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The Effect of Surface-Ion Attractive Forces on the Permeability of Bentonite

L'Effet des forces d'attraction "surface-ion" sur la perméabilité du bentonite

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SUMMARY

One-dimensional consolidation tests performed on commercial bentonite (Volclay) revealed that at certain pressures the test samples reached equilibrium at smaller interparticle distances than would be expected from consideration of the parallel-plate model and osmotic theory. Further tests on sodium bentonite, prepared from Volclay, confirmed that, at interparticle distances of the order of 50 Å, surface-ion attractive forces came into play tending to draw the plates together, with a corresponding expulsion of water from the saturated sample, until, at spacings of about 15–20 Å, the hydration and image forces between the platelets reversed the action.

SOMMAIRE

Des essais de consolidation unidimensionnelle effectués sur du bentonite de commerce (Volclay) ont montré qu'à certaines pressions les échantillons d'essai atteignaient l'équilibre à de moindres distances interparticulaires que n'aurait laissé prévoir la considération du modèle à paillettes parallèles et la théorie de l'osmose. De nouveaux essais sur bentonite de sodium, préparés à partir du Volclay, ont confirmé qu'à des distances interparticulaires de l'ordre de 50 Å, sont entrées en jeu des forces d'attraction tendant à attirer les ions vers les surfaces et, par conséquent, à rapprocher les paillettes, avec expulsion correspondante de l'eau contenue dans l'échantillon saturé, jusqu'au moment où, à des distances d'environ 15 à 20 Å, les forces d'hydratation et d'image entre les paillettes ont renversé l'action.

ONE-DIMENSIONAL CONSOLIDATION TESTS, performed on commercial bentonite as part of an investigation into the thixotropic characteristics of clays, showed that the large amount of compression undergone by the bentonite destroyed any possible structural changes resulting from thixotropic action, but were significant in revealing certain unexpected structural characteristics. To clarify the action, some of the tests were repeated on sodium bentonite. These tests revealed an apparent spontaneous collapse of the structure of the bentonite at small interparticle spacings.

PRELIMINARY TESTS ON COMMERCIAL BENTONITE (VOLCLAY)

Physical properties of the bentonite used are given in Data Sheet 202 issued by the American Colloid Company.

Two groups of samples were prepared. One group, at a liquidity index of approximately 1.0 ($w = 600\%$), was used in studies extending over sixteen weeks. The liquidity index of the second group varied from approximately 0.6 to 0.9 (Table I). Samples of the first group were prepared by remoulding with a spatula and placing in containers which

were then sealed and stored under water to prevent loss of moisture. Duplicate samples were then tested at periods of 0, 2, 6, and 16 weeks after remoulding. Individual samples of the second group were prepared in a similar manner and tested immediately after remoulding.

The results of these compression tests are presented in the standard manner in Figs. 1(a) and 1(b) and it will be seen that the time of storage had no effect on the compression characteristics of the bentonite. (Point M in Fig. 1(a) represents incomplete compression for one specimen, as the succeeding load increment was added before the specimen had reached 100 per cent of theoretical primary consolidation.) From the results of these compression tests the coefficient of permeability of the bentonite was calculated for each load increment, a mean value being taken for the eight tests of the first group (LI \approx 1.0). Fig. 2 shows that there is generally an exponential relationship of the form $k = ae^b$ between the void ratio e and the coefficient of permeability k , but, as will be seen from Table I, it is difficult to establish the trend of the values of the parameters a and b with varying initial moisture content.

An examination of Fig. 2 reveals that in every case the value of k corresponding to the final load increment ($e = 1.8$) appears to be anomalous (to avoid confusion only the test values for LI = 0.6 have been plotted *in toto* but all other results were no less consistent) and in order to investigate this anomaly it was decided to examine the compression curves from the aspect of osmotic theory as outlined by Bolt (1956). For this purpose a theoretical compression curve for Volclay was plotted (Fig. 1), using a median valency of 1.25 for the exchangeable cations. On this basis, and using the concept of an osmotic activity factor as outlined by Ruiz (1961), Fig. 3 was prepared. In simple terms, the activity factor $f(i)$, as plotted, is merely the ratio

