

## Mechanisms of osmotic flow and volume change in clay soils

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The mechanical behavior of compressible clay soils may be strongly influenced by physicochemical effects when concentrated pore fluids are introduced to the soil. Conceptual models have been used to explain the influence of pore fluid chemistry on the mechanical behavior of clays in a qualitative way. In this paper an alternate macroscopic description of the osmotic volume change behavior of a clay soil undergoing changes in pore fluid chemistry is provided.

Theoretical descriptions of two potential mechanisms of osmotic volume change (osmotic consolidation and osmotically induced consolidation) are presented. *Osmotic consolidation* occurs as a result of a change in the electrostatic repulsive-minus-attractive stresses,  $R - A$ , between clay particles. *Osmotically induced consolidation* occurs because of fluid flow out of the clay in response to osmotic gradients.

A numerical simulation is used to demonstrate the characteristic behavior of a clay soil undergoing either of these volume change processes. The results of a laboratory testing program on two clay soils exposed to concentrated NaCl solutions are used to illustrate that the dominant mechanism of osmotic volume change in surficial clay soils is osmotic consolidation.

*Key words:* physicochemical, osmosis, volume change, NaCl salt, montmorillonite, clay, stress state variables,  $R - A$ .

Le comportement mécanique des sols argileux compressibles peut être fortement influencé par des effets physico-chimiques lorsque des liquides interstitiels concentrés sont introduits. Des modèles conceptuels ont été utilisés pour expliquer l'influence de la chimie du liquide interstitiel sur le comportement mécanique des argiles de façon qualitative. Dans le présent article, l'on donne une description macroscopique alternative du comportement du changement de volume osmotique d'un sol argileux subissant des changements dans la chimie du liquide interstitiel.

L'on présente les descriptions théoriques de deux mécanismes potentiels de changement de volume osmotique (consolidation osmotique et consolidation induite par osmose). La *consolidation osmotique* résulte d'un changement dans la différence entre les contraintes de répulsion et d'attraction électrostatiques,  $R - A$ , entre les particules argileuses. La *consolidation induite par osmose* est due à l'expulsion de l'eau de l'argile par suite des gradients osmotiques.

Une simulation numérique est utilisée pour démontrer le comportement caractéristique du sol argileux subissant l'un ou l'autre de ces processus de changement de volume. Les résultats d'un programme d'essais en laboratoire sur deux sols argileux exposés à des solutions de NaCl concentrées sont utilisés pour illustrer le fait que le mécanisme dominant du changement de volume osmotique dans les sols argileux de surface est la consolidation osmotique.

*Mots clés :* physico-chimie, osmose, changement de volume, sel NaCl, montmorillonite, argile, variables d'état de contrainte,  $R - A$ .

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### Introduction

The mechanical behavior of clay soils may be strongly influenced by physicochemical interactions between the clay particles and the pore fluid. Conceptual and analytical models have been proposed to explain the influence of pore fluid chemistry on the behavior of clays. These models have been developed at a microscopic scale and have provided a qualitative description of interactions.

A quantitative description of the behavior of clay soils in response to changes in pore fluid chemistry is required in current geotechnical practice. For example, in the design of containment facilities for concentrated electrolyte solutions (i.e., brine) or organic fluids, the impact that these fluids have on the hydraulic and mechanical properties of soil must be known. In this paper, the impact that strong NaCl solutions have on the volume change behavior of clay soils is used to illustrate the influence of inorganic salts on the mechanical behavior of clay soils.

The time-dependent volume change behavior of soil is controlled by two properties: first, the compressibility of the soil, that is, its change in volume related to changes in stress state; and second, the ability of the soil to conduct pore fluid. The ability of physicochemical (i.e., osmotic)

effects to influence both soil compressibility and fluid flow through soil has been demonstrated in the literature.

The presence of osmotic flow in clay aquitards has been studied by numerous researchers, including Hanshaw and Zen (1965), Marine and Fritz (1981), and Neuzil (1986). The influence of osmotic flow processes on the transport of salt has been studied by Kemper and van Schaik (1966), Kemper and Rollins (1966), Elrick *et al.* (1976), Greenberg *et al.* (1973), and others. Deviations from Darcy's law have also been attributed to osmotic flow by investigators such as Kemper (1961), Low (1955), Bolt and Groenevelt (1969), and Olsen (1985).

Extensive studies of the compressibility and swelling of clays in response to changing pore fluid concentrations have been conducted over the last 30 years, and include the work of Bolt and Miller (1955), Bolt (1956), Warkentin *et al.* (1957), Aylmore and Quirk (1962), Blackmore and Miller (1962), Warkentin and Schofield (1962), and Mesri and Olson (1971). Bailey (1965) and Mitchell (1973a) have provided reviews of this literature.

Up to the present, a comprehensive theoretical description of the combined influence of osmotic flow and volume change on the behavior of clay soils has been lacking. The

objective of this paper is to provide a theoretical description of the two primary mechanisms for osmotic volume change, referred to in this paper as *osmotically induced consolidation* and *osmotic consolidation*.

When a clay is exposed to a concentrated salt solution the volume of the clay will change as a result of the combined influence of the processes of osmotic flow and osmotic compressibility. As salt is transported into the clay changes in the interparticle repulsive stresses will occur, which will in turn lead to suppression of the double layer and changes in void ratio. The time-dependent volume change associated with changes in the osmotic pressure of the pore fluid will be termed osmotic consolidation.

A second mechanism of volume change will occur, however, as a result of fluid flow that develops within the clay in response to osmotic gradients. As the clay is exposed to the electrolyte solution osmotic flow out of the sample will occur. These outward flows cause negative pore fluid pressures to develop within the sample, which then lead to increases in effective stress,  $\sigma - u_f$ , and consequently volume change. The volume change that occurs is still in response to changes in effective stress. However, because the changes in pore-water pressure are induced as a result of osmotic pressures, this form of volume change is called osmotically induced consolidation.

Characteristic features of these two processes are demonstrated through the use of a numerical model, and the relative importance of these two independent processes is demonstrated from the results of a laboratory testing program. An analysis of these test results illustrates some practical applications of the proposed approach to geotechnical engineering design in situations in which changes in pore fluid chemistry are of concern.

### Theoretical description of osmotic flow and compressibility

Consolidation is the result of the combined influence of fluid flow and soil compressibility. Changes in pore fluid chemistry may significantly influence both volume change and fluid flow through clay. Theoretical descriptions of these "osmotic" phenomena are presented in this section.

#### Osmotic flow

Osmosis is the term used to describe the phenomenon by which a solvent passes from a solution of lower solute concentration through a semipermeable membrane into a solution of higher solute concentration. A membrane is described as semipermeable if it allows the passage of solvent but not solute. If the flow of water is restricted, a pressure imbalance equal to the osmotic pressure difference between the two solutions would have to be present. The osmotic pressure can be calculated from thermodynamic principles (Robinson and Stokes 1968). The osmotic pressure can also be approximated by the van't Hoff equation (Metten 1966):

$$[1] \quad \Pi = RTC$$

where  $\Pi$  = osmotic pressure (kPa),  $C$  = sum of the molar concentrations in solution (mol/L),  $R$  = universal gas constant = 8.32 L·kPa/K·mol), and  $T$  = absolute temperature (K).

