MECHANISMS CONTROLLING THE PERMEABILITY OF CLAYS

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Abstract—Coefficients of permeability, calculated using Terzaghi's theory of one-dimensional consolidation, are reported for smectite, illite, and kaolinite, in water, methyl and ethyl alcohol, benzene, and carbontetrachloride. When the pore fluid was water the clays were homoionized to either the sodium or calcium form and the pore water electrolyte concentration was varied. The coefficients of permeability are evaluated in terms of both mechanical and physico-chemical variables. It appears that the coefficients of permeability are mainly influenced by mechanical effects, particularly the distribution of void sizes and the tortuosity of the channels. The coefficient of permeability is maximized if the flow channels consist of many small channels and a relatively few large ones, through which the main flow occurs. Physico-chemical variables exert great influence on the coefficient of permeability through their influence on dispersion or aggregation of the clay particles.

INTRODUCTION

ON A MICROSCOPIC scale the flow of fluids in a porous medium is governed by three equations, viz. the equation of continuity, the equation of state, and the dynamical equation of motion (Lamb, 1932). Unfortunately, the mathematical difficulties involved in solving these equations, and the difficulties involved in prescribing the nature of the flow channels and the interactions between the fluid and the porous media, are such that the classical approach to solving problems involving flow in porous media has not proved useful.

Instead, flow is considered on a macroscopic scale. Based on experiments, Darcy (1856) formulated a macroscopic flow equation which may be written:

$$v = -k \frac{dh}{ds} \qquad (1)$$

where v is the discharge velocity, k is the coefficient of permeability (often termed the conductivity in the scientific literature), h is the total head, and s is the distance of macroscopic flow measured in the direction of flow.

In consideration of its widespread use there have been numerous studies of Darcy's equation. These studies have been of two types: (1) studies of the general form of the equation, and (2) studies of the fundamental factors controlling the constant. This paper is concerned with the constant.

Numerous attempts have been made to study the coefficient of permeability of granular media through the use of capillary (Scheidigger, 1953) or hydraulic radius (Kozeny, 1927; Carman, 1941, 1948; Leibenzon, 1947) models. These analyses were simplified by the assumption of no chemical interaction between the fluid and the porous media. In contrast, there have been few attempts to develop suitable models for clays although the permeability of clays has been discussed extensively.

Terzaghi (1925) emphasized the importance of the nonuniformity of the voids on clay permeability and the substantial dependence of the permeability on the void ratio. He considered that the physical properties of the pore water changed in the immediate vicinity of the clay surfaces.

Macey (1942) measured the coefficient of permeability of clays in water and in nonpolar fluids. He reported rates of flow for benzene 100,000 to 1,000,000 times greater than for water in the same clay. He considered that particle spacing, particle size as influenced by aggregation or dispersion, particle arrangement, adsorbed layers, and interlamellar swelling all influenced the permeability but he considered the most important cause for the lower permeability in water to be the anomolous viscosity of the water near the clay surfaces.

Grace (1953) demonstrated that improved dispersion is the main reason for the marked reduction of permeability caused by use of certain electro-

lytes and he pointed out the importance of particle reorientation as consolidation progressed.

Michaels and Lin (1954) showed experimentally that the permeability of kaolinite decreased markedly as the polarity of the permeating fluid increased and that the most important factor controlling the permeability of kaolinite was the degree of dispersion of the kaolinite in the original suspending fluid. They also concluded (see also Michaels and Lin, 1955) that electroosmotic counterflow might be of some importance when aqueous solutions were used as the permeants but that such colloidal effects as adsorbed liquid surface films appeared to have little effect on the permeability.

Olsen (1960) measured hydraulic flow rates, electrical conductivities, and streaming potentials for flow through kaolinite, illite, and Boston blue clay, and concluded that electro-osmotic counterflow, high viscosity, and tortuous flow paths failed to account for the permeability characteristics he measured. He concluded that unequal pore sizes was the most important variable.

The foregoing studies and others as well, indicate that the most important variable influencing the permeability of clays is flocculation of the clay particles, which influences the distribution of void sizes and shapes.

