Quantitative Estimation of Particle Orientation of Montmorillonite by Optical and X-Ray Diffraction Techniques

by

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Introduction

An understanding of the concept of the microstructure of clays involves the study of the scalar and the vectorial properties (Sander, 1939). The scalar quantities are the physical and mechanical properties, *e.g.* specific gravity. The vectorial properties are the particle orientation, orientation distribution, directional features of voids, etc., with respect to a chosen co-ordinate system. These factors have to be determined in relation to the internal and the external boundary stresses. Generally, the vectorial properties of the clays are reflected in the analysis of its fabric (Gillott, 1970). The study of the microstructure of clays facilitates a better understanding of the long term and short term behaviour of clays (Foster and De, 1971).

The microstructural aspects of clays have been defined by various research workers of different disciplines to suit their requirements (Baver, 1956; Brewer, 1964). The following terms are defined for the purpose of clarity and interpretation :

Orientation:—The term orientation describes the general pattern of the arrangement of the clay particles with reference to a plane or co-ordinate system. It does not consider the size of the clay particles and the spatial distances between them.

Fabric:—The term fabric includes the aspects of interparticle distances with reference to a plane or co-ordinate system along with particle orientation.

Microstructure:—The term microstructure of clays includes the aspects of the fabric, environmental conditions, stress history, etc.

The versatile techniques, generally employed to quantify the average particle orientation of clays, are optical microscopy and X-ray diffraction.

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The latest trend is to synthesise such studies (Gillott, 1970; Mckyes and Young, 1971; Tchalenko, Burnett and Hung, 1971; and Venkateshwar Rao, 1972) to facilitate greater understanding of the clay microstructural behaviour. Reviews on these topics were made by Kahn and Bragg, (1966) and Morgenstern, Mitchell and Quigley, (1969). Electron microscopic study facilitates a qualitative understanding of the orientation at primary particle level (O'Brien, 1972), but it may mislead in certain cases when interpreted on macrosopic level (Mckyes and Young, 1971; Venkateshwar Rao, 1972).

Most of the investigations are limited to the study of the orientation of natural clays containing illite and kaolin and on the laboratory samples of commercial kaolin and illite. (Buessem and Nagy, 1954; Martin and Ladd, 1970). The particle orientation studies on montmorillonite clays are scarce and were studied only by a few (Englehardt and Gaid, 1963; Meade, 1961; Thiem, 1967). The present study is aimed at expressing the particle orientation of montmorillonite clay *quantitatively*.

Brief Review on Particle Orientation Studies

General

There are three distinct groups of research workers who analysed the microstructure of clays (Venkateshwar Rao, 1973). The first group putforward hypotheses based upon the inferences drawn from the physico-chemical and mechanical behaviour of the clays (Casagrande, 1932; Tan, 1959). The second group dealt with the evaluation of microstructure of clays qualitatively using direct method. (Brewer, 1964; Lambe, 1960; Mitchel, 1960; van Olphen, 1963). The third group attempted to quantify the microstructure of clays using direct methods, (Brindley, 1953; Morgenstern and Tchalenko, 1967).

Techniques for the estimation of particle orientation

Optical Microscopy—Direct observations of interparticle or interdomain disposition on low magnifications are possible by this method. The directional features of the skeleton grains, clay minerals, voids, crack pattern, etc, are observed by measuring the long axes of these features. These directional features are plotted on two or three dimensional co-ordinate polar diagrams to evaluate the general directional preferences of different constituents to predict the possible fracture planes (Fooks, 1965; Lafeber, 1972). Optical properties, *e.g.* crystallinity, transparency, birefringence, etc., are generally employed to determine the particle orientation in an indirect manner. The theory and the principles of interpretation of this method are described by Durrance (1967), Singh (1967), Smart (1966), Winchell (1958), Wahlstrom (1962) and Wu (1958). It is advantageous to employ optical microscope to study the overall features of the particle orientation of monomineralic or polymineralic systems with domains and broad discontinuities (Mitchell, 1956). However, the chief disadvantages in using this method are :

1. Sample preparation technique is cumbersome and disruptive. It is difficult to achieve uniform and identical thin sections from the same source of clay sample, even though large and ultra-thin sections of clays were prepared (Willoughby and Walsh, 1969).

2. Birefringence is a scalar quantity dependent upon the crystal orientations. The intensity of interference colours due to birefringence can only be measured by the photometric method, but not the actual birefringence of the clay particles (Lafeber, 1968).

3. Natural soils are polymineralic. Identification of the individual clay minerals by optical method is not often possible. The combined effect of the birefringence of the polymineral can alone be studied. It is not often possible to segregate these effects (Tchalenko, 1968). Therefore, until a synthesis of different monomineralic studies on polymineral systems is developed this limitation exists (Venkateshwar Rao, 1973).

X-Ray Diffraction Technique :—X-ray diffraction is an important technique employed to study the crystal systems, their orientation and distribution of orientation pattern with respect to a reference plane or co-ordinate system. Clay minerals orient under stress and enhance the basal plane reflections and reduce the prismatic reflections. This property is used in the quantitative estimation of the particle orientation by the X-ray diffraction analysis (Brindley, 1953). It requires a small amount of material for the analysis. The chief advantage of this technique is that it distinguishes each individual clay mineral in a polymineral system (say, natural soils) and permits the evaluation of particle orientation in a more rational way than by the optical microscopic method. However, the combined effect of the particle orientation in a polymineral system cannot be easily evaluated (Tchalenko, 1968). The chief disadvantage of this method is that delineation of the orientation of pores, cracks, dimensional features of skeleton grains and clay minerals is not possible. The dynamical and kinematical correlations between the orientation of clay minerals pore skeleton grains and their displacements cannot also be established directly (Lafeber, 1968). Quantitative evaluation of the clay particle orientation is carried out by adopting peak height ratio (Brindley and Kurtossy, 1961; Gipson, 1966), peak intensity ratio (O'brien, 1964; Odom, 1967), x^2 statics (Silverman and Bates, 1960), Orientation factor (Meade, 1961; Odom, 1967), peak count (Odom, 1967; Quigley and Thompson, 1966), diffraction ratio (Tchalenko, Burnett and Hung, 1971), orientation index (Diamond, 1971), mathematical model analysis (Sturm and Lodding, 1968), integrated intensity (Taylor and Norrish, 1966; Thiem, 1967) and three dimensional Schultz pole figure (Martin and Ladd, 1970). The superiority of one method over the other has not yet been established. In this investigation, peak intensity ratio method has been adopted to estimate the particle orientation of montmorillonite.