The osmotic flow of water in soil can be described using a flow law similar in form to Darcy's law:

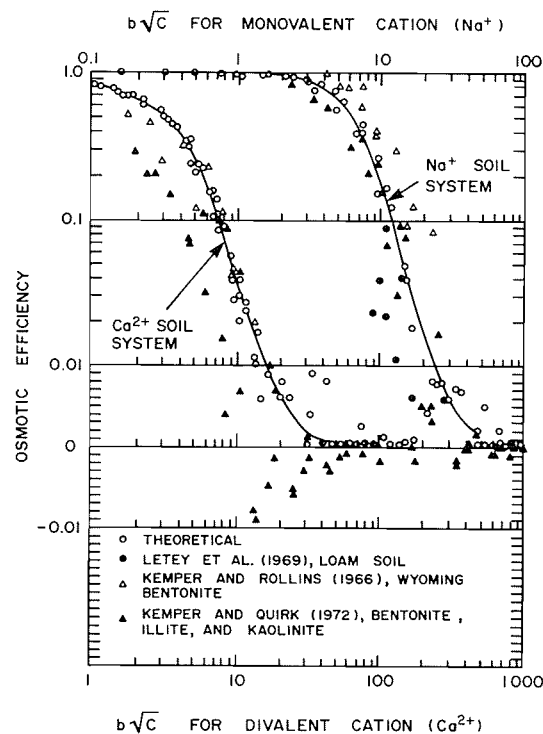


FIG. 1. Osmotic efficiency for a monovalent anion, unsaturated soil system, as a function of  $b\sqrt{C}$  ( $C$  is pore fluid concentration in normality,  $2b$  is the film thickness in angstroms, Bresler 1973).

$$[2] \quad q_{\pi} = k_{\pi} \frac{\Delta\pi}{\Delta x} = \sigma k_h \frac{\Delta\pi}{\Delta x}$$

where  $q$  = water flux (m/s),  $k_{\pi}$  = coefficient of osmotic permeability (m/s),  $k_h$  = coefficient of (hydraulic) permeability (m/s),  $\pi$  = osmotic pressure head =  $\pi/\rho_f g$  (m),  $\rho_f$  = pore fluid density ( $\text{kg/m}^3$ ),  $g$  = gravitational acceleration ( $\text{m/s}^2$ ),  $\sigma$  = osmotic efficiency, and  $x$  = distance (m).

If the soil behaves as a perfect semipermeable membrane, the coefficient of osmotic permeability will be equal to the coefficient of hydraulic permeability. In this case only pure water will flow in response to osmotic gradients. However, if the membrane is "leaky," the osmotic permeability will be equal to the hydraulic permeability multiplied by an osmotic efficiency, and water, carrying with it some dissolved salts, will flow in response to osmotic gradients. The osmotic efficiency is a measure of the degree to which the clay behaves as a perfect semipermeable membrane.

The osmotic efficiency of clays has been studied by Kemper and Rollins (1966), Bresler (1973), Olsen (1972), and others. Results of these studies (Fig. 1) illustrate that osmotic efficiency is strongly dependent on pore fluid chemistry, pore fluid concentration, void ratio, and interparticle spacing. High osmotic efficiencies have been observed only at low pore fluid concentrations, or low void ratios.

Figure 1 illustrates the dependence of osmotic efficiency of an unsaturated soil on pore fluid chemistry, pore fluid concentration, and fluid film thickness between soil particles. The film thickness is obtained by dividing the volumetric water content by half the soil surface area.

### Osmotic compressibility

Volume changes in clays may occur as a result of changes in pore fluid chemistry due to the alteration of the electrostatic interactions between the clay particles. A conceptual model of the nature of the repulsive interparticle pressures is provided by the osmotic pressure concept and diffuse double layer theory.

Most clay minerals have a net negative surface charge. In the case of electroneutrality, a diffuse double layer of cations (and anions) develops around the clay particle. The negatively charged clay surface and the charge distribution adjacent to the clay particle are termed the diffuse double layer. Theoretical descriptions of the diffuse double layer were first proposed by Gouy (1910) and Chapman (1913) and later modified by Stern (1924). Detailed developments of diffuse double layer theory are provided by Mitchell (1976) and van Olphen (1977). The distribution of charge density and electric potential in the diffuse double layer are described by an equation known as the Poisson-Boltzman equation.

The charge density and electric potential vary as a function of distance from the clay surface, surface charge density, surface potential, electrolyte concentration and valence, dielectric constant of the fluid, and temperature. The influence of these factors can be seen in the equation for the distance to the center of gravity of charge density surrounding a semi-infinite negatively charged clay particle. This distance can be viewed as being representative of the "thickness" of the diffuse double layer (Mitchell 1976):

$$[3] \quad 1/K = (DkT/8\pi n_0 \epsilon^2 \nu^2)^{1/2}$$

where  $1/K$  = "thickness" of the double layer (cm),  $D$  = dielectric constant,  $k$  = Boltzmann constant =  $1.380\ 662 \times 10^{-23}$  J/K,  $n_0$  = bulk solution electrolyte concentration (ions/cm<sup>3</sup>),  $\epsilon$  = unit electronic charge (esu),  $\nu$  = cation valence, and  $T$  = absolute temperature (K).

Equation [3] illustrates that the thickness of the diffuse double layer changes in response to changes in dielectric constant, concentration, and valence of the pore fluid. Changes in the behavior of clay soils due to changes in these pore fluid properties have been described by Mesri and Olson (1971).

In clay soils, long-range attractive and repulsive forces develop between particles. The long-range attractive force is primarily due to London van der Waals forces. The long-range repulsive force develops as a result of electrostatic repulsion between two adjacent clay particles. In many cases, the dominant long-range force between clay particles is electrostatic repulsion (Bailey 1965; Mitchell 1976).

The osmotic pressure concept can be used to calculate the repulsive stress between clay particles. The clay particle system is assumed to exist as a series of parallel clay particles. The Poisson-Boltzman equation for a single particle can be integrated to obtain the mid-plane electrolyte concentration and mid-plane potential between two clay particles. The half spacing between the clay particles can be estimated from the void ratio and the specific surface of the clay particles (Mitchell 1976).

In the osmotic pressure concept the overlapping diffuse double layers are considered as behaving as a semipermeable membrane. The difference in osmotic pressure between the bulk pore fluid and the fluid between the clay particles is then taken to be equivalent to the electrostatic repulsive

pressure between the particles. Detailed developments of the osmotic pressure concept are provided by Bolt (1956) and Mitchell (1976).

Researchers have attempted to use the osmotic pressure concept to predict the void ratio versus pressure relationship for clays containing low concentrations of homoionic pore fluid solutions. The osmotic pressure concept has been found, however, to provide a quantitative description of soil compressibility only for soils at high void ratios. The major limitation of this approach is the assumption of a perfectly dispersed parallel arrangement of individual clay particles, and the assumption that the dominant interparticle forces are the electrostatic repulsive force (Bailey 1965).

A second approach in dealing with osmotic compressibility considers the interparticle forces at a macroscopic scale. Barbour (1987a, b) and Barbour and Fredlund (1989) developed a set of stress state variables from a continuum mechanics approach for a saturated soil under physico-chemical as well as mechanical stresses. A three-phase soil system consisting of the soil particles, the pore fluid, and the diffuse double hull surrounding each soil particle was considered. The three-dimensional stress state for this element was expressed as follows:

$$[4] \quad \begin{bmatrix} \sigma_x - u_f - (R - A) & \tau_{xy} & \tau_{xz} \\ \tau_{yx} & \sigma_y - u_f - (R - A) & \tau_{yz} \\ \tau_{zx} & \tau_{zy} & \sigma_z - u_f - (R - A) \end{bmatrix}$$

where  $\sigma_x$ ,  $\sigma_y$ ,  $\sigma_z$  = total normal stresses,  $\tau_{xy}$ ,  $\tau_{xz}$ ,  $\tau_{yx}$ ,  $\tau_{yz}$ ,  $\tau_{zx}$ ,  $\tau_{zy}$  = shearing stresses,  $u_f$  = pore fluid pressure, and  $R - A$  = net interparticle repulsive-minus-attractive stress.