In the discussion to follow, values for the coefficients of permeability of kaolinite, illite, and smec-tite are reported as functions of void ratio and the type of pore fluid, and mechanisms controlling the permeability are discussed.

EXPERIMENTAL AND ANALYTICAL PROCEDURES

The experimental and analytical procedures have been presented in detail by Mesri (1969) and reviewed by Olson and Mesri (1970). In brief, homoionic slurries of kaolinite, illite, and smectite (Table 1) in water were prepared by repeated washing with concentrated solutions of NaCl or CaCl$_2$. Slurries in other fluids were prepared by washing air dried clay in ethyl alcohol, centrifuging, drying at 110°C, and then mixing with the desired fluid. The slurries were consolidated to pressures ranging from 90 to 125 psf in special sedimentation tubes (Mesri, 1969) and then transferred to 2·5-in. dia. consolidation rings where they were consolidated in increments to a pressure of 64,000 psf using a pressure ratio of two.

The coefficients of permeability were calculated by fitting Terzaghi's theory of consolidation (Terzaghi, 1943) to the observed laboratory time-settlement observations and extracting the coefficient of permeability from the calculated coefficient of consolidation. The fitting operation utilized the logarithmic method (Casagrande and Fadum, 1940) for all tests in which the pore fluid was water or alcohol. However, specimens in which the pore

Table 1. Properties of clays

Mineral (1)	Supplier source trade name (2)	Liquid limit in per cent (3)	Plastic limit in per cent (4)	Specific gravity (5)	Surface area in m² per g (6)	Cation exchange capacity m-equil. per 100 g (7)	Fraction finer than 0·002 (8)	Estimated ratio of dia. to thickness (9)
Kaolinite	Minerals and Chemicals Philipp Corp., Klondyke clay, McIntyre, Georgia	40 to 50	27 to 31	2·65	14	2·2	47	2–5
Illite	J. L. Eades, Dept. of Geology Univ. of Illinois, Marblehead, Wisconsin	83 to 104	31 to 32	2·80		28	100	10–50
Smectite	American Colloid Company, Wyoming, Volclay	190 to 1160	31 to 47	2·65 to 2·80	500 to 700	100	97	150–500

fluid was non-polar often underwent 50 per cent or more of their settlement during the first 6 sec after loading, thus precluding use of the logarithmic method. For these specimens the square root fitting method (Taylor, 1948) was used. In our experience, the standard square root method, in which fitting is performed at an average degree of consolidation of 90 per cent, usually yields coefficients of permeability that are higher than those calculated by either method when fitting is performed at 50 per cent consolidation. However, the coefficients of permeability in non-polar fluids were so much higher than in other fluids that a slight downwards adjustment in the data would have no effect on the interpretation.

A question may be raised as to how the coefficients of permeability calculated using Terzaghi's theory compare with values measured directly. Terzaghi (1923) made such comparisons when he first developed the theory; he found satisfactory agreement. Casagrande and Fadum (1944) reported that they always found satisfactory agreement provided the logarithmic fitting method was used and provided that there was a distinct change in curvature when the primary settlement curve merged with the secondary settlement curve. Taylor (1942) presented comparisons for remolded specimens of Boston Blue clay, based on the square root fitting method, and showed that the measured coefficients of permeability generally exceeded the calculated values. He attributed this difference in permeabilities to Terzaghi's assumption that the sole cause of delay in compression is the time required for the water to be squeezed out, i.e. t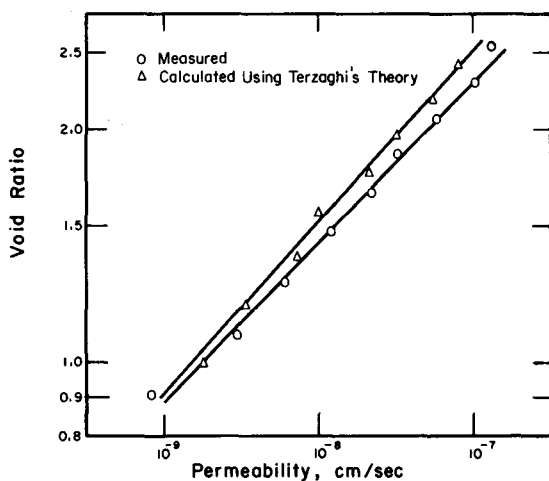o the permeability of the clay. Taylor concluded that the structure of clay itself possessed a time dependent resistance to compression so that the total resistance to volume change came partly from permeability and partly from the structural resistance of the clay itself. By attributing all of the resistance to low permeability, Terzaghi's theory must inevitably lead to an under-estimate of the permeability. It has been our experience, based on a number of comparisons between measured and computed coefficients of permeability on both remolded and undisturbed clays, that the calculated coefficients of permeability are low by only about 5–20 per cent provided that the clay is normally consolidated at the time of determination. All clays used in this investigation were normally consolidated. A comparison of measured and computed coefficients of permeability, which is typical of numerous other such comparisons, is shown in Fig. 1.*