TABLE I. INITIAL WATER CONTENT AND COEFFICIENT OF PERMEABILITY FOR SOILS USED IN COMPRESSION TESTS

Material	Number of samples	Identification (liquidity index)	Initial moisture content (per cent)	Coefficient of permeability (cm/sec $\times 10^{-13}$)
Natural Volclay	1	≈ 0.6	378	$3.32e^{1.16}$
Natural Volclay	1	≈ 0.7	413	$3.56e^{1.34}$
Natural Volclay	1	≈ 0.8	458	$4.52e^{1.08}$
Natural Volclay	1	≈ 0.9	548	$3.35e^{1.09}$
Natural Volclay	8	≈ 1.0	603	$4.08e^{1.02}$
Sodium bentonite	2	—	692	

of the applied equilibrium pressure to the theoretical equilibrium pressure at any void ratio. Assuming parallel-plate configuration and full saturation, the void ratio corresponds to a definite interparticle spacing ($2d$ Å).

It will be seen from Fig. 3 that, for high values of liquidity index, the curves show a characteristic "S" shape. The trough at low values of d , corresponding to an applied pressure of 6.63 kg/sq.cm., represents a lower void ratio at equilibrium than would theoretically be expected. In each case the maximum value of $f(i)$ (at the crest of the "S") occurred at interparticle spacings of the order of 40 Å, the minimum being at about 20 Å. Since a decreasing value of $f(i)$ indicates a smaller than normal interparticle separation it

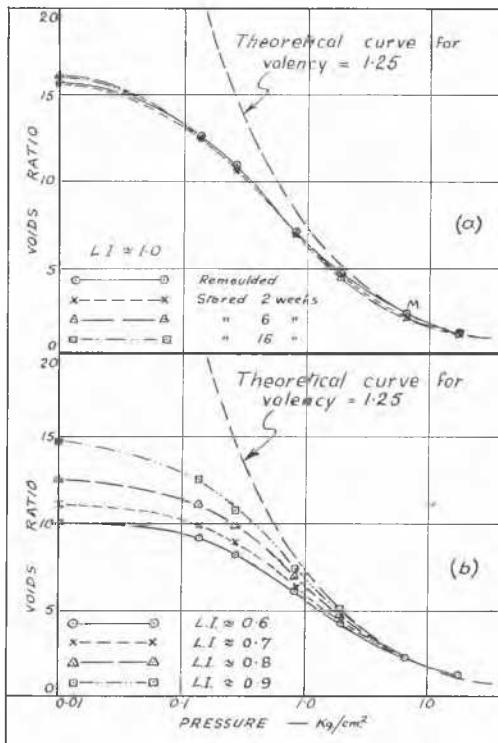


FIG. 1. Consolidation characteristics of Volclay bentonite.

is apparent that, at this stage of the compression cycle, some force comes into play tending to draw the plates together. Since the tests were not designed specifically to evaluate the interparticle forces it was felt that no purpose would be served by attempting to analyse the results obtained in too great detail. The specific position of the theoretical curves shown in Fig. 1 and of the activity factor curves in Fig. 3 was therefore regarded as being of only general significance and it was the shape of the activity factor curves which merited further study. Complete decompression curves were not established since they were not relevant to the original research project.

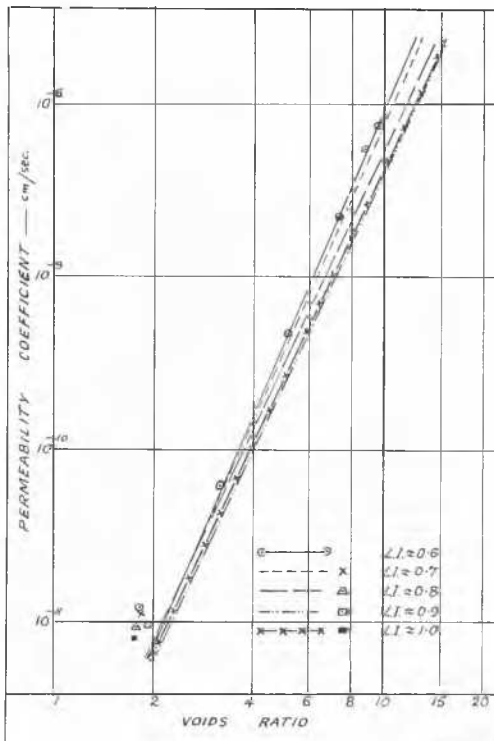


FIG. 2. Permeability - void ratio relationship for Volclay bentonite.