The stress state variable for the normal stress in [4] can be subdivided into two components. The first component is the conventional effective stress for a saturated soil,  $\sigma - u_f$ . The second component is the net interparticle repulsive-minus-attractive stress,  $R - A$ . The two stress components can be visualized as a single stress state variable,  $\sigma - u_f - (R - A)$ . This is an expression similar to that for the "true" effective stress proposed by Balasubramonian (1972) and Chattopadhyay (1972):

$$[5] \quad \sigma^* = \sigma - u_f - (R - A)$$

where  $\sigma^*$  = true effective stress. A similar expression based on static equilibrium along a wavy plane through a soil particle was proposed by Lambe (1960), Bailey (1965), and Mitchell (1962) also.

Two requirements need to be met to accept a particular combination of stresses as a single stress state variable. The first requirement is that change in the value of one of the components of the stress state variable must be as effective in producing behavioral changes in the soil mass as the same magnitude of change in one of the other components of the stress state variable. Fredlund and Morgenstern (1977) demonstrated that a null type test could be used to confirm the correct components for the stress state variables for unsaturated soils using this technique. Testing by Balasubramonian (1972), Morgenstern and Balasubramonian (1980), and Chattopadhyay (1972) demonstrated that if  $R - A$  could be calculated, changes in the effective stress were equal to changes in  $R - A$  when the volume of the specimen was maintained constant.

The second requirement for the stress state variable,

however, is that all components of the stress state variable be measurable. When using the true effective stress,  $\sigma^*$ , it is not possible to measure  $R - A$  directly and the prediction of  $R - A$  is possible only under ideal conditions. This is a severe restriction to the use of  $R - A$  as a component of the stress state. Because of this, it may be necessary that a deduced quantity, which is more easily measured than  $R - A$ , be used.

The osmotic pressure concept relates the change in repulsive stress between clay particles to the change in the osmotic pressure difference between the interparticle fluid and the bulk pore fluid. Mitchell (1962) suggested that "double layer interactions and the consequent repulsion between opposing particles are reflected by the osmotic pressure." The osmotic pressure is described as having two components: one associated with the free salt in the bulk solution and the other caused by the additional ions required to satisfy double layer requirements. The osmotic pressure of the bulk solution can be readily evaluated using samples of the pore fluid; the osmotic pressure of the interparticle fluid, however, cannot be measured. Consequently, the difference between the osmotic pressure of the bulk fluid and that between the clay particles cannot be evaluated.

In most practical problems, the change of osmotic pressure is of more interest in regard to the process of osmotic compressibility than the absolute value of the components of osmotic pressure. In problems of osmotic compressibility, changes in the osmotic pressure of the bulk solution will be large relative to the initial osmotic pressure within the sample. Consequently, it can be assumed that the change in the osmotic pressure of the bulk solution forms a satisfactory independent stress state variable. However, the osmotic pressure cannot be incorporated into the single stress state,  $\sigma - u_f - (R - A)$ , because osmotic pressure,  $\Pi$ , may not be as effective as  $\sigma - u_f$  in producing behavioral changes in the soil.

The change in the osmotic pressure of the bulk pore fluid is the driving force for osmotic flow. Consequently, the use of osmotic pressure as a stress state variable provides for consistency in the stresses used for the description of volume change as well as for the flow of the pore fluid.

*Constitutive relationship for volume change*

A general form for the relationship linking the stress state variables and volumetric strain can be written as follows:

$$[6] \quad \epsilon = -\frac{dV}{V} = -\frac{1}{V} \left[ \frac{\partial V}{\partial(\sigma - u_f)} d(\sigma - u_f) + \frac{\partial V}{\partial \Pi} d\Pi \right]$$

where  $\epsilon$  = volumetric strain and  $V$  = initial soil volume.

Equation [6] takes the form of a three-dimensional constitutive surface. The experimental data of Mesri and Olson (1971) illustrating the varying compressibility of a Na-montmorillonite clay with various concentrations of pore fluid are shown in Fig. 2a. Replotting this data using the osmotic pressure of the pore fluid as a stress state variable produces the three-dimensional constitutive surface shown in Fig. 2b. Along a plane in which  $\Pi$  is constant, the coefficient of volume change (compressibility),  $m_v$ , can be obtained. Along a plane in which  $\sigma - u_f$  is constant, the slope can be used to define the osmotic coefficient of volume change (osmotic compressibility),  $m_\pi$ . The equation corresponding to a point on the constitutive surface can be written as

$$[7] \quad d\epsilon = -m_v d(\sigma - u_f) - m_\pi d\Pi$$

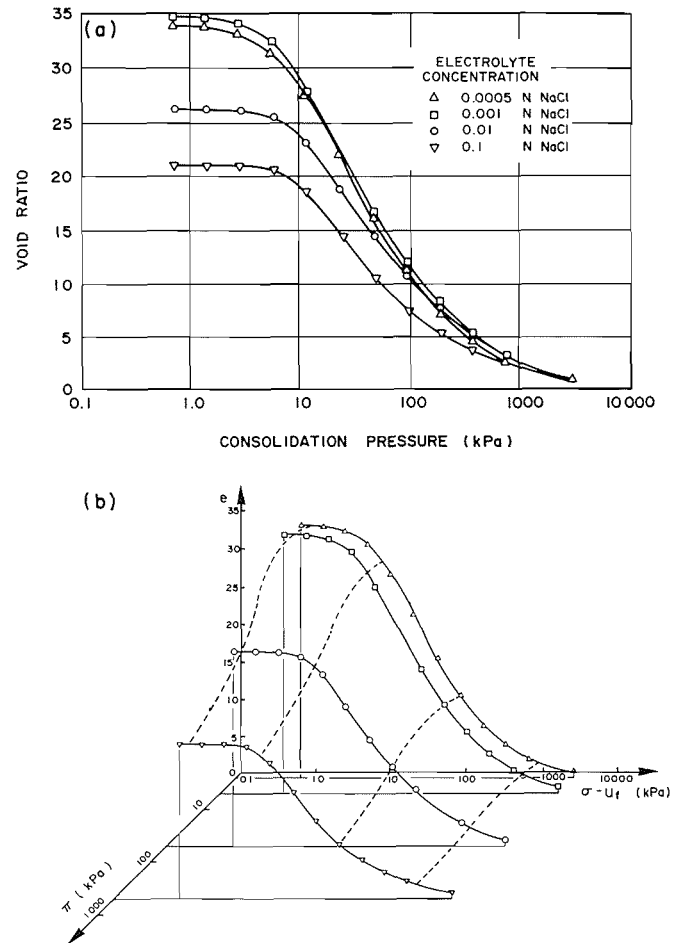


FIG. 2. (a) Consolidation curves for Na<sup>+</sup>-montmorillonite (after Mesri and Olson 1971); (b) three-dimensional constitutive surface for Na<sup>+</sup>-montmorillonite.

It should be noted that  $m_v$  and  $m_\pi$  are not necessarily material constants. Just as  $m_v$  must be evaluated over an appropriate stress range, so must  $m_\pi$  be evaluated over an appropriate change in pore fluid chemistry.

