

## EFFECT OF COMPACTION ON THE PORE FLUID CHEMISTRY OF MONTMORILLONITE

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(Received 3 November 1975; and in final form 12 March 1976)

**Abstract**—An apparatus for consolidating fine grained materials under controlled conditions of total stress, pore fluid pressure and temperature is described and the variation in chemistry of pore fluid expelled during compaction from a montmorillonitic material at 40°C is reported. The decrease in concentration of dissolved salts in the pore fluid is related exponentially to the effective axial pressure.

### INTRODUCTION

Large volumes of water are expelled from argillaceous sediments during compaction. In order to understand the effects that this water has on the surrounding deposits a knowledge is required of its composition and the way in which this changes with time. The main processes which alter the chemistry of interstitial water are summarised by Chilingarian and Rieke (1969) as (1) physical (compaction) (2) chemical (reactions between rock minerals, organic matter and interstitial solutions) (3) physicochemical (filtration through charged-net clay membranes, adsorption and base-exchange) and (4) biochemical.

Attempts have been made to study the diagenesis of interstitial water by sampling fluids from deep boreholes, e.g. Manheim and Sayles (1970) and Bischoff and Kur (1970). Even if the water samples are obtained without contamination in the field there is the inherent problem of having to assume how the water has changed with compaction without knowledge of the previous temperatures, pressures and initial composition. An alternative approach to this problem has been to study the effects of compaction on pore fluid chemistry by controlled laboratory tests. A summary of recent work is presented by Rieke and Chilingarian (1974). According to Chilingarian *et al.* (1969) the chemistry of expelled solutions depends on the types of clay present as most of the salts initially present in the interstitial waters trapped during sedimentation are squeezed out during the initial stages of compaction. The laboratory results mentioned in Chilingarian *et al.* (1973) obtained by Kryukov and Komarova (1954), Kryukov and Zhuchkova (1963), Von Engelhardt and Gaida (1963), Buneeva *et al.* (1964), Rieke *et al.* (1964) and Chilingarian and Rieke (1968) also showed that mineralisation of expelled solutions progressively decreases with increasing overburden pressure.

The change in interstitial fluids also has been predicted from theories such as that proposed by Warner (1964). This predicts from double-layer theory that the electrolyte content of expelled interstitial water

should decrease with increasing compaction pressure in the case of highly colloidal clays because of the expulsion of free interstitial water by the tighter bound double-layer water.

### EQUIPMENT

The diagenesis of natural materials is being investigated by the laboratory simulation of consolidation under controlled conditions of stress, pore fluid pressure and temperature. An apparatus has been developed (Wijeyesekera and de Freitas, 1975) which enables a sample to be consolidated over a range of stress from 0 to 700 kg/cm<sup>2</sup> at temperatures of 0–200°C while continuously monitoring the pore fluid pressure and degree of consolidation. The specimen of soil is contained in a stainless steel cylindrical cup of 10.16 cm internal diameter which is forced over a rigid cylindrical piston by hydraulic pressure, to permit consolidation. This facilitates smooth axial loading of the specimen with a minimum interference from sliding friction of the teflon-lined seals. Porous sintered stainless steel discs at each end of the specimen enable uniform drainage of expelled pore fluid from the sample. In this test drainage was only allowed to take place from the bottom disc and the top disc was used for measurement of pore fluid pressure. The pore fluid expelled during consolidation is collected by displacement of a syringe inserted into the drainage plate at the base of the specimen (Fig. 1).

### MATERIAL AND SAMPLE PREPARATION

The testing medium used in the experiments reported in this paper was Fuller's Earth from the Lower Cretaceous of Nutfield, Surrey (U.K.). The natural Fuller's Earth consists of calcium montmorillonite with trace quantities of quartz and accessory heavy minerals (Cowperthwaite *et al.*, 1972). This material was disaggregated in the laboratory and remixed with distilled, de-ionised, de-aired water and allowed to soak for 72 hr to permit the clay to swell

