The Mechanical Properties of Montmorillonitic and Illitic Clays related to the Electrolytes of the Pore Water

Les propriétés mécaniques des argiles montmorillonitiques et illitiques dépendant des électrolytes de l'eau interstitielle

by J. Moum and I. Th. Rosenqvist

Summary

Artificially sedimented and consolidated clays derived from illitic and montmorillonitic raw materials prove that replacement of sodium by potassium under constant load involves important changes in mechanical properties. Two series of illitic and montmorillonitic artificial clay sediments were prepared by effective consolidation, loads from 6 to 12 tons per sq. m for one year. After completed consolidation, half of the samples were subjected to percolation by equimolecular potassium chloride solutions. After two years of percolation, about 60 per cent replacement had taken place in the montmorillonites and about 90 per cent in the illites. The following observations were made:

1) the water content of the sediments was not affected by the replacement process;

2) the undisturbed shear strength was increased by replacement in all samples;

3) the remoulded shear strength increased in case of the illitic clays, but decreased in case of the montmorillonite clays. Thus the sensitivity of the illitic sodium clays dropped from 8 to 3·5, whereas the corresponding factor in the montmorillonite clays rose from 4·5 to 11. The basal plane distance of the consolidated montmorillonites dropped from 20·1 Å to 16·35 Å, corresponding to an expulsion of one monomolecular layer of water per unit cell. As the total moisture content was unchanged, this means a transfer from intergranular water to pore water;

4) the artificial K illites had properties resembling those of the weathered crusts of marine illitic clays. The K montmorillonites are similar to the clays of Mexico City.

Sommaire

Dans les argiles artificiellement sédimentées et consolidées, provenant de matériaux bruts illitiques et montmorillonitiques, le remplacement, sous charge constante, du sodium par du potassium, entraîne d'importants changements des propriétés mécaniques. Deux séries de sédiments artificiels argileux, illitiques et montmorillonitiques, ont été préparées et soumises pendant un an à des pressions effectives de consolidation variant de 6 à 12 t/m^2. Une fois la consolidation achevée, on a soumis la moitié des échantillons à une percolation par des solutions de chlorure de potassium équimoléculaires. Au bout de deux ans, le pourcentage de remplacement du sodium par le potassium avait atteint environ 60 pour cent dans les montmorillonites et 90 pour cent dans les illites. On a fait les observations suivantes:

1) la teneur en eau des sédiments n'a pas été affectée par le processus de remplacement;

2) la résistance au cisaillement de l'argile non remaniée a augmenté dans tous les échantillons;

3) lorsque des argiles ont été remaniées, la résistance au cisaillement des argiles illitiques a augmenté, tandis que celle des argiles montmorillonitiques a diminué. Ainsi, la sensibilité des argiles illitiques sodiques est tombée de 8 à 3,5, tandis que celle des argiles montmorillonitiques a augmenté de 4,5 à 11. Dans les montmorillonites consolidées l'épaisseur des feuillets a diminué de 20,1 Å à 16,35 Å, ce qui correspond à l'expulsion d'une seule couche monomoléculaire d'eau par feuillet. Puisque la teneur totale en eau est restée la même, cela veut dire que de l'eau absorbée a été transformée en eau interstitielle;

4) les illites potassiques artificielles ont des propriétés ressemblant à celles des croûtes d'altération des argiles illitiques marines. Les montmorillonites potassiques présentent une grande ressemblance avec les argiles de Mexico.

At the Fourth International Conference on Soil Mechanics (London 1957) the liquid limit of montmorillonitic and illitic clays was discussed in relationship to the polarizability of the exchangeable cations (Rosenqvist). Data were given for a Hungarian bentonite clay and two Norwegian illitic clays. The Hungarian bentonite consisted mainly of montmorillonite and quartz, and had an ionic exchange capacity equal to 110 m.e.q. per 100 gr.