*Theoretical description of osmotic volume change—osmotic consolidation and osmotically induced consolidation*

Osmotically induced consolidation occurs as a result of the flow of water out of a soil in response to chemical or osmotic gradients. This outward flow causes negative fluid pressure to develop within the clay, and consequently produces an increase in effective stress. The second osmotic volume change process is termed osmotic consolidation. In this process, volume change occurs because of a reduction in interparticle repulsive stresses as a result of changes in pore fluid chemistry.

Osmotically induced consolidation was addressed in a study by Greenberg (1971) and subsequently discussed by Greenberg *et al.* (1973) and Mitchell (1973b, 1976). The theoretical formulation of volume change due to osmotically induced consolidation was originally developed by Greenberg (1971) using the theory of irreversible thermodynamics. Barbour (1987a, b) formulated the problem from a phenomenological basis more familiar to geotechnical engineers. This formulation described transient volume

TABLE 1. Summary of theoretical relationships for osmotic flow and volume change

Flow laws	
For fluid:	$q_h = k_h(\partial h/\partial x)$ $q_\pi = k_\pi(\partial \pi/\partial x)$ $q_f = q_h + q_\pi$
For salt:	$q_s = q_f C - nD(\partial C/\partial x)$
Constitutive relationships	
	$\Delta e/(1 + e_0) = \Delta(\sigma - u_f)m_v$ $\Delta e/(1 + e_0) = \Delta \Pi m_\pi$
Material properties	
	$k_h$ = coefficient of permeability $k_\pi$ = coefficient of osmotic permeability $D$ = coefficient of diffusion $n$ = porosity $e$ = void ratio $m_v$ = coefficient of volume change $m_\pi$ = osmotic coefficient of volume change $C$ = concentration

change due to both osmotic and osmotically induced consolidation.

#### Formulation for osmotic volume change—osmotic consolidation and osmotically induced consolidation

To predict transient volume change, changes in the stress state variables with time are related to volume change through the constitutive equation. Continuity is used to relate these volume changes to the divergence of the fluid flux from the element. These fluid fluxes are in turn defined by the flow laws for the fluid phase. It is assumed that only hydraulically and osmotically induced flows of the fluid occur. The potential of ion streaming to produce electrical gradients is not considered. The rate of movement of the dissolved salt is described by the advection-diffusion equation (Freeze and Cherry 1979). It is assumed that no geochemical interactions of individual ion species occur with the soil solids; however, retardation of dissolved salts due to adsorption onto the soil solids may be accommodated in this formulation.

Table 1 summarizes the flow laws and constitutive equations required to develop a description of transient volume change of clay soils due to osmotic or osmotically induced consolidation. The development and solution of the governing differential equations describing these phenomena were developed in detail by Barbour (1987a).

#### Numerical simulation

The equations given in Table 1 describing transient osmotic volume change were used to develop a one-dimensional numerical solution describing transient fluid flow and salt migration during osmotic volume change (Barbour 1987a). The numerical model was verified by comparison of the results with the simulations of Greenberg (1971) and Mitchell (1973a, 1976) for osmotically induced consolidation, and with analytical solutions of one-dimensional conventional consolidation and contaminant transport. At present no models of osmotic consolidation exist against which the present model can be verified. Rather, this model will be used to illustrate behavior that is typical of clay specimens undergoing either osmotic or osmotically induced consolidation.

The simulation was conducted for a 1 cm thick specimen of clay exposed to a 4.0 M NaCl brine along its upper surface. Two cases with different boundary conditions were analysed (Table 2). In case 1, the base of the specimen was considered to be sealed so that no fluid flow occurred across the base. In case 2, a zero pore pressure condition was maintained at the base. Salt flux across the base was not permitted in either case.

Two sets of material properties were used for the simulation cases to represent situations in which osmotically induced consolidation or osmotic consolidation was the dominant mechanism of volume change. In the first situation, osmotically induced consolidation will be the principal mechanism of volume change. The clay was assumed to behave as a semipermeable membrane with an osmotic efficiency of 0.005 (i.e.,  $k_\pi$  is equal to  $0.005k_h$ ). In the second situation, osmotic consolidation is the dominant mechanism of volume change. The clay was given an osmotic compressibility equal to 0.005 times the conventional compressibility of the sample, but with an osmotic efficiency of zero. These values of osmotic compressibility and osmotic efficiency are typical of those obtained in laboratory tests described later in the paper.

The results of the simulations are illustrated in Figs. 3 and 4. For case 1 (Fig. 3) it is apparent that the time-deflection behavior is characteristically different in the two mechanisms. Osmotically induced consolidation occurs within the same time frame as conventional consolidation. The maximum deflection of the specimen occurs at a time factor of approximately 1. Rebound then occurs as the negative pore pressures induced by osmotic gradients dissipate as the salt diffuses into the soil specimen. Osmotic consolidation, however, develops slowly in response to the diffusion of salt into the clay.

In case 2 (Fig. 4), in which fluid flow across the base of the specimen was permitted, the results from the two simulations are quite different. For the case of osmotic consolidation (Fig. 4b), fluid is expelled from the base, as the advancing salt front causes a reduction in volume. However, in the case of osmotically induced consolidation (Fig. 4a), strong upward flows across the base begin at a time factor of approximately 1.0 in response to the strong osmotic gradients near the top of the specimen. The rate of flow through the base becomes nearly constant with time as the salt distribution reaches an equilibrium position. The final flow rate is approximately equal to the osmotic permeability times the osmotic pressure gradient across the soil specimen.

#### Laboratory testing

Laboratory testing was conducted to establish the dominant mechanism of osmotic volume change in two clay soils. A simple modified oedometer was used to consolidate a clay slurry to various confining stresses. The specimen was then exposed to concentrated NaCl solutions along either the upper soil surface only or the top and bottom of the specimen simultaneously. Measurements of specimen deflection were made for both cases. In the first case, the amount of osmotic flow across the base of the specimen or the pressure that developed along the base of the sample when flow was prevented was also measured. The specimens tested were between 0.5 and 1.7 cm thick.

One natural soil and one artificial soil mixture were used in the tests. The natural soil was the minus No. 10 sieve frac-

TABLE 2. Numerical simulation example cases

General properties		Specific properties
$k_h = 1.0 \times 10^{-10}$ m/s $m_v = 5.0 \times 10^{-4}$ kPa <sup>-1</sup> $m_{vs} = 5.0 \times 10^{-5}$ kPa <sup>-1</sup> $c_v = 2.0 \times 10^{-4}$ cm <sup>2</sup> /s $D = 5.0 \times 10^{-10}$ m <sup>2</sup> /s $n = 0.5$ $m_{vs}$ = coefficient of volume change during swelling		Osmotically induced consolidation $k_\pi = 5.0 \times 10^{-13}$ m/s $m_\pi = 0$ Osmotic consolidation $k_\pi = 0$ $m_\pi = 2.5 \times 10^{-6}$ kPa <sup>-1</sup>
Geometry and boundary conditions		
Sample thickness:	1 cm	
Initial conditions:	$u = 0, C = 0$ for all $y; t \leq 0$	
Boundary conditions:	Case 1	Case 2
Top-flow boundary	1st type; $u = 0; t > 0$	1st type; $u = 0; t > 0$
Bottom-flow boundary	2nd type; $q_f = 0; t > 0$	1st type; $u = 0; t > 0$
Top-salt transport boundary	1st type; $C = 4 M; t > 0$	1st type; $C = 4 M; t > 0$
Bottom-salt transport boundary	2nd type; $\partial C / \partial y = 0; t > 0$	3rd type; $qC = 0; t > 0$ $C = 0; t > 0$