TESTS ON SODIUM BENTONITE

In order to investigate the problem more rigorously, within the limits of the equipment available, a sample of sodium bentonite was prepared from Volclay by repeated washing with $\frac{1}{2}N$.NaCl, followed by washing with distilled water through dialysis tubing. Washing was continued for two weeks after a silver nitrate test revealed no chloride ions present and it is considered that the free electrolyte content of the sodium bentonite was effectively zero.

Because of the limited time available and lack of facilities for separating conveniently and accurately the minus 0.2 micron fraction, it was decided to use the prepared sodium bentonite without further treatment. One-dimensional consolidation tests were performed on two identical samples of this material, using a greater number of smaller load increments up to the same maximum value of 17.92 kg/sq.cm. The samples were prepared in the same manner as in the previous tests and were placed in the oedometers at the moisture content existing after the washing (approx. 690 per cent). At this figure the material would be, to all intents and purposes, completely dispersed.

Fig. 4 shows the conventional e vs. $\log p$ curves for these two tests and the deviation of the actual curves from the theoretical in the vicinity of 10 kg/sq.cm. can clearly be seen. (The reason for the vertical shift between the two experimental curves is obscure.)

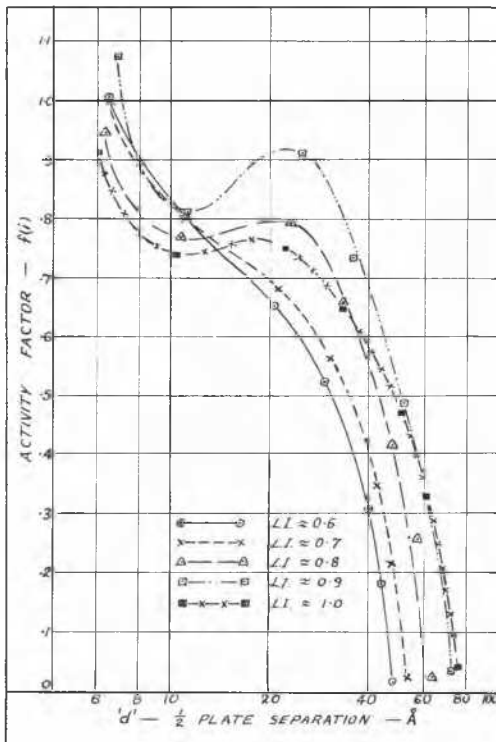


FIG. 3. Activity factor - interparticle spacing relationship for Volclay bentonite.

Fig. 5 shows the relationship between the coefficient of permeability and the void ratio for the sodium bentonite and in each case the deviation of the straight lines at a void ratio of 5.6 (corresponding to a value of $2d \approx 54 \text{ \AA}$) is clearly apparent. The expression for the upper section of the graph, based on an average value is $k = 2.48 e^{3.75} \times 10^{-13} \text{ cm/sec}$; that for the average of the lower sections is $k = 9.41 e^{2.08} \times 10^{-13} \text{ cm/sec}$.

In Fig. 6 the full line curves show the activity factor, $f(i)$, plotted against interparticle spacing and the same "S" shape is again evident, this time with a crest at about $2d = 50-60 \text{ \AA}$ and a trough at about $15-20 \text{ \AA}$. Since, as is shown below, the apparently increased permeability of the samples at lower void ratios is equivalent to an excess pressure increment for the same change in void ratio, this has been used to calculate an apparent activity factor $f'(i)$ which is plotted in Fig. 6 with broken lines. It will be noticed that, in each case, when the interparticle spacing is reduced to approximately $2d = 60 \text{ \AA}$, the apparent activity factor $f'(i)$ reaches a value which remains more or less constant until the upward trend begins at 20 \AA . The significance of this is outlined in the discussion which follows.

THEORETICAL CONSIDERATIONS

For the bentonite the value of γ_s has been taken at 2.59, as recommended by Quirk, and the specific surface has been

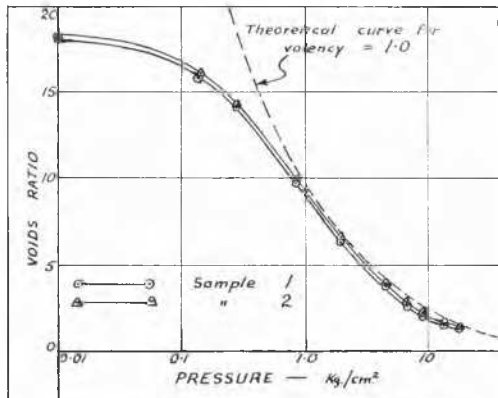


FIG. 4. Consolidation characteristics of sodium bentonite.