The Norwegian hydrous mica clays, consisting of 90 per cent and 40 per cent illite respectively, had ionic exchanging capacities of 30 m.e.q. and 17 m.e.q. per 100 g. It was demonstrated that the liquid limit increases with increasing size of the monovalent cations $\mathrm{Li} < \mathrm{Na} < \mathrm{Rb} < \mathrm{Cs}$ in the case of the illitic clays. The gradient of increase was proportional to the content of illite, or the ionic exchange capacity.

In the case of the montmorillonitic clays it was found that the liquid limit dropped from Li to Na and from Na to K, but increased on passing from K to Rb and from Rb to Cs. The gradient of increase from K to Cs was regular in the same way as for the hydrous mica and proportional to the ion exchange capacity. In a discussion in the American Society of Civil Engineers (1959), the variations in the absorbtion of the interstratified water were suggested to be a function of the ionic state of the system. The force of absorbtion is higher with Li than with Na, and still lower with K, whereas it seems to remain constant in the case of K, Rb and Cs. The force of absorption in the three latter cases is small in comparison with the force of absorption of the smaller alkali ions. As the liquid limit of a clay expresses the difference in moisture content between a clay/water mixture of low shear strength and the material dried at 110° C, intercrystalline as well as infracrystalline water is involved in the case of the montmorillonites. As the amount of intercrystalline water at the liquid limit depends upon the bonding strength between the minerals, this amount will increase in the following manner; $\mathrm{Li} < \mathrm{Na} < \mathrm{K} < \mathrm{Rb} < \mathrm{Cs}$, whereas the amount of infracrystalline water, being a function of the absorbtion force, varies in the manner $\mathrm{Li} > \mathrm{Na} > \mathrm{K} = \mathrm{Rb} = \mathrm{C}$. When these two curves are summed up, the variation in the liquid limit indicated above, is obtained (Fig. 1).

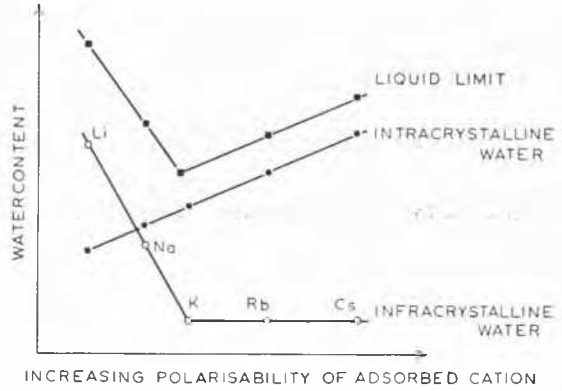

Fig. 1 Diagram showing the nature of water present in alkali montmorillonites as the function of adsorbed cations.

Diagramme montrant la nature de l'eau existant dans les montmorillonites alcalines en fonction des cations absorbés.

In the case of the illites we have no interstratified water which is driven off at 110° C, so only the bonds between the minerals and the hydration, of the ions themselves control the liquid limit. This seems to he the reason why a regular increase in the liquid limit occurs in the whole series from Li to Cs. In order to investigate the importance of infra and intercrystalline water upon the shear strength properties of montmorillonitic and illitic clays, a series of sedimentation and consolidation tests has been carried out. The equipment used is the same as that described by Bjerrum and Rosenqvist in 1957. Sedimentation was carried out in cylinders having a cross sectional area of 140 cm^2. Natural illitic clay from Åsrum and the same montmorillonite as referred to above were used. The cylinders contained a solution of NaCl containing 13,5 g. Na per litre which is equivalent to the content of 35 g NaCl per litre in normal sea water. The sediments were prepared from suspension of the respective clays which were added to the cylinders in small portions during one week in the case of the bentonite, and in two weeks in the case of the hydrous mica clays. The clay mineral suspensions which were added flocculated and settled fairly rapidly.