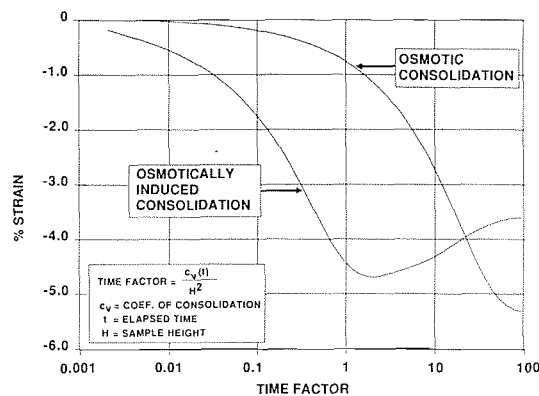


FIG. 3. Consolidation curves from numerical simulation of osmotic volume change for case 1 (no fluid flow across the base).

tion of air-dried samples of Regina Clay. The artificial soil was a mixture of 20% Na-montmorillonite and 80% Ottawa Sand. The slurries were prepared by mixing the air-dried soil to water contents 10–20% above the liquid limit. Tables 3 and 4 provide a summary of the physical and chemical characteristics of the two soil types. In contrast to the Na-montmorillonite – sand mixture, the Regina Clay is predominately a Ca-montmorillonitic soil.

**Test results**

Figure 5 illustrates a deflection curve for a specimen of Regina Clay undergoing normal effective stress consolidation from 100 to 200 kPa loading. At an elapsed time of 8000 min the upper surface of the specimen is exposed to a 4.0 M NaCl solution and further consolidation of the specimen takes place. Typical curves of specimen deflection versus time due to osmotic consolidation for the soils tested are illustrated in Figs. 6 and 7. These curves have been corrected for secondary consolidation as illustrated in Fig. 5. For the Regina Clay, the consolidation process is extremely slow and appears to be controlled by the rate of diffusion of salts into the specimen. The specimens of Na-montmorillonite – sand

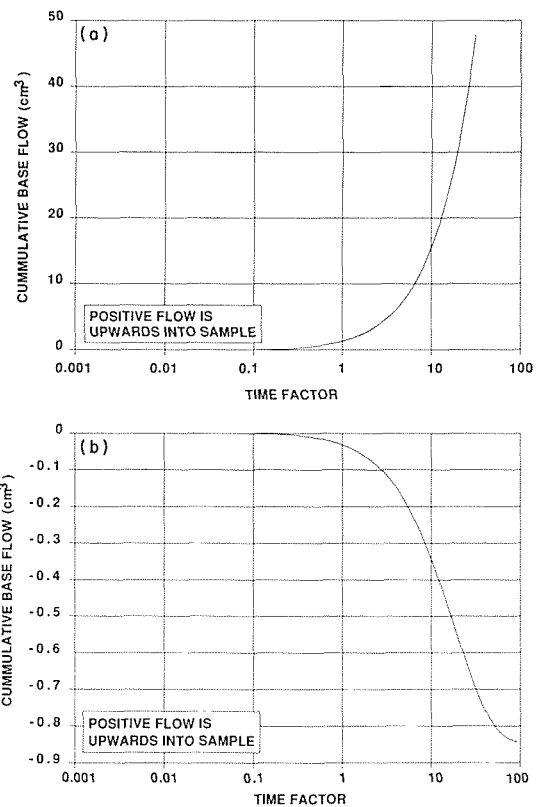


FIG. 4. Cumulative base flow curves from numerical simulation for case 2, (fluid flow across the base permitted): (a) Osmotically induced consolidation case; (b) osmotic consolidation case.

appear to undergo more rapid consolidation than the Regina Clay specimens. At first observation, this rapid consolidation would appear to be similar to that anticipated for osmotically induced consolidation (Fig. 3).

Figure 8 illustrates the effect of osmotic volume change as viewed on a conventional void ratio versus effective stress,  $\sigma - u_f$ , diagram. After osmotic volume change, the

TABLE 3. Summary of classification tests

Test	Regina Clay	Na-montmorillonite	Ottawa Sand	Mixture of sand-montmorillonite
Specific gravity	2.83	2.56	2.65	2.63
Atterberg limits				
Liquid limit	75.5%	—	—	62.1%
Plastic limit	24.3%	—	—	Nonplastic
Plasticity index	51.2%	—	—	—
Grain-size distribution				
Sand sizes	0%	0%	100%	80%
Silt sizes	34%	0%	0%	0%
Clay sizes	66%	100%	0%	20%
Mineralogical composition— less than 2 $\mu\text{m}^a$				
Montmorillonite	45.2%	80% <sup>b</sup>	—	—
Illite	27.7%	7%	—	—
Kaolinite	17.7%	7%	—	—
Trichlorite	9.4%	0%	—	—
Quartz	0.0%	8%	—	—
Gypsum	0.0%	3%	—	—
Carbonate	0.0%	2%	—	—
Specific surface ( $\text{m}^2/\text{g}$ )	53 <sup>c</sup>	700-840 <sup>d</sup>	—	140-168
Exchange capacity (mequiv./100 g) <sup>e</sup>	31.7	80-150 <sup>d</sup>	—	—
Total exchangeable bases by $\text{NH}_4\text{Ac}$ leaching (mequiv./100 g)				
Magnesium	15.3	—	—	—
Calcium	54.4	—	—	—
Potassium	0.59	—	—	—
Sodium	1.77	—	—	—

<sup>a</sup>Test performed by Saskatchewan Research Council.<sup>b</sup>Quigley (1984).<sup>c</sup>Test performed by Department of Soil Science, University of Saskatchewan, by ethylene glycol sorption (Black 1965a, Part 1).<sup>d</sup>Mitchell (1976).<sup>e</sup>Fredlund (1975).TABLE 4. Chemical analysis of saturated extract<sup>a</sup>

Test result	Regina Clay	Na-montmorillonite
Water content of extract (%)	88	550
pH	7.4	8.8
Conductivity (mS/cm)	3.7	4.0
Ions in extract ( $\mu\text{g}/\text{mL}$ )		
$\text{Na}^+$	210	910
$\text{Ca}^{2+}$	546	23
$\text{Mg}^{2+}$	183	7
$\text{K}^+$	35	9
$\text{Cl}^-$	133	81
$\text{SO}_4^{2-}$	1700	1400
Sodium adsorption ratio	2.0	42.2

<sup>a</sup>Analyses performed by Saskatchewan Soil Testing Laboratory (after Black 1965b).

specimen appears to be overconsolidated to some higher stress level. Upon reloading, the specimen recompresses back towards the original virgin branch.

For the cases where only the top of the specimen was exposed to brine, measurements were made of the flow across the base of the specimen. The reservoir for this flow was maintained at the same elevation as the brine level on the top of the specimen. Figure 9 illustrates typical patterns of cumulative flow with time for the Regina Clay and Na-montmorillonite - sand specimens. In the case of the Regina Clay, small flows into the specimen begin at time factors of approximately 1.0 and build to nearly constant flow rates. For the Na-montmorillonite - sand specimens, initial flow was out of the specimen. These flows reverse at a time factor of approximately 1.0 and begin to flow into the specimen, again increasing to essentially fairly constant flow rates.

#### Analyses of results

The test results may be interpreted in light of the theory presented previously, in order to evaluate the role played by osmotic flow (as represented by the osmotic efficiency) and osmotic compressibility in producing volume change.