EXPERIMENTAL RESULTS AND DISCUSSION

The coefficients of permeability of kaolinite, illite, and smectite, are shown as functions of void ratio in Figs. 2–4, respectively. For the cases where water was the pore fluid, all data are indicated without regard to the electrolyte concentration. The scatter in data for the sodium forms of the illite and smectite is mainly caused by the variations in electrolyte concentration, a variable that will be discussed subsequently.
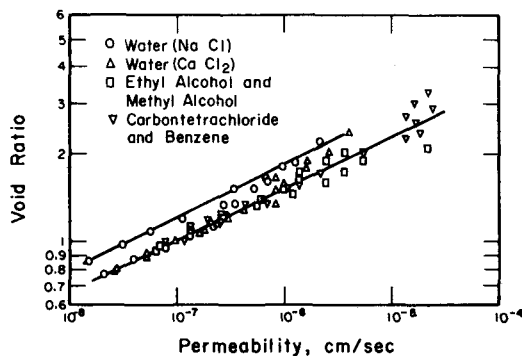
Fig. 2. Coefficients of permeability of kaolinite.

In discussing the data in Figs. 2–4, it is convenient to separate the variables that influence the coefficients of permeability into two broad and overlapping groups, viz. mechanical variables and physico-chemical variables.

Fig. 1. Coefficients of permeability of a 0·01 N calcium illite.

*The illite used in this comparison came from Fithian, Illinois, and has slightly different properties from the illite used in the main series of experiments to be reported in this paper.
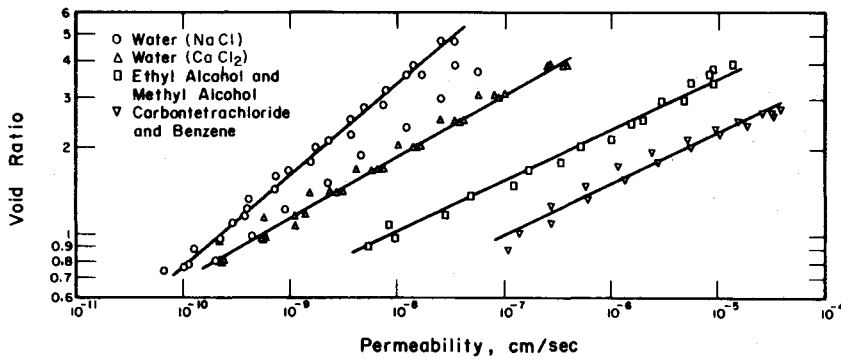
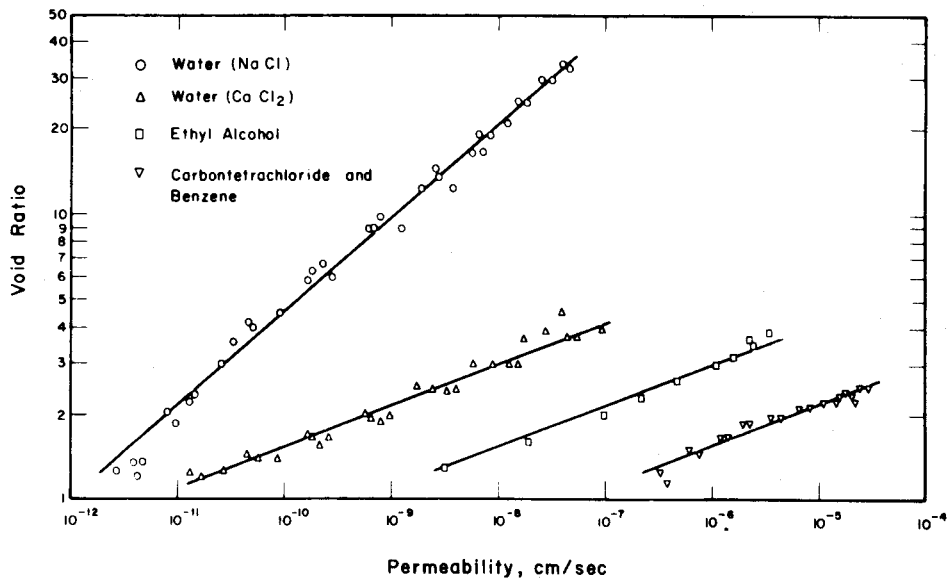Fig. 3. Coefficients of permeability of illite.