The suspensions were allowed to settle for one month. Subsequently, a filter paper was placed on top of the sediments over which 100 g of fine silt was deposited. After consolidation for one week another 100 g of silt was added, then after three days 240 g of coarse silt, and finally after a further three days 500 g of sand ; the cylinders were then left to consolidate for 5 days. The gain of strength in the clay was now sufficient to permit the support of a perforated brass plate with a weight of 900 g. Upon this brass plate the loading frame was gradually built up, and during a period of 4 months an increasing load was applied in order to consolidate the sediment. After 6 months under full load, the settlement had decreased to such an extent that the actual experiment could start. In half of the cylinders the supernatant NaCl solution was removed and replaced by KCl solution. At the same time a slight suction of about 0·1 atm. was applied to the bottom af all sediments. The cylinders were left in this state for 26 months before further examinations were undertaken. The cylinders were then opened and the mechanical and chemical properties of the clays determined.

In each sediment the top and bottom parts were examined separately, because as a result of the suction below the sediment there was a difference in the effective consolidation stress applied during the time of percolation.

The data obtained are shown in the attached table. The main results are included in Fig. 2 and 3, from which it is seen how the shear strength in the undisturbed and remoulded

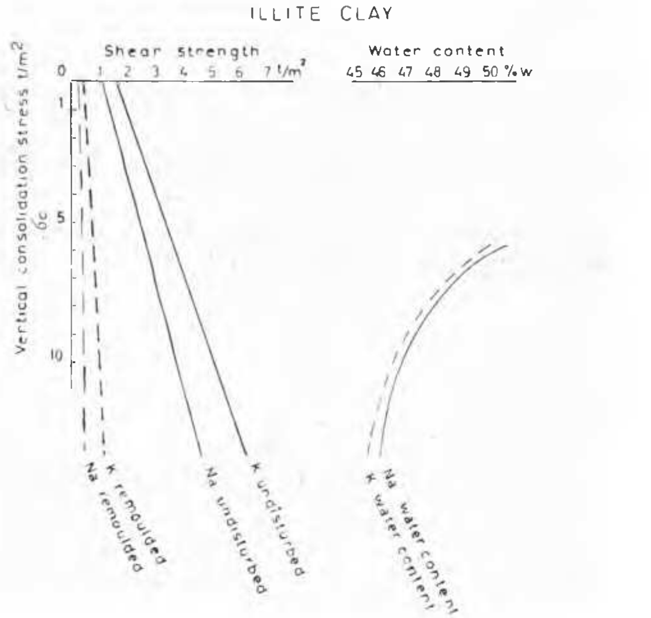

Fig. 2 Undisturbed and remoulded shear strength of illitic clays as a function of the effective overburden pressure for different adsorbed alkali cations. The variation in water content is also shown.

Résistance au cisaillement — cas d'échantillons non remaniés, et d'échantillons remaniés — dans des argiles illitiques, en fonction de la pression efficace de consolidation pour les différents cations d'alcalis absorbés. En outre, la variation de la teneur en eau est représentée.

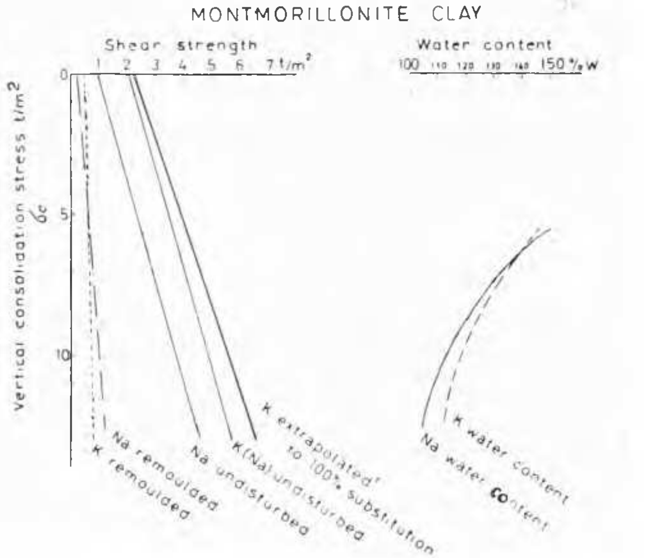

Fig. 3 Undistrubed remoulded shear strength of montmorillonitic clays as a function of the effective overburden pressure for different adsorbed alkali cations. The variation in water content is also shown.