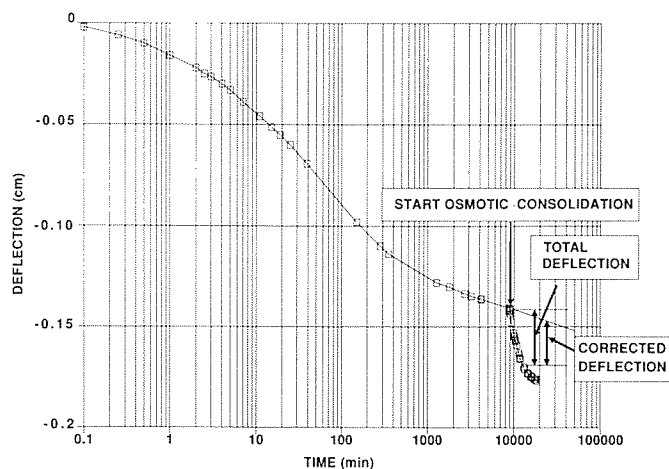


FIG. 5. Effective stress consolidation and osmotic volume change of Regina Clay.

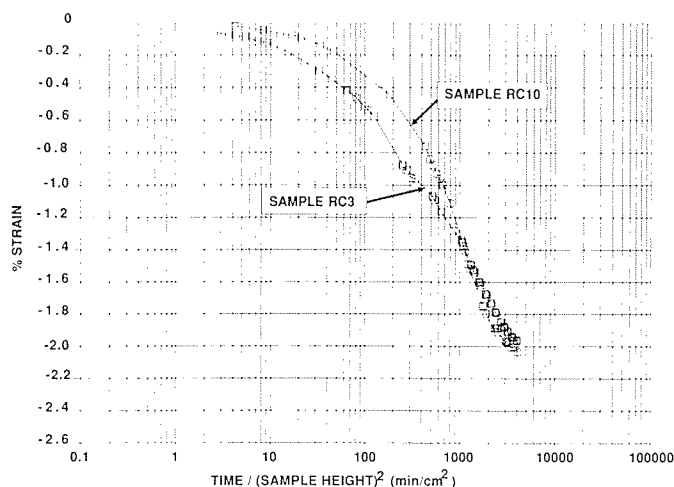


FIG. 6. Time-dependent volume change of Regina Clay by 4.0 M NaCl solution (specimen preconsolidated to 200 kPa).

#### Osmotic efficiency

Nearly constant osmotic flow rates were developed in both soil types after the completion of volume change within the specimen. The osmotic flow rates were observed during exposure to different NaCl solution concentrations. Using [2], the osmotic efficiency of the soils at different confining stresses and different solution concentrations was calculated. These efficiencies are shown in Figs. 10 and 11 along with osmotic efficiencies calculated from the work of Bresler (1973). The data of Bresler require an estimate of the film thickness of the solution between two charged soil particles. This value was obtained using estimates of specific surface, and the equation for interparticle spacing between packets of particles as given by Shainberg *et al.* (1971). With different values of specific surface the lines of efficiency shift to the right or left on the graph.

In spite of considerable scatter, a number of significant observations can be made. First, fairly high efficiencies are obtained at low solution concentrations and low efficiencies are obtained at high solution concentrations. This is consistent with the results described by Kemper and Rollins (1966). Second, the induced negative fluid pressures that would have occurred due to the observed flows are small. These pressures were calculated from [2] using the osmotic

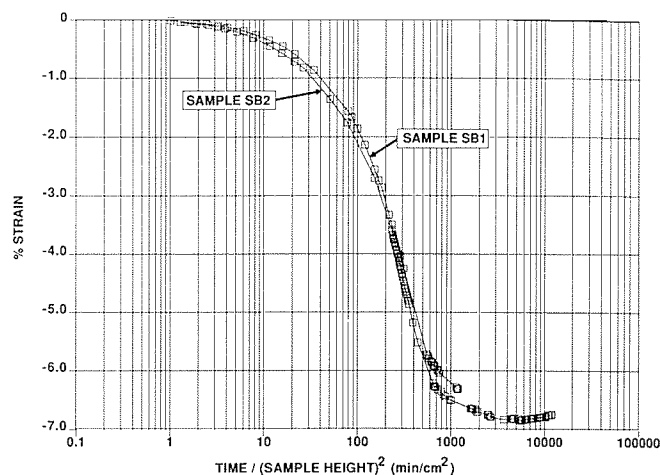


FIG. 7. Time-dependent volume change of sand-montmorillonite by 4.0 M NaCl solution (specimen preconsolidated to 200 kPa).

pressure calculated using [1], the full specimen thickness for  $\Delta x$ , and the measured value of  $k_h$ . Efficiencies of 0.01 at 0.01 M and 0.001 at 1.0 M would only provide a negative pressure response in the range 0.5–5 kPa. This would not be sufficient to account for the magnitude of volume change observed in Fig. 8. Finally, Fig. 10 indicates that for the Regina Clay, the number of particles bound into packets decreases as higher concentrations of NaCl are used. Although there are insufficient data, the pattern in the Na-montmorillonite – sand seems to indicate that the number of particles bound up in packets remains unchanged. Regina Clay is predominately a Ca-montmorillonite; consequently, the clay would be expected to form packets of clay particles bound together by adsorbed  $\text{Ca}^{2+}$  during sedimentation. However, as the pore fluid becomes concentrated these packets would begin to open up because of the dominance of  $\text{Na}^+$  in the double layer, resulting in fewer particles per packet.

#### Osmotic compressibility

The analyses of the osmotic flow and pressure measurements indicate that the predominate mechanism of osmotic volume change for both types of soil cannot be osmotically induced consolidation. However, if osmotic consolidation is the dominant mechanism, then the rate of consolidation will be controlled by the rate of salt transport into the specimen by diffusion.

Steady state diffusion tests were conducted on a single specimen of each material. The combined coefficient of diffusion for  $\text{Na}^+$  and  $\text{Cl}^-$  from tests on both materials was approximately  $4 \times 10^{-6} \text{ cm}^2/\text{s}$ . The coefficient of diffusion from steady state diffusion tests does not take into account the retardation of  $\text{Na}^+$  migration due to adsorption onto the soil solids. In that the Regina Clay is a  $\text{Ca}^{2+}$ -dominated clay, it would be likely that the effective coefficient of diffusion during transient conditions would be lower than that measured in the steady state tests.

This lower rate of diffusion in the Regina Clay samples may in part explain the more rapid changes in volume as a result of osmotic consolidation for the Na-montmorillonite – sand samples (Fig. 7) over those for the Regina Clay samples (Fig. 6). The difference in the time to 100% consolidation for the two soil types is very large, however, and



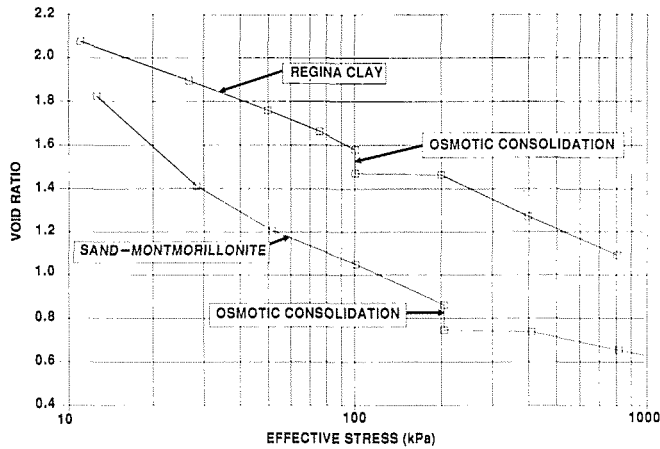


FIG. 8. Influence of osmotic consolidation on the virgin compression branch.