Fig. 4. Coefficients of permeability of smectite.

Mechanical variables

Various mechanical variables may be used but perhaps the most convenient are the size, shape, and geometrical arrangement of the clay particles. These variables in turn determine the geometrical properties of the pore system.

The smaller the particle size the smaller the size of the individual flow channels, all other variables being constant. The simplest theoretical models of porous media (Lamb, 1932) show that the coefficient of permeability is directly proportional to the second power of the diameter of the flow channel. The great range in size of the flow channels is one reason for the large differences in the coefficient of permeability of the three clay minerals. The smectite has the smallest particle size and the kaol-

inite the largest. The coefficients of permeability of the three clays, in the sodium form and in water are shown in Fig. 5. The illite is about 200 times more pervious than the smectite, and the kaolinite is about 200,000 times more pervious, at the same void ratio. Variables other than channel size also contribute to the differences in Fig. 5 as will be discussed subsequently.

The geometry of the flow channel is a function of the shape and orientation of the clay particles. For the platey clay particles, application of pressure, under fully drained conditions and without lateral strain, results in an orientation of the plates normal to the direction of maximum principal stress (Lambe, 1953; Norrish, 1954; Emerson, 1956; Mitchell, 1956; Quigley and Thompson, 1966)
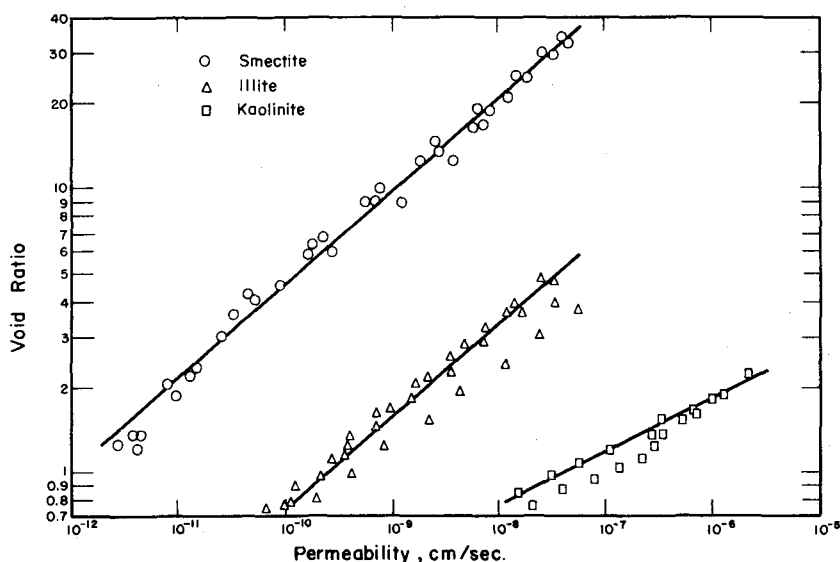
Fig. 5. Coefficient of permeability of all three sodium clays in water.

and leads to an increasingly tortuous flow path in the direction of the applied pressure. This particle orientation, or tortuosity effect, is larger for particles with larger diameter to thickness ratios. The diameter to thickness ratio (Table 1) increases from kaolinite to illite to smectite and is another source of the difference in the coefficients of permeability of the three clays (Fig. 5). Also, particle orientation increases with increasing consolidation pressure (decreasing void ratio) causing a reduction in permeability with reduction in void ratio. Of course, the coefficient of permeability also decreases with void ratio simply because of the reduction in total void space.