Résistance au cisaillement — cas d'échantillons non remaniés, et d'échantillons remaniés — dans des argiles montmorillonitiques, en fonction de la pression de consolidation pour les différents alcalis absorbés. En outre, la variation de la teneur en eau est représentée.

state varies for the two types of clay, both as a function of the load and with the alkali ion substituted. With the illite clay the variations are relatively regular. It is seen that the strength of the potassium clay at any load will be about 1·55 times the strength of the undisturbed sodium clay, whereas the strength of the remoulded potassium clay is about 2·1 tiles the strength of the remoulded sodium clay. The substitution of sodium by potassium has been nearly complete in all the cases. It involves an increase in the undisturbed as well as in the remoulded strength. The sensitivity decreases from an average of about 7·5 in the case of sodium clays to less than 4·5 in the case of potassium clays. In the montmorillonitic clays the substitution has not been complete. This lack of completeness is particularly striking in clays consolidatid under maximum loads. The regularity in the shear strength values of the montmorillonite clays is not so good as in the case of the illites, but it is obvious that the shear strength in the undisturbed state has increased and in the remoulded state has decreased as a result of replacement. Thus the sensitivity of the potassium montmorillonite is markedly higher than in the case of the sodium montmorillonite.

Examining the values given for the potassium/sodium ratio in the four montmorillonitic clays, namely 13·5/2·5 — 13·5/2·5 — 10·0/7·5 and 8·5/6·5, we observe that the substitution decreases from about 85 per cent in the least loaded clays to 55 per cent in the clays which carried the maximum loads. If we use equivalent units instead of weight units, the substitution in the last-mentioned case is only 44 per cent. It therefore seems appropriate to augment the increase in undisturbed shear strength observed with incomplete substitution. To find out what the increase might be with complete substitution, the increase in shear strength may be extrapolated proportionalety to the degree of substitution. Thus the calculated undisturbed shear strength at 11 T/m^2 consolidation stress is found to be about 7 T/m^2 for 100 per cent replacement, compared to the average value of 5·5 T/m^2 for incomplete substitution, and 4 T/m^2 for the sodium montmorillonites with the same consolidation stress. The ratio between shear strength for the undisturbed potassium montmorillonite and the undisturbed sodium montmorillonite should thus be of the order of 1·7, whereas the remoulded potassium clay must be considerably softer than the corresponding remoulded sodium clay. The most probable value of the shear strength for the remoulded potassium clay would be about half of its value in the sodium state. As a consequence of this calculation it is found that the sensitivity increases from 4 with the sodium montmorillonite to 14 with the potassium montmorillonite.

Previous experience has indicated that neither the illitic clays nor the montmorillonites would have any shear strength at zero consolidation load. In the case of all the clays studied here, however, a straight line extrapolation gives zero shear strength at a negative consolidation load of the order of 2 T/m^2.

The explanation of this anomaly probably lies in the technique used for these experiments. Most probably the boundary conditions during loading introduce an error into the calculation of the effective consolidation stress. The parts of the specimen which have been examined have probably carried a higher effective load than that calculated on the basis of the cross section of the cylinder and the loads employed. It has not been possible to determine or recalculate this error with any accuracy, but rough estimates indicate that it may be as high as a few tons per m^2. Furthermore, there is the possibility that with the clays in this system a certain amount of cementation has occurred between single mineral grains. This may especially be the case with the illitic clays, as it seems here as if the shear strength of the remoulded clays will be zero in the neighbourhood of the origin of the system. This factor is unlikely to apply to the montmorillonitic clays, and it is considered most likely that in this case the calculated consolidation stresses have been somewhat too low. Whatever the reason, it is possible to express the increase in shear strength as a function of the consolidation stress. This ratio may be denoted as $(c/p)'$ and has the following values :

	$(c/p)'$
Sodium illite	0·29
Potassium illite	0·44
Sodium Montmorillonite	0·29
Potassium Montmorillonite extrapolated to 100 per cent substitution	0·46