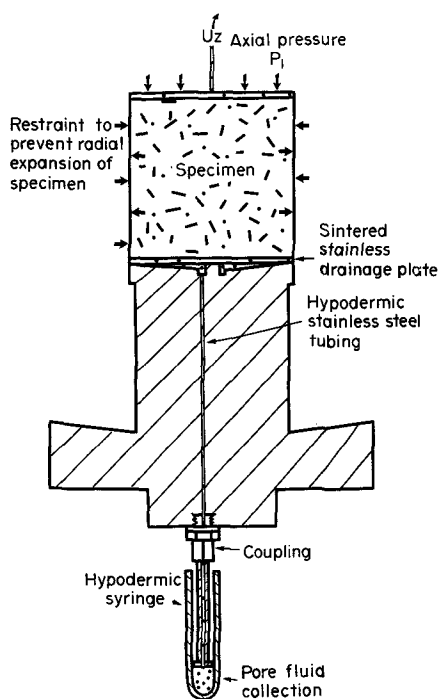


Fig. 1. Simplified diagram of testing apparatus to illustrate collection of pore fluid from a specimen and measurement of pore fluid pressure ( $u_z$ ) during consolidation under an applied axial pressure ( $P_1$ ).

and attain equilibrium with the water. The resulting slurry was then de-aired under a vacuum of 0.5 Torr. The slurry was then poured into a sedimentation column fitted to the top of the cylindrical sample cup and the clay allowed to settle out of suspension under gravity for a period of 72 hr. No drainage of water was allowed from the specimen during this period

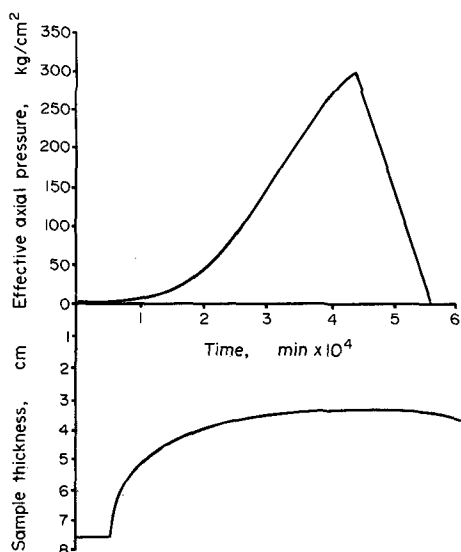


Fig. 2. Variation of effective axial pressure and thickness with time for consolidation of montmorillonite at a constant temperature of 40°C. Loading of the specimen commenced 5000 min after the test began and unloading commenced after 44000 min.

of settling into the sample cup so that the resulting specimen had a fabric related only to the mechanism of deposition and undisturbed by pore fluid flow or transportation. The sedimentation column was then removed and the top of the sample trimmed flush with the top of the cup. The cup was then inserted into the pressure vessel and the cell assembled.

For this experiment the axial pressure was applied at a constant loading rate of 0.7 kg/cm<sup>2</sup>/hr at a constant temperature of 40°C. After the maximum total axial pressure of 350 kg/cm<sup>2</sup> had been reached the specimen was unloaded at 1.4 kg/cm<sup>2</sup>/hr (Fig. 2).

ANALYTICAL TECHNIQUES

The pore fluid expelled during consolidation was collected in syringe bottles. The syringes were cleaned by thorough soaking in a decontaminating surface active agent and the apparatus in contact with the pore fluid was boiled before assembly in water of the same initial chemistry as that used in the preparation of the testing material. The syringe bottles containing the expelled pore fluid were refrigerated at 5°C until required for analysis. The pore fluids were analysed for their concentration of K, Na, Ca, Mg, SO<sub>4</sub> and Cl which originated from the original fuller's earth testing material, principally as adsorbed ions on the montmorillonite. The cations were determined using atomic absorption spectrophotometry. The anions were too dilute to use the standard titration analytical techniques and so a method has been devised based on the turbidity of a solution on the addition of a salt to induce precipitation of either the sulphate or chloride ions. The turbidity was then measured using the scattering of a light beam from a nephelometer. The system was calibrated against a set of standard solutions prepared with the same reagents as used in preparing the anion solutions.

RESULTS

The accompanying graphs (Figs. 3 and 4) show the variation of concentration of K, Na, Ca, Mg, SO<sub>4</sub>

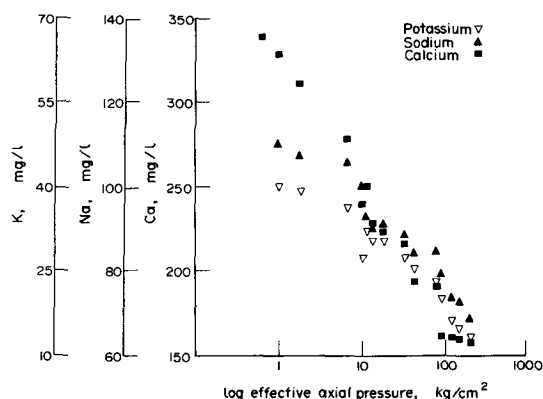


Fig. 3. Variation of K, Na and Ca concentrations in expelled pore fluid with effective axial pressure at 40°C.