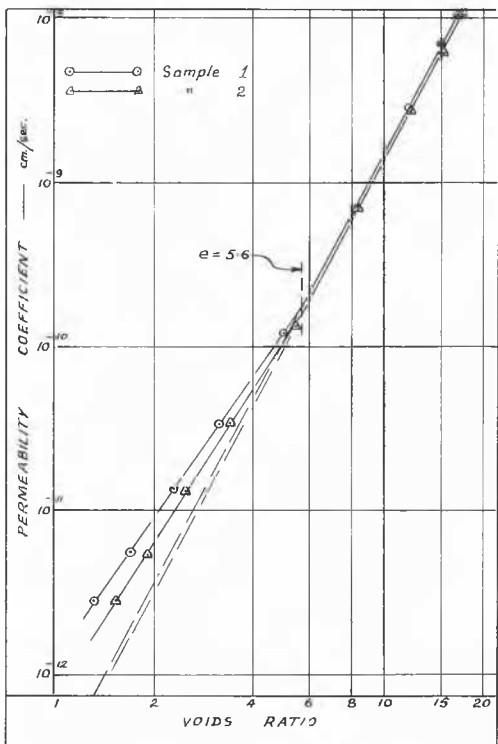


FIG. 5. Permeability - void ratio relationship for sodium bentonite.

taken as 800 sq.m./gram as given by Bolt. The coefficient of permeability was calculated using the expression

$$k = \gamma_v C_v (e_0 - e_{100}) / [1 + e_0] \Delta P \quad (1)$$

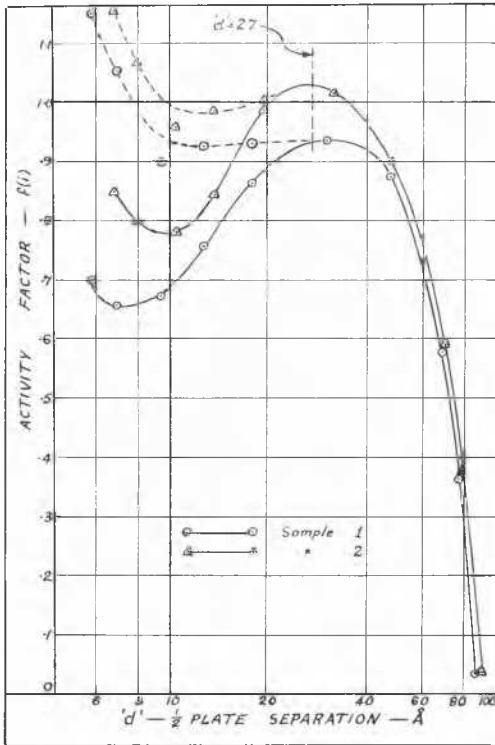


FIG. 6. Activity factor - interparticle spacing relationship for sodium bentonite.

and c_v , e_0 , and e_{100} were obtained using the modified Naylor and Doran method (Arnold, 1952).

Values of k were plotted against the mean void ratio (e_{10}) for each load increment and standard regression methods were used in determining the parameters of the best-fitting straight line, the correlation coefficient in every case being not less than 0.998.

Theoretical compression curves were computed using the van't Hoff equation

$$P_s = RTC_0 \left(\frac{C_r}{C_0} + \frac{C_0}{C_c} - 2 \right) \text{ atm} \quad (2)$$

and the interparticle spacing equation as given by Bolt

$$v \sqrt{\beta C_0} (x_0 + d) = 2 \sqrt{\frac{C_0}{C_c}} \times \int_0^{\phi} d\phi / \sqrt{\left[1 - \left(\frac{C_0}{C_c} \right)^2 \sin^2 \phi \right]} \quad (3)$$

Using these expressions, the following expression for the activity factor, $f(i)$, was developed:

$$f(i) = P_a / P_s = 4.05 \left[4 + (ev/\gamma_s S) \times 10^8 \right]^2 P_n \times 10^{-1} \quad (4)$$

where P_a = applied pressure in kg/sq.cm. and P_s = theoretical swelling pressure in kg/sq.cm.