Physico-chemical variables

The data in Figs. 2–4 indicate that the coefficients of permeability are largest for nonpolar fluids, smaller for polar fluids of low dielectric constant, and lowest for water, which is polar and has a high dielectric constant. Further, the coefficient of permeability is generally lower when the adsorbed cations are monovalent rather than divalent. Generally, a reduction in electrolyte concentration (Fig. 6) tends to reduce the coefficient of permeability but the effect of electrolyte concentration diminishes as the valency of the cations increases, and is smaller in sequence from smectite to illite to kaolinite. It is convenient to consider these effects as physico-chemical.

The physico-chemical variables may be taken as the surface charge density and distribution, valency of the adsorbed cations, and such pro-perties of the fluid as dielectric constant, dipole moment, and viscosity. Other variables could be chosen and some of the above variables, e.g. viscosity of the fluid, could just as well be considered mechanical variables.

The existence of a net negative charge within the clay particles leads to the adsorption of cations. For pore fluids of low dielectric constant, such as the alcohols, benzene, and carbon tetrachloride used in this study, the cations are held tightly against the surface of the particles and have a minimal tendency to block the flow channels. Further, when nonpolar fluids are used, there is little tendency for the fluid to be adsorbed by either the cations or the surface and the flow channels are almost completely open. Thus, the coefficient of permeability should be largest for benzene and carbon tetrachloride (Figs. 2–4). When a fluid such as an alcohol is used, the fluid is adsorbed either by the cations or, more likely, by the formation of hydrogen bonds with the surface, and a part of the flow channels is likely to be blocked, thus reducing the permeability (Figs. 2–4). However, these effects seem clearly inadequate to explain the very large differences between the coefficients of permeability of the smectite and illite in nonpolar pore fluids, in alcohols, and in water. Further, these effects do not explain the fact (Fig. 7) that all of the clays yielded the same void ratio vs. permeability relationship when nonpolar fluids were used.

Since there is a negligible adsorption of the non-polar fluids and no formation of diffuse double
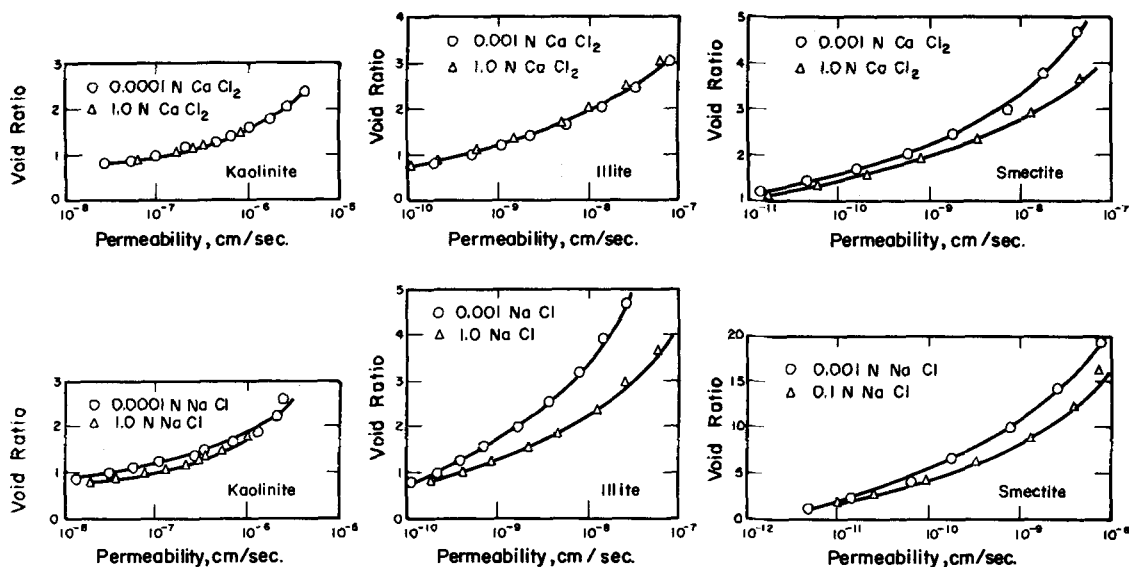
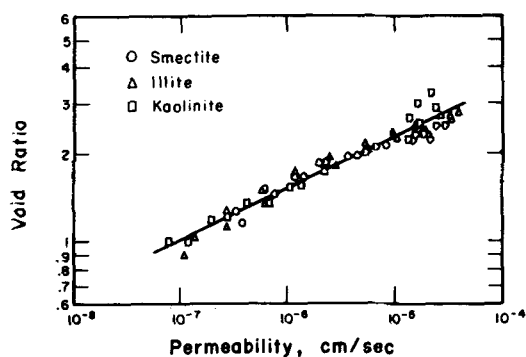Fig. 6. Coefficients of permeability of all three clay minerals in water.