Scope of the Present Study

Quantitative estimation of the particle orientation of montmorillonite is aimed at by considering the ideal random and ideal parallel orientations of the clay particles using the X-ray diffraction technique. The particle orientation is expressed quantitatively as the percentage of average particle orientation based upon the X-ray diffraction analysis.

In the laboratory investigation, montmorillonite specimens were consolidated and rebounded at various pressures in oedometer. Samples were extruded from these specimens and impregnated with carbowax 6000 to study the degree of particle orientation in terms of applied external pressure using X-ray diffraction technique. The estimated values of the particle orientation of montmorillonite by the X-ray diffraction technique are compared with the values of particle orientation obtained by the optical microscopic method for the same samples.

A New Method to Express Particle Orientation

From the X-ray diffraction analysis, the average particle orientation in clays is expressed in two dimensional form as the percentage of the relative orientation from the datum of particle orientations varying from ideal random to ideal parallel orientations. The expression is as follows :

$$\begin{array}{l}\text{Percentage}\\ \text{of relative}\\ \text{average}\\ \text{particle}\\ \text{orientation,}\\ \text{`}R\text{'}\end{array} = \left[\frac{(O.F.\ \text{of Specimen}) - \begin{pmatrix}O.F.\ \text{of ideal random}\\ \text{orientation considered}\\ \text{as unity}\end{pmatrix}}{\begin{pmatrix}O.F.\ \text{of ideal}\\ \text{parallel orien-}\\ \text{tation}\end{pmatrix} - \begin{pmatrix}O.F.\ \text{of ideal random}\\ \text{orientation considered}\\ \text{as unity}\end{pmatrix}}\right] \times 100$$

where, *O.F.* is the orientation factor, which is equivalent to

(*OOL*/*OLO*) peak intensity ratios on horizontal plane or on vertical plane or ratio of both

$L = 1, 2, 3$, integers which refer to Miller indices.

Horizontal plane : Plane perpendicular to the direction of applied pressure which provides the arrangement of clay particles on vertical cross section of the clay sample.

Vertical plane : Plane parallel to the direction of applied pressure, which provides the arrangement of clay particles on horizontal cross section of the clay samples.

The above expression for '*R*' implies that *R* is directly proportional to *O.F.* of the clay minus unity. This is satisfactory in as much as the X-ray diffraction peak heights (or intensities) are directly proportional to the number of basal planes (or number of clay particles) which diffract the X-rays at a particular Bragg angle (Brindley, 1961). The above expression is an improvement over the existing expressions developed for kaolinitic and illitic clays by Martin and Ladd (1970), Diamond (1971) and Quigley and Ogunbadejo (1972). It considers the entire range of degrees of particle orientation in the denominator as an accepted datum. The *R* values are hundred and zero for the ideal parallel and the ideal random particle orientations respectively. They also provide upper and lower bound values for the particle orientations. The particle orientation of clays estimated by this method is considered similar to the relative density of cohesionless soils.

Experimental Investigation

General

Commercial bentonite containing chiefly montmorillonite clay mineral was used in this investigation. It contains small amounts of feldspar and silt (See Figure 1.). The bentonite is hereafter called as montmorillonite. The important properties of the montmorillonite are presented in Table. I.

TABLE I

Properties of Bentonite (montmorillonite)

I. Engineering Properties :	
(*i*) Specific gravity	2.40
(*ii*) Liquid limit	420 %
(*iii*) Plastic limit	36 %
(*iv*) Shrinkage limit	10 %
(*v*) Percentage of free swell (at O.M.C.)	177
(*vi*) Maximum swell pressure (at O.M.C.)	7.38 kg/cm^2
II. Chemical properties :	
(*i*) pH value	9
(*ii*) amount of dissolved Salts	1.16 %
(*iii*) Organic content (wet combustion method)	1.17 %
(*iv*) Base exchange capacity (Ammonia distillation method)	89.60 meq/100 gm
(*v*) Percentage of exchangeable ions	
Na+ (flame Photometer method)	58 %
K+ (flame Photometer method)	2 %
Ca++ (Oxalate method)	34 %
Other ions	6 %

Preparation of samples to represent ideal random and ideal parallel particle orientations

Preparation of the clay samples to represent ideal random particle orientation is very difficult (Thomson, Duthie and Wilson, 1972). Samples were prepared in this investigation by impregnating the montmorillonite dry powder with the carbowax 6000 as suggested by Martin (1966) and by Quigley and Ogunbadejo (1972) for kaolinitic and illitic clays respectively. Similarly preparation of clay samples to represent ideal parallel particle orientation is also difficult (Brindley, 1961; Gibbs, 1965; Schaw, 1972). These samples were prepared by modifying the procedures described by Diamond (1966, 1971), Gibbs (1965) and Spoljanic (1971) to suit for montmorillonite clays based upon the findings of preliminary investigations to determine the speed and the time of centrifuging as well as the amount of clay in suspension for the preparation of well oriented aggregates. The preliminary investigation showed that, for the montmorillonite studied, the samples prepared by centrifuging the clay suspension (5 percent) on to the glass slides in centrifuge tubes for a period of 30 minutes yield a maximum *O.F.* value of 30. It is assumed that the clay particle represents maximum (ideal) parallel orientation at this maximum *O.F.* value.