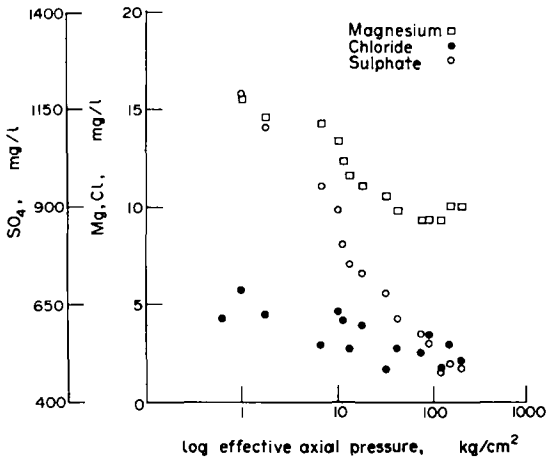


Fig. 4. Variation of Mg, Cl and SO<sub>4</sub> concentrations in expelled pore fluid with effective axial pressure at 40°C.

and Cl with axial effective stress. The total axial stress is equivalent to the hydraulic pressure applied to the piston containing the clay specimen within the cylindrical sample chamber. The axial effective stress is equal to the total axial stress minus the average pore fluid pressure within the specimen. In this series of tests, the back pressure against drainage was set at atmospheric pressure and the average pore fluid pressure has been taken as half the pore fluid pressure measured at the undrained end of the sample.

The initial concentration of all the analysed ions decreases rapidly with increasing stress during the initial loading and thereafter the rate of decrease declines markedly. The decrease in concentration is exponential but shows a break in curvature indicating two distinct phases of pore fluid expulsion. The first, with a relatively low gradient for the cations on Fig. 3, holds from the initial stages of compaction up to about 10 kg/cm<sup>2</sup> effective stress and the second, with a relatively high gradient, covers compaction from 10 kg/cm<sup>2</sup> up to 300 kg/cm<sup>2</sup> (the maximum effective stress used to expel pore fluids in this series of experiments). The change in gradient takes place at a stress which is equivalent to a change in the pattern of mechanical compaction of the clay. Figure 2 shows the variation in sample thickness and effective axial stress with time. The initially rapid phase of compaction is seen to be reduced to a slower phase at about 10 kg/cm<sup>2</sup>. At lower stresses a large volume of fluid

is expelled from the specimen as reflected by the rapid decrease in thickness. The rate of fluid expulsion is then reduced and the specimen attains a more stable equilibrium condition with sufficient drainage of pore fluid to prevent additional increase in pore fluid pressure. This state corresponds to the linear increase of effective axial pressure with time at the same rate of 0.7 kg/cm<sup>2</sup>/hr as the increase of total axial pressure. The slight decrease of gradient of effective axial pressure at the end of the loading period may be the result of a building up of pore fluid pressure as the permeability of the specimen is reduced by the low void ratio resulting from the high degree of compaction at these stress levels. However, the small volume of pore fluid expelled at these high stress levels did not produce a significantly large enough number of analyses to indicate a change in chemical behaviour.

Some comparisons can be drawn between the results of the change in expelled pore fluid concentration and overburden pressure reported in Rieke *et al.* (1964), Chilingarian *et al.* (1973) and the data reported herein (Table 1). In both these papers the total mineralisation of solutions squeezed out of montmorillonite clay saturated in sea water was determined at different overburden pressures. Although the author's results were obtained from the compaction of montmorillonite saturated in distilled water the data confirms the general tendency of the interstitial fluid to decrease in concentration over a range of pressure of 10–300 kg/cm<sup>2</sup>. However, at relatively low pressures of 0–35 kg/cm<sup>2</sup> the results of Chilingarian *et al.* (1973) indicate a decrease in concentration only of K<sup>+</sup> and Na<sup>+</sup> with Ca<sup>2+</sup> and Cl<sup>-</sup> remaining constant and Mg<sup>2+</sup> and SO<sub>4</sub><sup>2-</sup> increasing. This corresponds to the slow change of concentration up to 10 kg/cm<sup>2</sup> followed by a substantial decrease observed by the author. This rate of decrease is of the same magnitude as that observed for the higher pressure range of 10–300 kg/cm<sup>2</sup>. Therefore the conclusion of Chilingarian *et al.* (1973) that the concentration of expelled solutions during the initial stages of compaction is higher than the initial interstitial fluid was not confirmed but initially the fluid did maintain a constant concentration.

