

$$\frac{1}{K} = \left(\frac{D\kappa T}{8\pi n_0 e^2 \upsilon^2} \right)^{0.5}$$

where $1/K$ = thickness of the double layer, D = dielectric constant, κ = Boltzmann constant, T = temperature (Kelvin), n_o = bulk solution electrolyte concentration, e = unit electronic charge, ν = cation valence. The equation shows that the thickness of the double layer decreases inversely as the square root of the concentration, other factors remaining constant. Long range interparticle repulsive forces depend on the interaction between adjacent double layers. In general, the thinner the double layer at a given particle separation, the smaller will be the repulsive force and therefore, the lower the strength. The thickness of double layers in the bentonite component of buffer decreases when concentrated solutions are introduced to the specimens. Strengths therefore decrease when the density is held constant. If the material is allowed to consolidate under drained compression, then the density of the salt-rich material should increase. However, the strength behavior is unclear.

Fig. 7 compares stress-strain behavior of two sets of specimens. Each set consisted of one specimen made with deionized water (low osmotic suction), and the second made with 1.0 M NaCl (elevated osmotic suction). All specimens were formed at 19.40% water content. To compare the effect of compaction, two different test procedures were used. One specimen from each set was sheared in a conventional undrained triaxial compression test with confining pressure of 0.2 MPa. The remaining specimens were formed by compacting loose buffer to a density of about 1.4 Mg/m^3, significantly looser than the 1.67 Mg/m^3 of the first specimens. They were then compressed in triaxial cells at 3 MPa for

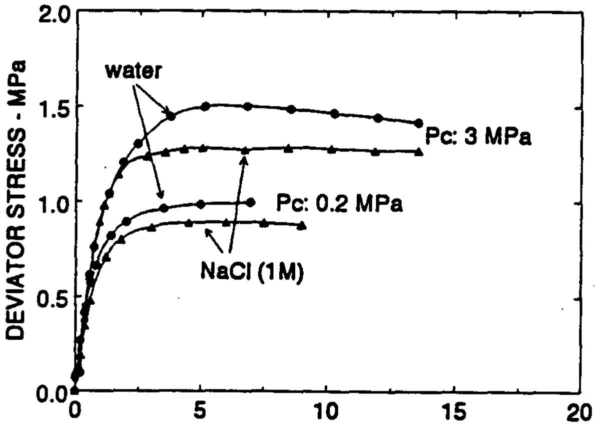

Figure 7. Deviator stress *vs.* axial strain.

12 hours with open drainage for the air phase, and finally sheared to failure with the same confining pressure of 3 MPa. In Fig.7, specimens with NaCl pore fluid (and hence, elevated osmotic suctions) have consistently lower strength. Delage and Graham (1996) showed that microstructure was controlled by hydrating water content, and influenced to only a lesser extent by compaction effort or, here, compression.

SUMMARY AND CONCLUSIONS

Understanding the suction-dependent mechanical properties of sand-bentonite buffer requires an understanding of the separate effects of matric suction and osmotic suction. Osmotic suctions change with hydrating water content and with the chemistry of the pore water.

Subject to the restriction that specimens have the same dry density, the strength and stiffness of buffer decrease when total suctions are increased by osmotic agents. Elevated total suctions introduced by salt in the pore fluid produce small decreases (not increases) in shear strength or stiffness. This is believed to be due to compression of potential fields round charged particles with increasing pore fluid concentration. Larger increases in strength and stiffness accompany similar increases in matric suction. Roles played by osmotic suction and matric suction in controlling strength and stiffness are not quantitatively equivalent. While matric and osmotic suctions combine algebraically, it is unlikely that total suction can be used as a single stress state variable.

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