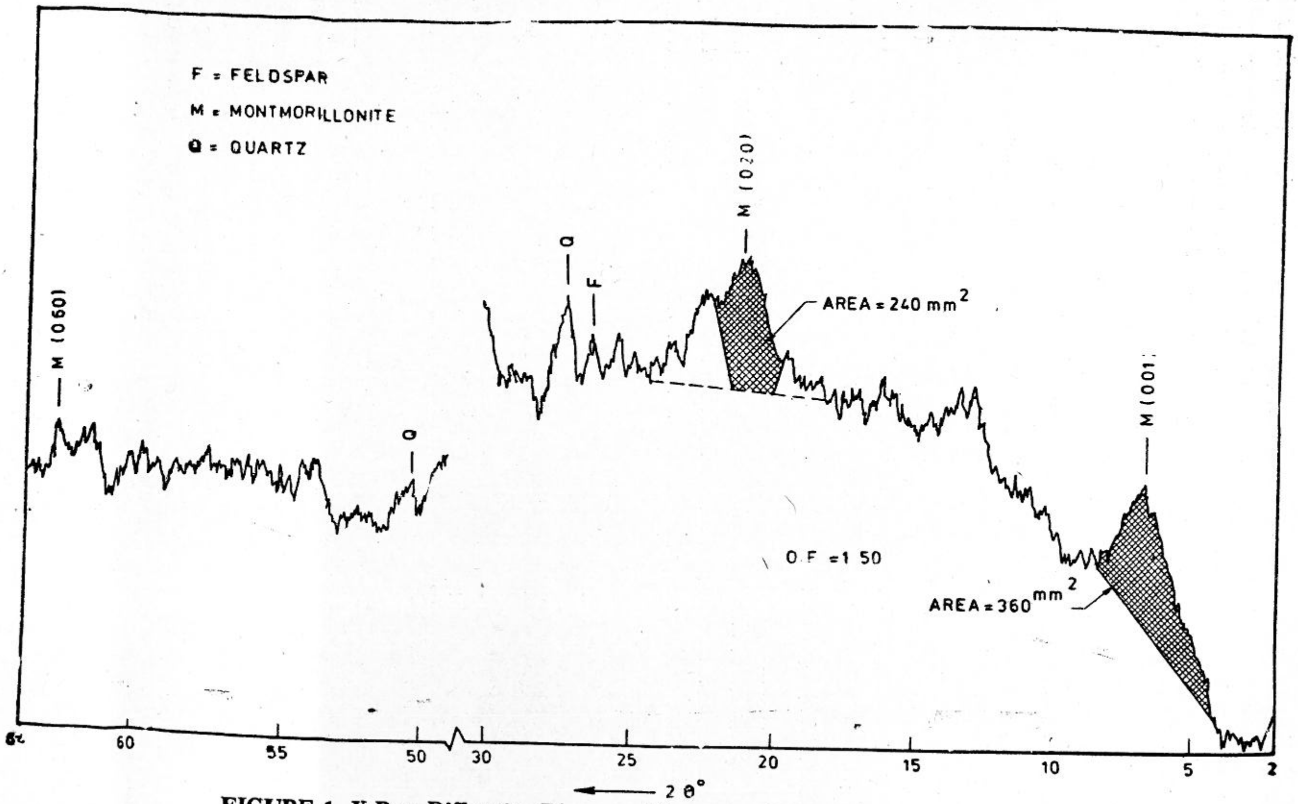

FIGURE 1. X-Ray Diffraction Diagram of Bentonite (Montmorillonite, Powder Method)

One dimensional consolidation and rebound tests

One dimensional consolidation tests were carried out on the montmorillonite slurry from liquidity index of 1.55 in the conventional oedometers. The clay slurry was cured for more than 15 days prior to testing. Nine numbers of identical consolidation tests were carried out to represent the various maximum consolidation pressures of 0.1, 0.2, 0.4, 0.8, 1.6, 3.2, 6.4, 13.8, and 18.0 kg/cm². A load increment ratio of unity was used at 24 hours interval.

Six numbers of identical rebound tests were also carried out from a maximum consolidation pressure of 6.4 kg/cm² to represent various rebound pressures of 3.2, 1.6, 0.8, 0.4, 0.2, 0.1 kg/cm². The rebound characteristics of the clays are studied from 6.4 kg/cm² consolidation pressure, because the limiting equilibrium pressure of the clay is about 6.4 kg/cm². The consolidation (compression) and the rebound (swelling) characteristics of the clay are governed by the physico-chemical forces within this range of pressure (Sankaran and Venkateshwar Rao, 1974; Venkateshwar Rao, 1973). Typical consolidation and rebound characteristics of the montmorillionite are shown in Figure 2.

Impregnation

After the equilibrium condition for the clay specimen at the desired maximum consolidation pressure in the oedometer is attained, the load was removed immediately and the specimen was not allowed to imbibe water. The sample was extruded carefully from the consolidation ring and cut into about 3.8 cm x 3.8 cm pieces by fine wire. Thereafter, these samples were immersed in the molten carbowax 6000 at about 55°C-65°C using the technique of Mitchell (1956). This procedure may cause the elastic rebound of strains with clay particles to some extent. As sufficient time is not allowed for the clay to imbibe water, the diffuse double layers between the clay particles in the clay mass might not have re-formed. Shrinkage forces developed may also restrict the elastic recovery of the strains upon immersion in the hot molten carbowax 6000. Therefore, it is surmised that the resultant particle orientation due to these effects is very close to the in situ particle orientation.

X-ray diffraction studies

Sufficient care was taken to prepare the impregnated samples, before mounting into X-ray diffractometer as suggested by Barden and Sides (1971) and Quigley and Thompson (1966). An automatic X-ray diffractometer, Russian model *y P C* 50, was used with an input voltage of 30 *KVA* and amperage of 10 *ma*. Copper target *Cuk* α was used to produce a beam of X-rays of wave length $\lambda = 1.54\ A^{\circ}$. An average 2-3 chart recordings were taken for each of the clay samples. The details of the test procedure are described by Venkateshwar Rao (1973).