The results reported above imply that the mechanisms responsible for releasing ions into the pore fluid and allowing their expulsion are closely related to the mechanical compaction and dewatering

Table 1. Concentration of expelled interstitial fluid at maximum pressure expressed as a percentage of the concentration of the fluid expelled at the minimum pressure of the range indicated

	10–300 kg/cm <sup>2</sup>		0–35 kg/cm <sup>2</sup>	
	Rosenbaum	Rieke <i>et al.</i> (1964)	Rosenbaum	Chilingarian (1973)
K <sup>+</sup>	—	—	72	76
Na <sup>+</sup>	68	25–87	82	95
Ca <sup>2+</sup>	64	50–62	63	100
Mg <sup>2+</sup>	74	60	67	129
SO <sub>4</sub> <sup>2-</sup>	50	67–81	55	113
Cl <sup>-</sup>	40	40–82	50	98

of the sample. Theories accounting for such behaviour have recently been summarised by Rieke and Chilingarian (1975).

Further consolidation tests on montmorillonite are currently in progress at Imperial College using the apparatus described in this paper. These tests are being conducted over a range of temperatures from 20 to 80°C.

*Acknowledgements*—I would like to thank M. K. Armstrong, B. A. Clarke, M. H. de Freitas and B. J. Hough for help in conducting this series of experiments and for criticism during the preparation of this paper. The research reported herein has been sponsored in part by the United States Army through its European Research Office and the author has been supported by a Natural Environmental Research Council research studentship. Both awards are gratefully acknowledged.

#### REFERENCES

- Bischoff, J. L. and Ku, Teh-Lung (1970) Pore fluids of recent sediments—I. Oxidising sediments of 20°N Continental Rise to Mid-Atlantic Ridge: *J. sedim. Petrol* **40**, 960–972.
- Buneeva, A. N., Kryukov, P. A. and Rengarten, E. V. (1964) Experiment in squeezing out of solutions from sedimentary rocks: *Dokl. Akad. Nauk. SSSR* **57**, 707–9.
- Chilingarian, G. V. and Rieke, H. H. (1968) Data on consolidation of fine grained sediments: *J. sedim. Petrol* **38**, 811–6.
- Chilingarian, G. V. and Rieke, H. H. (1969) Some chemical alterations of subsurface waters during diagenesis: *Chem. Geol.* **4**, 235–252.
- Chilingarian, G. V., Sawabini, C. T. and Rieke, H. H. (1973) Effect of compaction on chemistry of solutions expelled from montmorillonite clay saturated in sea water: *Sedimentol.* **20**, 391–8.
- Cowperthwaite, I. A., Fitch, F. J., Miller, J. A., Mitchell, J. G. and Robertson, R. H. S. (1972) Sedimentation, petrogenesis and radioisotopic age of the Cretaceous Fuller's earth of Southern England: *Clay Min* **9**, 309–328.
- Kryukov, P. A. and Komarova, N. A. (1954) Concerning squeezing out of water from clays at very high pressures: *Dokl. Akad. Nauk. SSSR* **99**, 617–9.
- Fukov, P. A. and Komarova, N. A. (1954) Concerning micral phenomena associated with driving out of solutions from rocks. In present day concepts of bound water in rocks: *Izd. Akad. Nauk. SSSR, Lab. of Hydrogeol: Problems*, pp. 95–105. F. P. Savarenskiy, Moscow.
- Manheim, F. T. and Sayles, F. L. (1970) Brines and interstitial brackish water in drill cores from the deep Gulf of Mexico: *Science* **170**, 57–61.
- Rieke, H. H., Chilingar, G. V. and Robertson, J. O. (1964) High pressure (up to 500,000 psi) compaction studies on various clays: *22nd Int. Geol. Cong.*, New Delhi **15**, 22–38.
- Rieke, H. H. and Chilingarian, G. V. (1974) *Compaction of Argillaceous Sediments*, 424 pp. Elsevier, Amsterdam.
- von Engelhardt, W. and Gaida, K. H. (1963) Concentration changes of pore solutions during the compaction of clay sediments: *J. sedim. Petrol.* **33**, 919–930.
- Warner, D. L. (1964) An analysis of the influence of physical-chemical factors upon the consolidation of fine-grained clastic sediments. Ph.D. thesis, University of California, Berkeley.
- Wijeyesekera, D. C. and de Freitas, M. H. (1975) High pressure consolidation of kaolinitic clay: *Bull. Am. Ass. Petrol. Geol.* (in press).