Fig. 7. Coefficients of permeability of all three clay minerals in nonpolar fluids.

layers, the mechanisms controlling the coefficients of permeability of the clays in nonpolar fluids should be mechanical. The existence of essentially the same permeability for the three minerals (Fig. 7) then implies the existence of voids of similar size and shape. Apparently the particles of illite and smectite formed stable aggregates, and the main fluid flow took place in the large voids between the aggregates. Thus, for a constant void ratio, the channels between these aggregates would be considerably larger in size than channels between evenly spaced particles and the rate of flow would be expected to increase with some power of the channel diameters. The main effect of using alcohol or water as a permeant is then to break down the aggregates, thus reducing the size of the largest

flow channels at a constant void ratio, and reducing the coefficient of permeability. The existence of large aggregates was previously used to explain the swelling characteristics of the same clays (Olson and Mesri, 1970).

The clay particles in aggregates in nonpolar fluids are believed to be randomly arranged and the aggregates are probably more-or-less spherical in shape. When the pore fluid is water, it is believed that aggregates of parallel particles, termed domains (Emerson, 1959; Aylmore and Quirk, 1959), are formed for smectite and possibly for illite as well. In the case of calcium smectite the water is adsorbed between the layers until the basal spacing is about 19 Å (Norrish, 1954; Senich, Demirel and Handy, 1967) and then adsorption stops. Since the layers cannot swell sufficiently to form diffuse double layers, it is believed that domains of perhaps eight to ten layers form (Blackmore and Miller, 1961) with larger voids between the domains. The fluid trapped within the domains is probably essentially immobile until high pressures are applied. Thus, the fluid flows mainly through the larger interdomainal voids. The calcium smectite in water is more dispersed than the smectite in alcohol, but because of the presence of domains is less dispersed than the sodium smectite in water, and thus has intermediate coefficients of permeability. A similar argument apparently may be used with the illite. The kaolinite particles are too large and too nearly equidimensional to form domains that influence the permeabilities significantly.

For calcium clays a diffuse double layer probably

forms on the exposed surfaces of clay particles (Norrish, 1954) but in the case of kaolinite and illite the void spaces are so large that the diffuse double layers do not block the main flow channels significantly. Thus, changes in double layer thickness associated with variations in pore water electrolyte concentration do not influence the permeability by measurable amounts (Fig. 6). However, in the case of calcium smectite with its much smaller void spaces, the apparent double layer thickness influences permeability significantly (Fig. 6). Further, in the case of calcium smectite, it is probable that thicker domains form when the electrolyte concentration reaches about 1N so, for a given void ratio, the interdomainal voids are larger and a higher permeability results.

For a sodium clay in water, diffuse double layers form and the clay disperses. The lower the electrolyte concentration the greater is the tendency of the clay to disperse uniformly through the available space without the occurrence of any larger voids. Thus, electrolyte concentration, through its influence on dispersion, influences the coefficient of permeability.