Optical microscopic observations

From each sample that was exposed to X-ray radiation, two identical thin-section (30μ thick) slides were prepared to represent particle orientation on horizontal plane i.e., on plane perpendicular to the direction of consolidation pressure and two identical thin sections to represent particle orientation on vertical plane, i.e., on plane parallel to the direction of

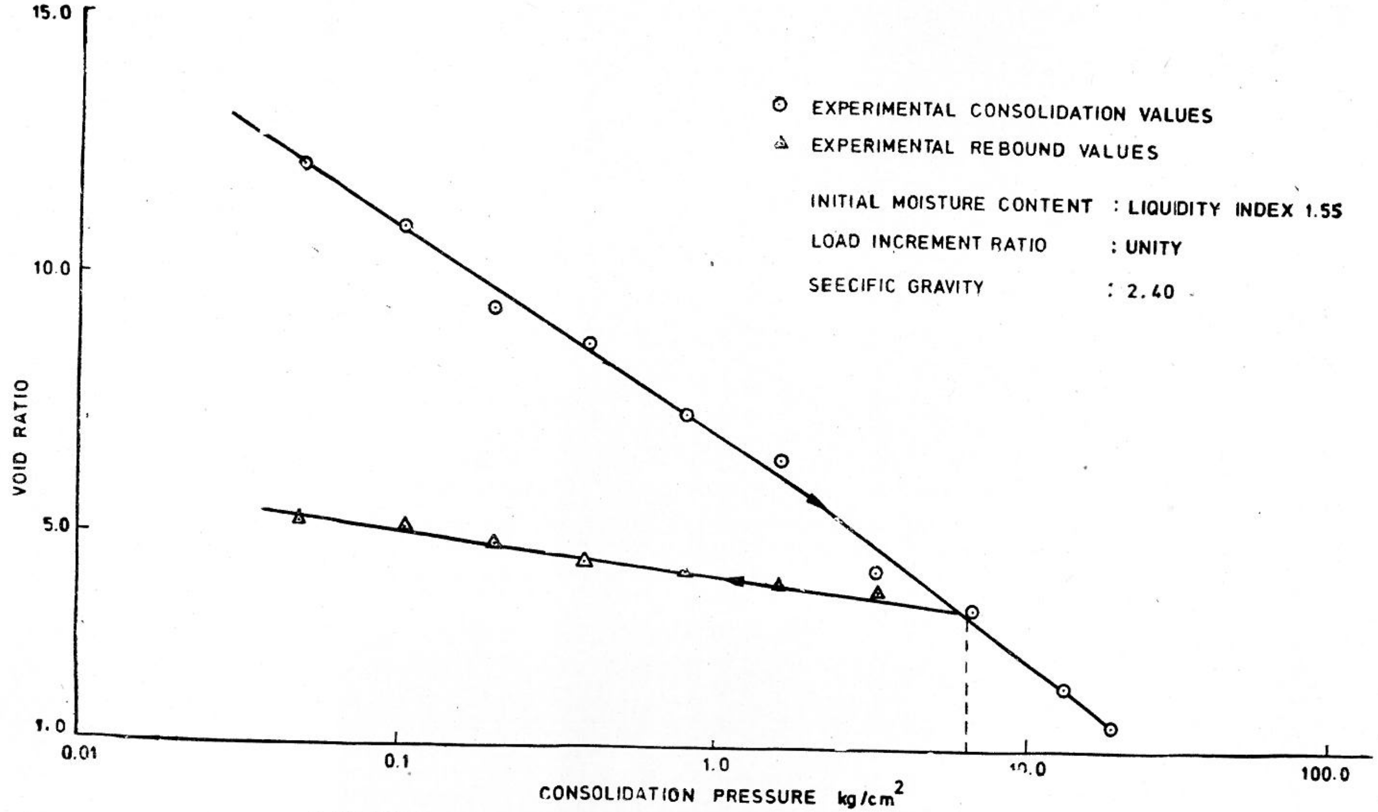

FIGURE 2 : Consolidation and Rebound Characteristics of Montmorillonite

consolidation pressure. These thin-sections were prepared in accordance with the petrofabric standard procedure (Kerr, 1959; Singh, 1967). Intensities of light transmitted through the thin-sections in a polarising microscope (Leitz-wetzler Co. Ortholux-1), using cross nicols under constant input of light-source at extinction (I_{min}) and at illumination (I_{max}) conditions, were measured by means of a photocell in terms of galvanometer deflections. Minimum of 5 observations were made for each slide at different places as suggested by Mitchell (1956). The details of test procedure adopted in this investigation are described by Venkateshwar Rao (1973). The method of estimation of the particle orientation by the two and three dimensional theories proposed by Morgenstern and Tchalenko (1967) based on 'Birefringence ratio', $\beta = (I_{min}/I_{max})$, *is utilised in this investigation for* montmorillonite. The zero and hundred of the '*R*' values correspond to the values one and zero of orientation ratios of Morgenstern and Tchalenko (1967).

Test Results and Analysis

General

It is appropriate to discuss the limitations of the laboratory investigations before presenting the results and their analysis. An ideal condition to study the particle orientation would be under in situ load conditions. To preserve the clay fabric at in situ conditions by replacement of pore water by a suitable embedding medium is a difficult task. Therefore, the particle orientation studies were carried out on samples impregnated with carbowax 6000 immediately after the in situ load was removed. This process causes the following inherent defects in the preparation of samples.

1. The most important defect of the carbowax 6000 impregnation is the shrinkage effect on the sample. The samples which were subjected to maximum consolidation pressures greater than 0.4 kg/cm² in a given test series did not show any signs of cracks or disintegration, when immersed in the molten carbowax 6000 at 55°-65° C. But the samples which were subjected to consolidation pressures less than 0.4 kg/cm² were found to show, invariably, signs of cracks upon immersion. The carbowax 6000 impregnation for minimum of 7 days and cooling at room temperature resulted in volume and axial shrinkages of the samples as shown in Figure 3. The shrinkage effect in the impregnated montmorillonite is an unavoidable feature, and is the characteristic of typical montmorillonite clays. Degree of shrinkage in clays is dependent upon the initial particle orientation. Clays possessing initial ideal parallel particle orientation shrink more in the direction perpendicular to the preferred orientation, and shrink less in the plane of preferred orientation. Thus, the relative particle orientation system may not alter. Similarly, ideal random particle orientation in clays causes uniform shrinkage in all directions, and the relative particle orientation is not expected to alter. Therefore, it is assumed that the carbowax 6000 impregnation preserves the relative particle orientation of the clay for intermediate degrees of particle orientation as in in-situ conditions, even though there is a change in the spatial distances.
2. Grinding procedure for the preparation of the thin-sections on flat surface might have also altered the particle orientation at the