It is well known that a substitution of sodium by potassium in the case of montmorillonites may change the montmorillonite lattice into a mixed layer of mica lattice. In order to investigate the swelling properties of the clays X-ray photographs were taken of the montmorillonite sediments in their undisturbed state on broken vertical section in order to determine the basal plane distance and the random nature of the mineral arrangement. The result of the investigation was that the *hk*0 values were unaffected by the substitution, whereas the 001 values had undergone a marked change from the basal plane distance of 20·1 Å in case of the sodium montmorillonite consolidated at 11 t/m^2 to 16·25 Å after partial substitution by potassium. In both cases the minerals were found to be distributed haphazardly with no preferred orientation. The difference in basal plane distance, namely 3·75 Å, is very nearly equal to half the *C*-axis for ice, which ia 7·41 Å.

As two layers of water are present along the hexagonal cell axis in ice, the thickness of 3·75 Å is attributed to a monomolecular layer of water, which corresponds to the change in basal plane distance resulting from a partial substitution of sodium by potassium. The distances found by X-ray examinations indicate that, with the consolidation stresses here employed, the minerals in their sodium state possess a trimolecular layer of water between the backbone sheets of the montmorillonite structure and two molecules in the mixed sodium-potassium state. Thus an expulsion of water corresponding to about 18 per cent of the volume of the mineral has taken place, this water having been transferred from the internal parts of the single minerals to the pore water, whereas the distances along the *a* and *b* axes have not been changed.

In Fig. 4 the liquidity indices are given in relation to the

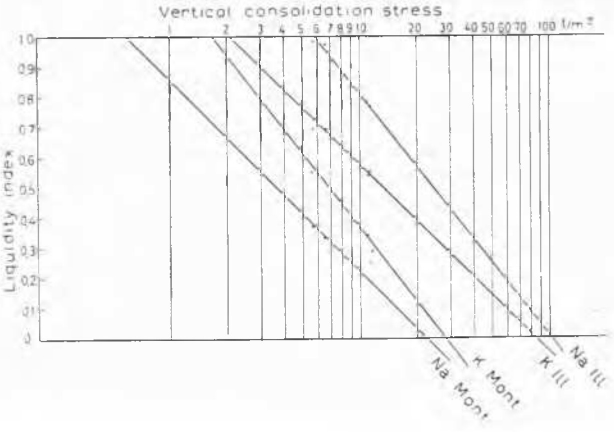

Fig. 4 Variation in liquidity index as a function of the vertical consolidation stress for illitic and montmorillonitic clays containing different adsorbed cations.

Variation des indices de liquidité en fonction de la pression verticale de consolidation pour les argiles illitiques et montmorillonitiques contenant différents cations absorbés.

K/Na g/litre *in pore water*	*Effective consolida-tion pressure* t/m^2	*Undisturbed shear strength* t/m^2	*Remoulded shear strength* t/m^2	*Water content* per cent	*Liquid limit* per cent	*Plastic limit* per cent	*Plasticity index* per cent	*Liquidity index*
Illitic Clays saturated with Sodium								
0·05/13	5·8	2·4	0·32	50·6	51·5	23·4	28·3	0·98
0·05/13	6·6	3·1	0·33	49·1	50,7	23·4	27·3	0·97
0·62/13	11·1	4·0	0·55	45·7	52·0	23·4	28·6	0·78
0·37/13	11·9	4·2	0·49	46·6	53·4	23·2	30·2	0·77
Illitic Clays after Substitution by Potassium								
15/1·4	5·8	3·75	0·75	49·8	60·2	27·2	33·0	0.685
14/1·5	6·6	5·0	0·75	49·4	60·9	26·5	33·5	0·685
16·5/0·5	11·1	6·2	1·1	45·5	60·5	27·3	33·7	0·54
16·5/1·4	11·9	6·3	1·3	46·1	61·3	26·9	34·4	0·555
Montmorillonitic Clays saturated with Sodium								
0·055/13·0	5·7	2·4	0·7	147	300	58	242	0·37
0·05/13·5	6·5	3·0	0·8	136	302	58	244	0·33
0·06/12·5	11·2	3·65	1·0	111·2	318	57	261	0·25
0·34/14·5	12·0	4·9	1·1	102·8	299	58·8	240	0·14
Montmorillonitic Clays after Substitution by Potassium								
13·5/2·5	5·6	3·7	ca. 0·6	141	ca. 200	70	ca. 130	0·55
13·5/2·5	6·4	3·8	« 0·6	148	« 200	70	« 130	0·60
10/7·5	11·1	4·5	« 0·7	120·5	« 220	68	« 150	0·345
8·5/6·5	11·9	6·0	« 0·8	113·9	« 230	65	« 165	0·298