SUMMARY AND CONCLUSIONS

The coefficients of permeability of clays are controlled by variables that may be classified as mechanical and physico-chemical. The mechanical variables of main interest are the size, shape, and the geometrical arrangement of the clay particles. The reduction in the coefficient of permeability, at constant void ratio, from kaolinite to illite to smectite is largely the result of a reduction in the size of individual flow channels and an increase in the tortuosity of the flow paths.

Physico-chemical variables exert great influence on the coefficient of permeability by controlling the tendency of the clay to disperse or to form aggregates. Aggregation leads to the existence of many tiny flow channels through which there is likely to be little flow, and a smaller number of relatively large channels through which the main flow occurs. Dispersion leads to channels that are all of nearly the same size, and tend to be nonequidimensional, and thus reduces fluid flow.

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Résumé — Les coefficients de perméabilité calculés à l'aide de la théorie de la consolidation unidimensionelle de Terzaghi sont donnés pour une smectite, une illite et une kaolinite en présence d'eau, d'alcool méthylique, d'alcool éthylique, de benzène et de tétrachlorure de carbone. Lorsque le fluide remplissant les pores est l'eau, les argiles ont été rendues homoioniques, soit sous forme sodium, soit sous forme calcium et l'on a fait varier les concentrations en électrolyte de la solution des pores. Les coefficients de perméabilité ont été évalués à la fois en terme de variables mécaniques et physicochimiques. Il apparaît que les coefficients de perméabilité sont essentiellement influencés par les effets mécaniques, en particulier, la distribution des dimensions des vides et la tortuosité des pores. Le coefficient de perméabilité passe par un maximum lorsque les pores assurant l'écoulement sont constitués par de nombreux petits canaux et relativement peu de grands, à travers lesquels la plus grande part du débit s'établit. Les variables physico-chimiques exercent une grande influence sur le coefficient de perméabilité par le biais du rôle qu'elles jouent dans la dispersion et l'agrégation des particules d'argile.

Kurzreferat — Es wird über Permeabilitätskoeffizienten, berechnet unter Verwendung der Theorie von Terzaghi über eindimensionale Verdichtung, für Smectit, Illit und Kaolinit in Wasser, Methyl- und Äthylalkohol, Benzol und Tetrachlorkohlenstoff berichtet. Wenn die Porenflüssigkeit Wasser war wurden die Tone entweder in die Natrium-oder die Calciumform homoionisiert und die Elektrolytkonzentration des Porenwassers wurde variiert. Die Permeabilitätskoeffizienten wurden mit Hilfe mechanischer sowie physikalisch-chemischer Variabler eingeschätzt. Es scheint, dass die Permeabilitätskoeffizienten in erster Linie durch mechanische Wirkungen beeinflusst werden, besonders die Verteilung von Hohlraumgrössen und die Gewundenheit der Kanäle. Der Permeabilitätskoeffizient wird maximiert wenn die Strömungskanäle aus vielen kleinen Kanälen und verhältnismässig wenigen weiten bestehen, durch welche die Hauptströmung stattfindet. Physikalisch-chemische Variable üben infolge ihrer Wirkung auf die Dispersion oder Zusammenballung der Tonteilchen einen beträchtlichen Einfluss auf den Permeabilitätskoeffizienten aus.

Резюме — Приведены коэффициенты проницаемости для смектита, иллита и каолинита (по отношению к воде, метиловому и этиловому спиртам, бензолу и четыреххлористому углероду), подсчитанные в соответствии с теорией одномерной консолидации Терцаги. В тех случаях, когда наполнителем пор служила вода, глины гомоионизировались до натриевой или кальциевой форм, и концентрация водного электролита пор изменялась. Коэффициент проницаемости оценивался как в отношении механических, так и в отношении физико-химических переменных. Как оказалось, на коэффициент проницаемости влияют, главным образом, механические эффекты, и в особенности — характер распределения пор по размерам и степень извилистости каналов. Коэффициент проницаемости достигает максимума в тех случаях, когда каналы, по которым проходит поток жидкости, состоят из множества мелких каналов и сравнительно небольшого числа крупных. Физико-химические переменные оказывают влияние на коэффициент проницаемости лишь в той степени, в которой они влияют на дисперсию или агрегацию глинистых частиц.