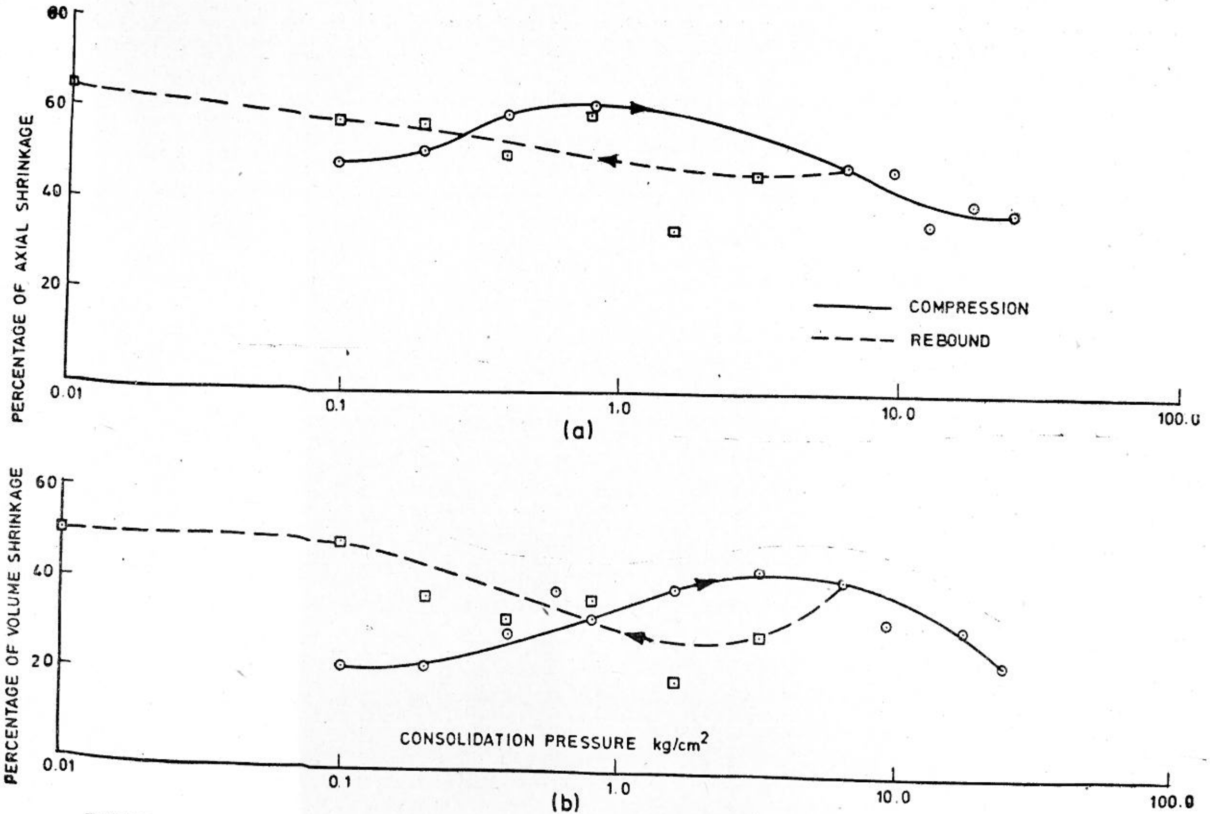

FIGURE 3 : Shrinkage Characteristics of Montmorillonite upon Impregnation with Carbowax 6000 and Cooling to room temperature : (a) Volume shrinkage (b) Axial shrinkage

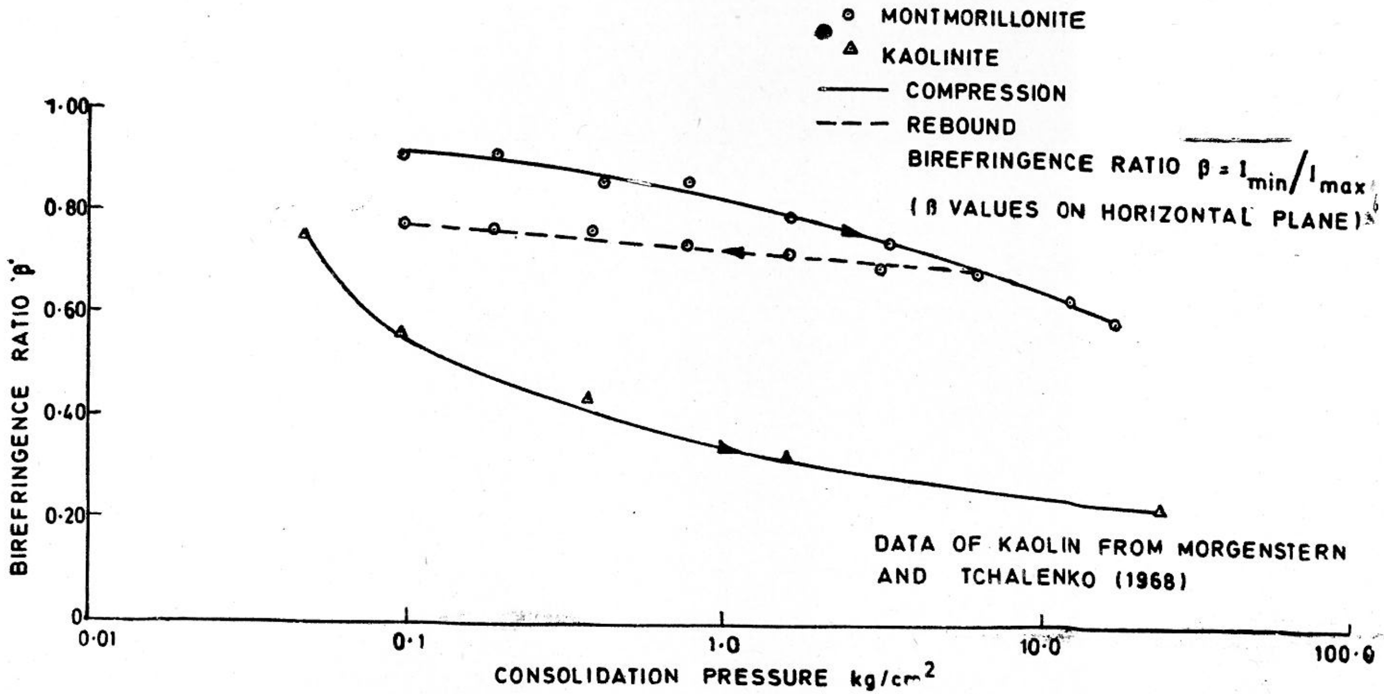

FIGURE 4 : Relation between Birefringence ratio β and consolidation pressure

surface of the samples by smearing effect. However, according to Barden and Sides (1971), the effect of such grinding is only to a depth of 1 μ, and does not affect the particle orientation studies by the X-ray diffraction technique. However, the smearing affects the optical microscopic observations.