consolidation stresses for the four types of clay, sodium illites, potassium illites, sodium montmorillonites and mixed sodium/potassium montmorillonites. The consolidation stresses required to give liquidity indices equal to 1 and zero will be 5·5 t/m^2 and 100 t/m^2 respectively, and in'case of sodium illites, 2 t/m^2 and 85 t/m^2 in case of potassium illites, 0·6 t/m^2 and 23 t/m^2 in case of sodium bentonites, and 1·7 t/m^2 and 27 t/m^2 in case of the mixed sodium-potassium bentonites. The greater sensitivity of the montmorillonites towards consolidation stresses is well known. The increase in this factor by a substitution of sodium by potassium is a new result of this investigation. It tallies with the changes in lattice dimensions.

The substitution experiments carried out prove the connection between the composition of the electrolytes, and consequently the ionic state of the mineral and the mechanical properties of the soils. The sensitivity by remoulding is one of these mechanical factors which has been the chief aim of investigations carried out in Norway during the last decades. The simplest possible chemical change is that which takes place in some Scandinavian quick clays. Here we have a replacement of saline pore water by fresh water. In the case of illitic clays this involves a decrease in the undisturbed shear strength and a pronounced increase in sensitivity, provided no other chemical or mineralogical changes take place. With montmorillonitic clays such a simple replacement will give opposite effects. It is, however, not only by leaching that sensitivity may arise in illitic clays. Even by *addition* of certain electrolytes, for instance phosphates, silicates and other dispergents, the sensitivity may increase in case of illites, as earlier described by ROSENQVIST 1955, and SODERBLOM 1959.

In such cases it is mainly the anions which are active, but even changes in the cations, from highly polarizable cations to less polarizable cations may leas to the same result, although no examples of such replacements are known in nature. The opposite effect is, however, very frequent, namely the replacement of less polarizable cations by more polarizable, as for instance the replacement of sodium by potassium. This change takes place in the weathered crust of all Scandinavian marine clays, where the potassium ions are furnished by the weathering of potassium bearing minerals. These natural changes resemble those described by the authors. Many properties of the weathered crusts of the Scandinavian clays are reproduced in the artificial potassium illites mentioned above, whereas the mixed potassium/sodium montmorillonites resemble the Tacubaya clays of the Mexico City sediment series (ZEEVAERT, 1957 and FOREMAN, 1955). The weathered crusts of Scandinavian marine clays and the clays of Mexico City originated through partial weathering of eroded material. In the Scandinavian clays the mineralogy is determined by the illlites and chlorites, whereas in the Mexico soils the montmorillonitic mineralogy with a high .percentage of biogen material and unweathered volcanic glass is characteristic. The Scandinavia clays were deposited in more or less highly saline sea water, whereas the Mexican sediments were deposited in a salt lake of variable low salinity.

Conclusions

The liquid phase of the sediments will normally change their composition by natural processes during geological time. Among the ions of marine and several lacustrine sediments, potassium and sodium are the most important. In order to investigate the influence of the substitution of these ions upon the properties of various clay sediments, investigations have been made on representatives of the non-expanding hydrous mica minerals and the expanding montmorillonite minerals. In the experiments, comprising artificial sedimentation, consolidation and replacement, a Norwegian hydrous mica and a Hungarian bentonite were used. The experiments, including the consolidation and substitution of sodium by potassium in half of the series, lasted for about three years.