Assuming that a unique relationship between permeability coefficient and void ratio should hold throughout the complete range of void ratios (as shown by the broken lines of Fig. 5) the excess permeability represented by the decreased slope shown in the lower part of Fig. 5 must be due to the excess pressure increment, the magnitude of which is given by

$$P_c - P_a = \Delta P_a (k_a/k_c - 1) \quad (5)$$

where P_c = computed pressure required to account for the greater coefficient of permeability under the same void ratio conditions, k_a = actual coefficient of permeability, and k_c = coefficient of permeability given by broken lines of Fig. 5 at relevant void ratios.

DISCUSSION OF TEST RESULTS

Figs. 3 and 6 indicate that the bentonite conforms in a general way with the work of Hurst and Jordine (1964) who computed that for montmorillonite, interparticle spacings of between 30 Å and 10 Å represented a net attractive force due to the dominance of the surface-ion component, probably involving a collapsing of structure in the consolidated sample, with a spontaneous reduction of interparticle spacing from 30 Å to 10 Å. It is most probable that the critical spacing is reached at different stages of loading throughout the sample and this would account for the appearance of the phenomenon at a net spacing of about 60 Å and its continuation to about 15 Å. For the collapse to occur under saturated conditions there must be a greater expulsion of water and this would manifest itself as an apparent increase in the coefficient of permeability. A greater expulsion of water for identical void ratio conditions would imply an increased hydraulic gradient which would in turn require increased pore pressures and increased total stresses which could only be supplied by the attractive surface-ion forces coming into play at a specific interparticle spacing.

The deviation of the actual compression curves from the theoretical in the low pressure range is probably due to the effect of edge-to-face attractions between particles, which modify the parallel-plate model in the range down to about 80 Å spacing. In the range from 60 Å to 20 Å, the constant value of the apparent activity factor $f'(i)$ would seem to indicate that the parallel plate model is valid provided the concept of an osmotic activity factor is accepted. Since osmosis without membrane, as occurs in a clay dispersion, is a consequence of the kinetic energy of the dispersed ions, the forces that play the role of a semi-permeable membrane must necessarily decrease the osmotic activity of the ions by restraining their freedom of movement and hence their kinetic energy. Thus the osmotic pressure holding the plates apart is decreased and the system reaches equilibrium at a void ratio less than the theoretical.

It is also probable that osmotic theory cannot be used when considering the minimum spacing shown since electric double layers break down when interparticle spacings are reduced to the order of 0 Å-10 Å and, also, hydration shells of Na ions limit the distance of approach to the particle surfaces to 5-6 Å (Norrish and Rausell-Colom, 1961). At spacings of this order Jordine, *et al.* (1962), have shown that the forces between platelets become repulsive due to hydration and image forces and this would account for the upward trend of the curves in Figs. 3 and 6 at the lowest particle separations.

In assessing the test results presented, it must be realized that the computations made involve numerous assumptions.

The assumptions made in computing the coefficient of permeability from the results of consolidation tests are well established, while those used in computing the activity factor have been outlined by Bolt in his analysis of the compressibility of pure clays, the most uncertain factor being the calculation of the interparticle spacing from the void ratio and specific surface of the bentonite.

Some difference of opinion exists as to the correct value to adopt for the specific surface of bentonite. If the value of Edwards and Quirk (1962) is accepted at between 560 and 600 sq.m./gram this would cause all curves of Figs. 3 and 6 to be shifted to the right by a factor between 1.33 and 1.43 and in Fig. 5 a void ratio of 5.6 would correspond to a value of $2d = 72$ to 77 \AA . At the lower plate separations this factor would not be of great significance. The greatest discrepancy would occur by virtue of the fact that the theoretical curve of Fig. 4 would be displaced to the left and most of the activity factor values would be greater than unity. For $S = 600$ sq.m./gram the activity factor for sample 1 would be increased by a factor of 1.44 at the smallest plate separations to 1.66 at the crest of the "S".

In the light of the comments made previously it is felt that actual numerical values of the activity factor are of little concern and it is the shape of the curves and relative particle separations which are of interest.

CONCLUSION

These compression tests on bentonite indicate that, at least for the material used, there is no unique relationship between

the coefficient of permeability and void ratio but that at a certain critical interparticle spacing, which is most probably dependent on the material, surface-ion attractive forces come into play to alter the relationship.

Since, in the performance of the consolidation tests, no time-expansion readings were taken, it has not been possible to examine the permeability - void ratio relationship during increasing interparticle spacing.

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