Summing up, it may be stated that the particle orientation of in-situ clays is affected by the sample preparation technique, but not very significantly to affect the analysis. Any way, until better sampling techniques are evolved, the above limitations have to be reckoned with.

Analysis of the test results by optical microscopy

The computed values of 'β' from the thin-sections prepared on vertical planes of the clay samples have shown erratic values. This is because thin vertical particles on this plane have a natural tendency to orient in the plane of thin-section by smearing effect during the process of grinding at 30 μ thickness. Hence, 'β' values of thin sections on vertical planes are not considered in this investigation. The particle orientation analysis is carried out from the 'β' values of thin sections on horizontal planes.

It can be seen from Figure. 4 that the 'β' value decreases in a consistent pattern with the increase of consolidation pressure. Similar observations have also been obtained by Morgenstern and Tchalenko (1967) for kaolin. A typical relationship between the *R* value and the consolidation pressure computed by the two and three dimensional theories is shown in Figure. 5.

There is a consistent pattern of increase in the '*R*' value as the consolidation pressure increases and hence, an increase in the particle orientation with the increase of consolidation pressure as one would expect. The '*R*' values computed based upon the three dimensional theory are found to be higher than those computed by the two dimensional theory. Two distinct slopes are observed on the semilog plot of '*R*' versus consolidation pressure. The slope of the curve changes at a consolidation pressure of about 1.6 kg/cm^2 indicating that the rate of change of '*R*' is high at small pressures.

During rebound, the slope of 'β' as well as of '*R*' versus consolidation pressure on a semilog plot are found to be flat (Figures 4 and 5), suggesting little change in the degree of particle orientation during rebound.

The data on birefringence ratio from Morgenstern and Tchalenko (1967) on kaolinite are also used to compute the '*R*' values, and the results are also plotted in Figures 5 and 6. The rate of change of '*R*' value shows a similar trend to that of montmorillonite. Kaolinite attains 100 percent parallel particle orientation at relatively lower consolidation pressure than montmorillonite.

The above analysis suggests that the montmorillonite clay particles prepared from suspension state require high consolidation pressure (when compared to kaolinite), to achieve 100 percent parallel particle orientation. This is due to the fact that montmorillonite clay particles are thin, flexible, less crystalline and more active than kaolin, as also explained by Bohor

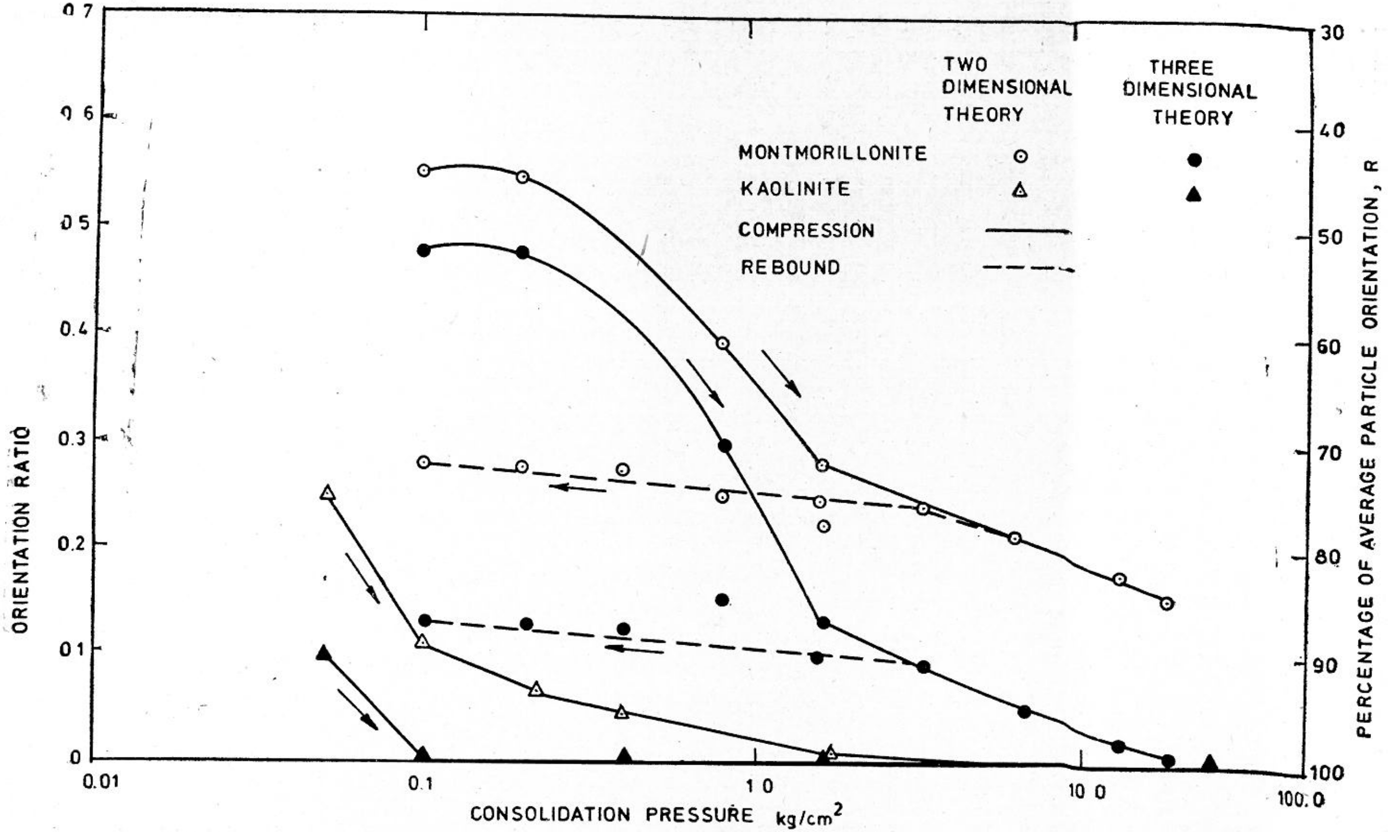

FIGURE 5 : Relation between the average particle orientation, R (by optical method) and consolidation pressures

and Hughes (1970) and Borst and Keller (1969) from their electron microscopic studies.