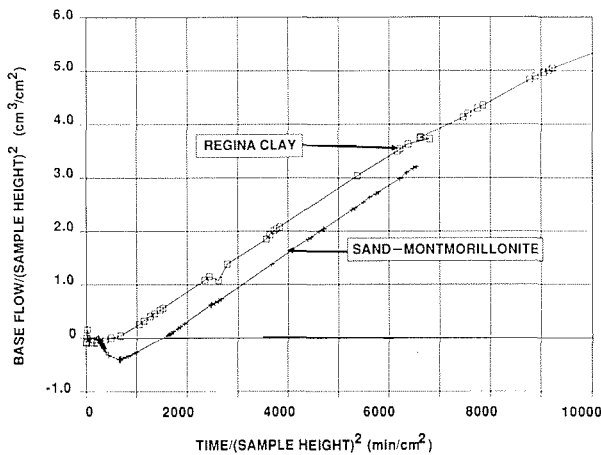


FIG. 9. Cumulative base flow for Regina Clay and sand-montmorillonite exposed to 4.0 M NaCl solution along upper surface (specimen preconsolidated to 200 kPa).

it is unlikely that this difference could be completely explained on the basis of the differences in the effective coefficients of diffusion.

A second explanation for the difference in the rates of osmotic volume change is that the magnitude of the osmotic compressibility of the two samples is substantially different. The osmotic compressibility for each of these materials is illustrated in Fig. 12. Because of the high osmotic compressibility of the Na-montmorillonite - sand mixture at low salt concentrations, a majority of the osmotic volume change within the specimen would have occurred prior to full equalization of salt concentrations within the specimen.

In the case of Regina Clay, the constant value of osmotic compressibility would require nearly full equalization of salt concentrations within the specimen before osmotic consolidation is complete. Consequently, the time to the completion of osmotic consolidation of the Regina clay was much longer than that for the sand-montmorillonite mixture.

The nearly linear relationship between strain and concentration for the Regina Clay does not appear to be consistent with the suppression of the double layer around individual clay particles. This linearity may be due to the dispersion of the clay packets within the Regina Clay as the concentration increases, as was also indicated by the osmotic efficiency data. The shapes of Na-montmorillonite - sand and

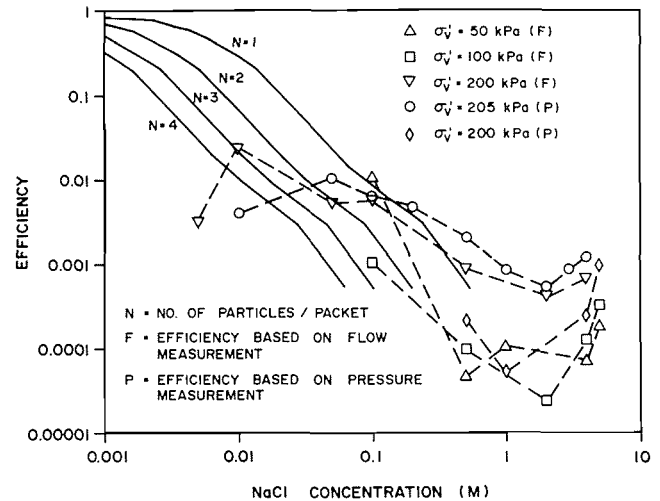


FIG. 10. Theoretical and experimental osmotic efficiencies for Regina Clay.

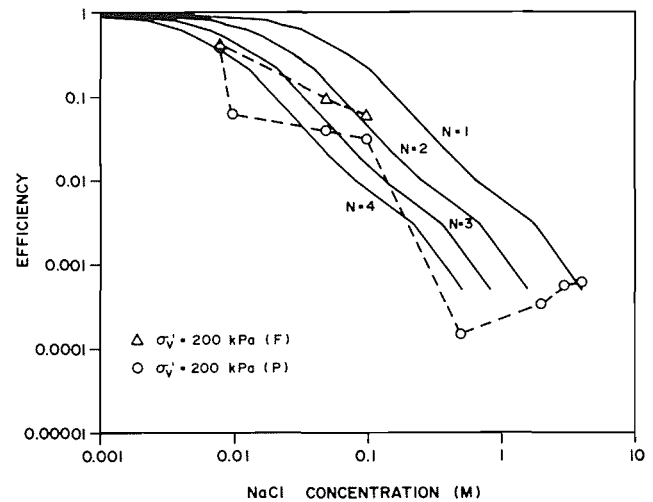


FIG. 11. Theoretical and experimental osmotic efficiencies for sand-montmorillonite.

Regina Clay curves (Fig. 12) are consistent with data described by Aylmore and Quirk (1962) for pure Na-montmorillonite and Ca-montmorillonite, respectively. These authors also attributed the anomalous behavior of the Ca-montmorillonite (similar to the Regina Clay) to the presence of clay packets.

*Consolidation rates*

When osmotic consolidation is the dominant mechanism, the time to 100% consolidation should coincide with the time for 100% equalization of salt concentrations within the specimen by diffusion. The osmotic deflection versus time data for the Regina Clay was analyzed using this assumption, and the coefficient of diffusion for combined Na<sup>+</sup> and Cl<sup>-</sup> was found to range from 1 × 10<sup>-6</sup> to 9 × 10<sup>-6</sup> cm<sup>2</sup>/s with an average of approximately 5.0 × 10<sup>-6</sup> cm<sup>2</sup>/s. Direct steady state diffusion testing was conducted by maintaining different salt concentrations within reservoirs above and below the specimen and measuring the salt flux through the specimen. The results of this testing indicated diffusion coefficients for combined Na<sup>+</sup> and Cl<sup>-</sup> of between 3.6 × 10<sup>-6</sup> and 4.2 × 10<sup>-6</sup> cm<sup>2</sup>/s with an average value of 3.9 × 10<sup>-6</sup> cm<sup>2</sup>/s for the Regina Clay

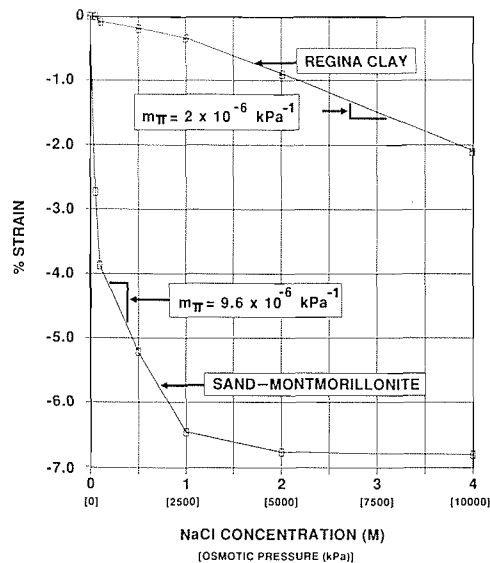


FIG. 12. Osmotic volumetric strain versus solution concentration.

specimens. These values of diffusion were not corrected for the small advective flux of salt due to osmotic flow, which would oppose the diffusive salt flux.

#### Apparent $R - A$ stress

The stress path followed during consolidation, osmotic consolidation, and then reloading, as viewed on the three-dimensional constitutive surface presented previously, is shown in Fig. 13a. When this pathway is viewed on the void ratio versus  $\sigma - u_f$  plane (Fig. 13b), a path different from that presented in Fig. 8 develops. The actual observed behavior suggests that upon reloading, the osmotic pressure of the bulk pore fluid is not constant but is decreasing. In fact, the pore fluid concentration is unchanged during reloading. This behavior is consistent, however, with a view of using  $R - A$  as the stress state variable representing physicochemical interactions. It would be reasonable that upon reloading, small decreases in void ratio would occur, which would produce an increase in the net repulsive stress between the particles as they are forced closer together.