In all clays the shear strength increased with increase in consolidation stress. The substitution of sodium by potassium produced a systematic increase in the shear strength of the illites as well as in the montmorillonites in their undisturbed state. The moisture content was not markedly influenced by the substitution. In the case of the illitic clays the shear strength in the remoulded state increased more than in the undisturbed state. Thus the sensitivity decreased from 7·5 to 4·5 as a result of the substitution. With the montmorillonite clays the increase in shear strength as a function of the consolidation stress was of about the same order as for the hydrous mica clays. This held for the sodium as well as for the potassium state. Even here the water content of the sediment was uninfluenced by the substitution. In contrast to the illitic clays, however, remoulded potassium montmorillonite has a much lower shear strength than the corresponding sodium montmorillonite has a much lower shear strength than the corresponding sodium montmorillonite. Thus the sensitivity of the montmorillonites increased from 4 to 14. This is connected with a decrease in liquid limit as a result of the substitution. The basal plane distance of the montmorillonites decreased from 20·1 to 16·35 Å as a result of the substitution of about half of the sodium by potassium. This involves an expulsion of a monomolecular layer of water from its interstratified position in the montmorillonite lattice. As the total moisture content was constant, this means that some of the internal water changed into external water.

From these experiments the conclusion is drawn that the shear strength in an undisturbed clay is a function of attracted van der Waalske forces between the adsorbed cations of one mineral and the ions of the adjacent minerals. By substitution of the sodium ion with less polarizability into a potassium ion of higher polarizability, the attractive forces and bonds of the flocculated system are increased. This increase seems to be the same in case of illitic and montmorillonitic clays. By remoulding, where the short distance contacts are broken by the mechanical treatment, we arrive at a shear strength in the remoulded state which is no longer a function of the stress history of the sediment, but depends mainly upon the mineralogical and chemical composition as well as the volume ratio between mineral phase and liquid phase. With montmorillonites some of the water molecules leave their position in the mineral lattice, thus increasing the volume of the liquid phase, at the same time decreasing the volume of the mineral phase. The above-mentioned ratio is then altered. With illitic sediments no such effect is possible. The substitution will consequently not influence the volume ratio between minerals and water.

References

[1] Foreman, F. (1955). Paleontology in Southern North America. Part II. Geological Society of America. *Bulletin*, Vol. 66, pp. 475-510.

[2] Michaels, A. S. Discussion on : Rosenqvist, I. T. Physico-chemical properties of soils ; properties of soil-water systems. American Society of Civil Engineers. *Proceedings*, Vol. 85, No. SM 2, pp. 91-102.

[3] Moum, J. and Rosenqvist, I. T. (1955). Kjemisk bergartsforvitring belyst ved en del leirartsprofiler. *Norsk geologisk tidsskrift*, Vol. 34, pp. 167-174. (Norwegian Geotechnical Institute. Publ., 8).

[4] Rosenqvist, I. T. (1957). Discussion on : Earth dams, slopes and open excavations. International Conference on Soil Mechanics and Foundation Engineering, 4. London. *Proceedings*, Vol. 3, pp. 257-258.

[5] — (1955). Investigations in the clay electrolyte water system. Oslo. 125 p. *Norwegian Geotechnical Institute*. Publ., 9.

[6] — (1960). Physico-chemical properties of soils ; properties of soil-water systems. American Society of Civil Engineers. *Proceedings*, Vol. 86, No. SM 1, pp. 75-77.

[7] Söderblom, R. (1959). Aspects on some problems of geotechnical chemistry. Geologiska föreningen i Stockholm. *Förhandlingar*, Vol. 81, No. 4, pp. 727-732.

[8] Zeevaert, L. (1957). Foundation design and behaviour of Tower Latino Americana in Mexico City. *Géotechnique*, Vol. 7, No. 3, pp. 115-133.