Analysis of the test results by X-ray diffraction Technique

The characteristic peak reflections of the montmorillonite are (001) basal reflection and (020) and (060) general reflections. These reflections were obtained invariably on the horizontal planes of the clay samples. The (001) and (020) reflections on horizontal planes are found to be very sensitive and show sharp reflections. In the samples which were subjected to high pressures, the (020) reflections are more distinct than (060) reflections. Therefore, the (001) and (020) peak reflections are considered in the analysis to compute orientation factors. Typical peak reflections are shown in Figure 6.

Similar observations have also been made by Guide and Hathaway (1961). Distinct peak reflections are not observed on vertical planes of most of the clay samples even at low speeds of scan. Therefore, the peak reflections of (001) and (020) basal reflections on vertical planes of the clay samples are not considered. Carbowax 6000 has shown peak reflections at $2\theta=15°$, $19°-15'$, $23°-22'$ and $27°—17'$ and they do not affect the peak reflections of montmorillonite.

The montmorillonite samples representing ideal random particle orientation have yielded an *O. F.* value of 2.72 by carbowax 6000 impregnation method. Various research workers (Martin, 1966; Silverman and Bates, 1960) have also obtained the *O. F.* values for kaolinite and illite greater than two for ideal random particle orientation by employing rigorous experimental techniques. Therefore the montmorillonite samples prepared to represent the ideal random orientation are satisfactory. Samples of dry clay powder packed in the aluminium mount of *X*-ray diffractometer have yielded an average *O. F.* value of 1.50 as shown in Figure 1. This value is very close to unity obtained by the theoretical considerations (Fisher, 1953). Therefore, the *O. F.* value of unity is considered in this analysis for the ideal random orientation.

A maximum value of *O. F.* of 30 is obtained on the horizontal planes of samples representing ideal parallel particle orientation by the centrifuge technique. Peak reflections on vertical planes of these oriented samples could not be obtained. Therefore, the *O. F.* value of 30 on horizontal plane is considered in this analysis for ideal parallel orientation.

A typical relationship between the '*R*' value and the consolidation pressure is shown on semilog plot in Figure 7. As the consolidation pressure incrceases, there is a consistent pattern of increase in the '*R*' value; in other words, particles have become more oriented towards parallel orientation in a consistent manner as consolidation pressure increases. This is the expected behaviour. Two distinct slopes are observed on the semilog plot of '*R*' versus consolidation pressure. The slope changes at about 1.6 kg/cm^2 consolidation pressure. Similar, observation has also been made in the analysis by the optical method. At low pressures, the rate of change of '*R*' value with pressure is observed to be less than that at high pressures. At the maximum consolidation pressure of 18 kg/cm^2 the '*R*' value is 76.70 per cent. On rebound from 6.4 to 0.1 kg/cm^2, the slope of

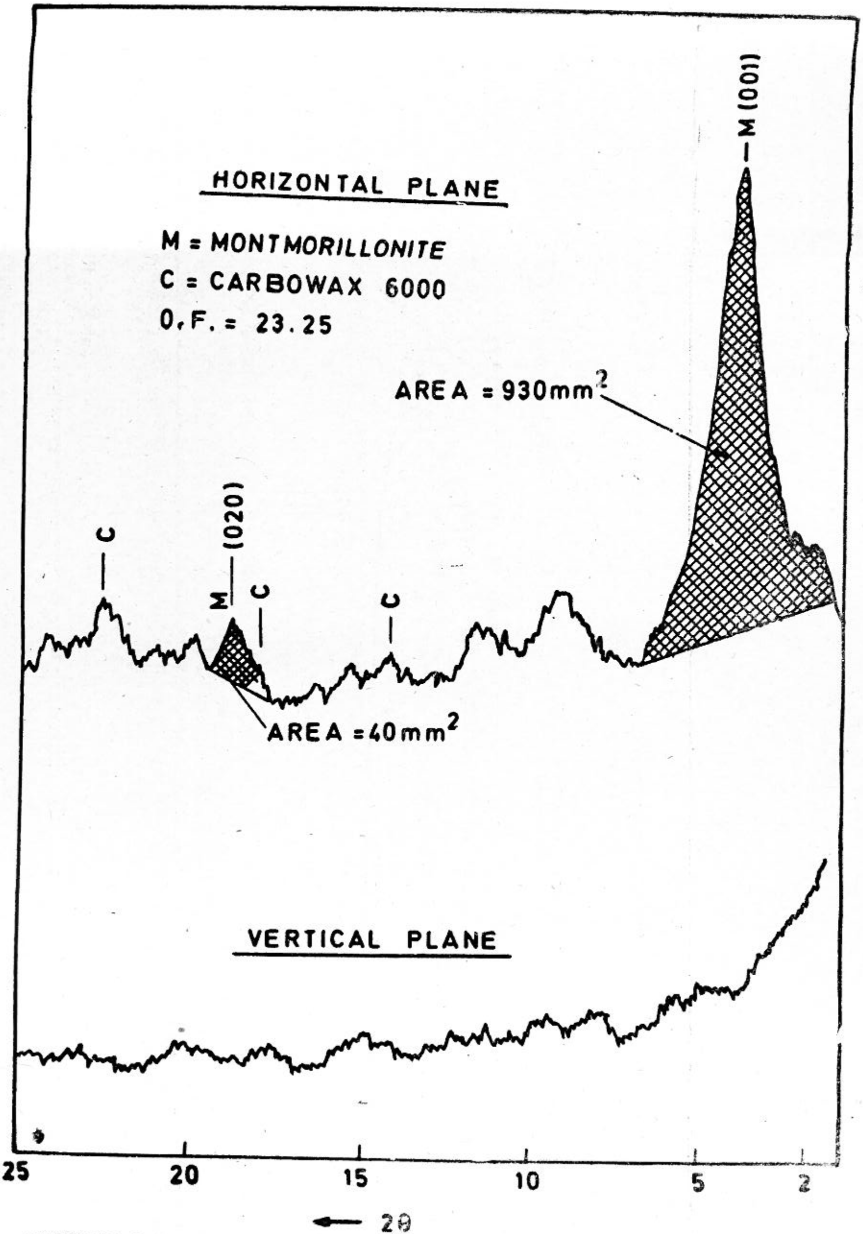

FIGURE 6: X-Ray Diffraction Diagram of Montmorillonite Sample Subjected to 18 kg/cm² Consolidation Pressure