An indirect estimate of the change in the net repulsive stress within the soil mass could be obtained from the void ratio versus stress plots. Figure 14 illustrates a typical virgin branch for the two soil types tested. A second line, below the virgin branch, was drawn by subtracting the volume changes due to osmotic consolidation under a 4.0 M NaCl solution (Fig. 12) from the virgin branch. The difference between these two virgin branches can be taken as a measure of the change in the  $R - A$  stress present in the specimen at a particular void ratio when the soil is exposed to a 4.0 M NaCl solution.

Figure 15 illustrates how the change in the repulsive stress within the two soils varies with confining stress. Also shown is the predicted repulsive stress based on the osmotic pressure concept for the Na-montmorillonite - sand specimens. It is of interest that in the case of the Na-montmorillonite - sand specimens the theoretical values of  $R - A$  agree reasonably well with the measured values when a value of specific surface equal to 580  $\text{m}^2/\text{g}$  is selected. Agreement between the theoretical and measured values for  $R - A$  for Regina Clay was not obtained. Barbour (1987a) indicated that this may

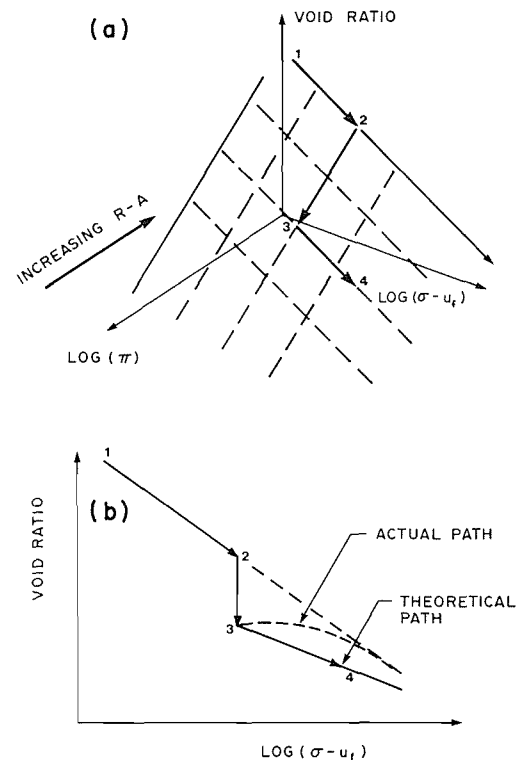


FIG. 13. Stress paths during effective stress and osmotic consolidation: (a) three-dimensional surface; (b) projection onto effective stress axis.

be due to the fact that the clay particles are bound up in packets and do not exist as individual parallel particles, as is assumed with the osmotic pressure concept method of calculating  $R - A$ .

#### Conclusions

The design of waste containment facilities requires that the mechanical and hydraulic properties of the barrier materials be defined. In many instances, alteration of these properties upon permeation with waste fluids will occur. Consequently, a methodology in which to incorporate these changes into design is required.

The theoretical development highlighted in this paper provides for a generalized description of osmotic flow and volume change based on a continuum mechanics approach consistent with existing practice. The volume change behavior of a soil undergoing osmotically induced consolidation and osmotic consolidation can be described using the following properties: osmotic compressibility ( $m_\pi$ ), compressibility ( $m_v$ ), coefficient of permeability ( $k_h$ ), coefficient of osmotic permeability ( $k_\pi$ ), and coefficient of diffusion ( $D$ ) for the dissolved salts. Laboratory tests on modified conventional oedometers can be used to establish the osmotic compressibility and permeability with techniques similar to those used to test for conventional compressibility and permeability.

In the soil specimens tested, the dominant mechanism of volume change associated with brine contamination was osmotic consolidation. This process is characterized by a slow rate of volume change controlled by the rate of transport of the dissolved salt into the specimen. Osmotically induced consolidation was shown to be of little significance with respect to volume change, although osmotic flow did occur.

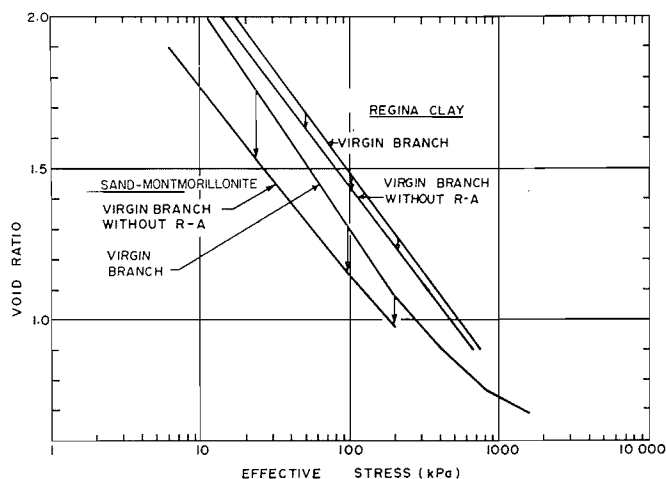


FIG. 14. Indirect evaluation of net  $R - A$  stresses from the virgin curve.

The results of this study provide a comprehensive approach to dealing with the volume change processes that occur during permeation of clay soils with concentrated pore fluids. This understanding can be used to predict the magnitude and rate of associated volume changes. A second possible use of this approach is the incorporation of the effects of changing repulsive stresses within the soil mass into conventional geotechnical analyses through the use of true effective stress and an estimate of  $R - A$ . Barbour (1987b) illustrated the potential use of this approach in analyses of the alteration of the permeability and shear strength of clays after permeation with brine.

The microscopic, mechanistic models such as diffuse double layer theory and the osmotic pressure concept have provided valuable insights into the role that pore fluid chemistry has on soil behavior. Building on this understanding, a macroscopic, phenomenological model provides a way for this understanding to be quantified in design. Description of physicochemical interactions using the same phenomenological framework as conventional soil mechanics would allow existing analyses to include physicochemical interaction in design.

Further work is required to verify the use of "physicochemical" stress state variables and material properties. The present thrust of research into physicochemical interactions has to be expanded from "compatibility" testing of the permeability of clay barriers to general characterization of hydraulic and mechanical behavior of clay soils within a verifiable, theoretical phenomenological framework.

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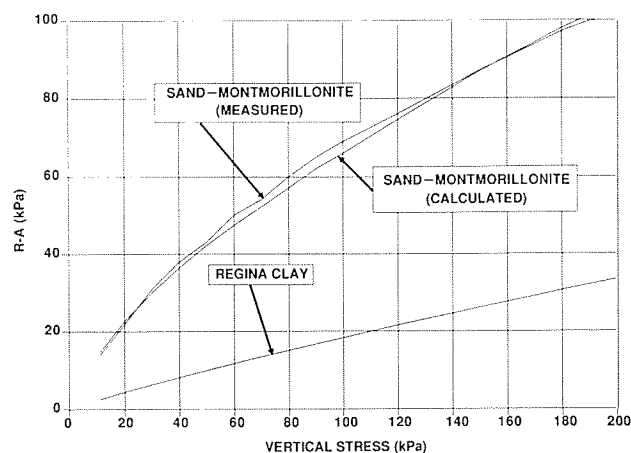


FIG. 15.  $R - A$  versus vertical effective stress.

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