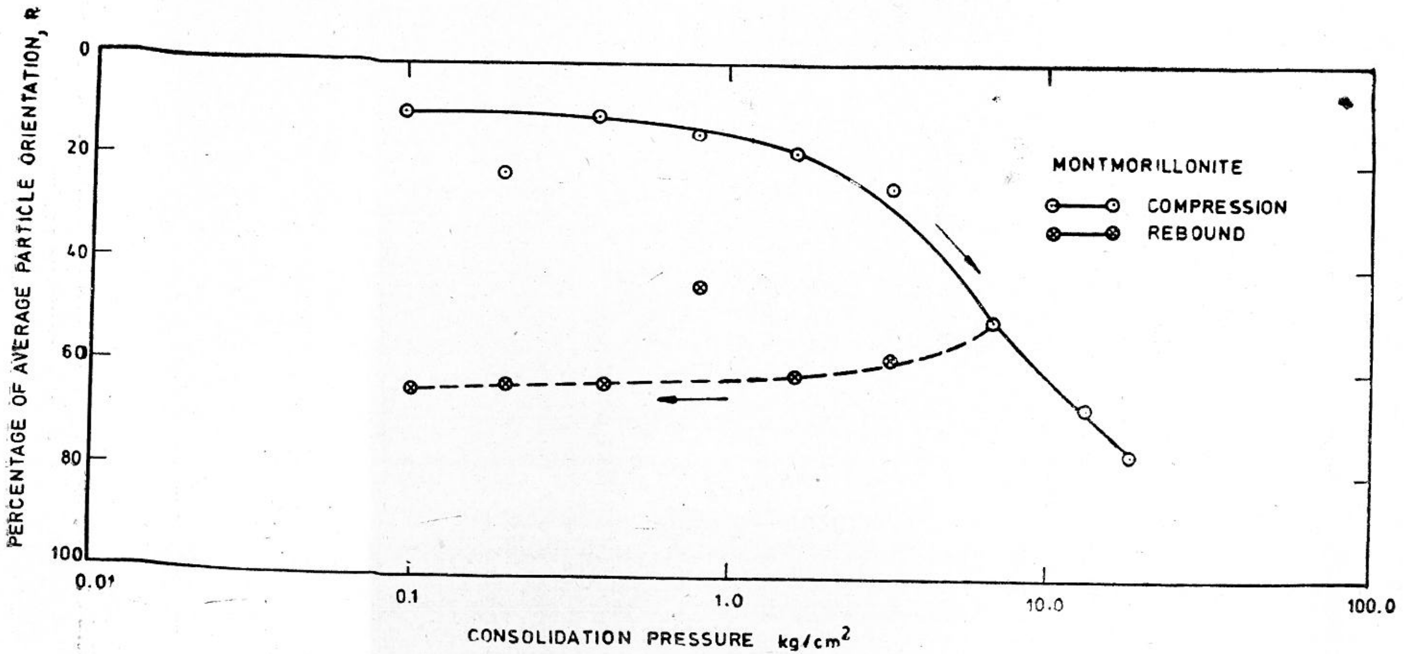

FIGURE 7 : Relation between the percentage of average particle orientation R (By X-Ray Diffraction Analysis) and consolidation pressures.

'*R*' versus log pressure is a straight line with a flat slope suggesting that the net change in particle orientation due to rebound is very small as theoretically shown by Sankaran and Venkateshwar Rao (1973).

Comparison between the Optical Observations and the X-ray Diffraction Analysis

The values of '*R*' at various consolidation and rebound pressures estimated by the optical microscopic method are found to be higher than those obtained for the same samples by the *X*-ray diffraction analysis as can be seen from Figures 5 and 7.

The values of '*R*' obtained by the optical microscopic method using two dimensional theory of particle orientation are relatively closer to those obtained by the *X*-ray diffraction analysis.

The rate of change of '*R*' with pressure is found to be higher at lower range of pressures than that at higher range of pressures, in both the optical methods, whereas in the *X*-ray diffraction analysis the rate of change of '*R*' at lower range of pressures is found to be less than that at high pressures. It is well known that montmorillonite clay particles have a tendency to remain in random orientation and resist becoming parallel oriented. Only at higher pressures, they may become closer to parallel orientation. The *R* values obtained from *X*-ray diffraction analysis are in accordance with the conclusions drawn earlier. The attainment of higher rate of '*R*' values by optical methods, especially in the lower range of pressures, is attributed to the smearing effect during grinding at 30μ thickness. It is, therefore, felt that the *X*-ray diffraction analysis provides a more realistic estimation of particle orientation as pointed out by Venkateshwar Rao (1972, 1973).

The above discussion suggests that the new method of expressing the degree of average particle orientation appears to be satisfactory for montmorillonite clays, and that the particle orientation of montmorillonite can be expressed *quantitatively*. The optical microscopic method, apart from overestimating the *R* value, does not appear to express the change in particle orientation with increase in pressure satisfactorily.

The concept of expressing the average particle orientation *quantitatively* by the *R* value for montmorillonite clays can also be applied to find the average particle orientation of various clay mineral systems after the evaluation of *O. F.* values for ideal random and ideal parallel orientations of the clay particles of various mineral systems. The rigorous analysis of this method has been described else where by Venkateshwar Rao (1973).

Determination of the Average Thickness of Clay Particles

An attempt is made to estimate the average thickness of clay particles under various consolidation and rebound pressures using Scherrer's equation with Warren's Correction (Cullity, 1967; Azaroff, 1968). Typical results are presented in Figure 8. It can be seen from Figure 8 that the clay particles breakdown to smaller units initially under consolidation pressure, and aggregation takes place at high consolidation pressure. The average thickness of the clay particles and the amount of degree of particle orientation for different rebound pressures during rebound are found to be

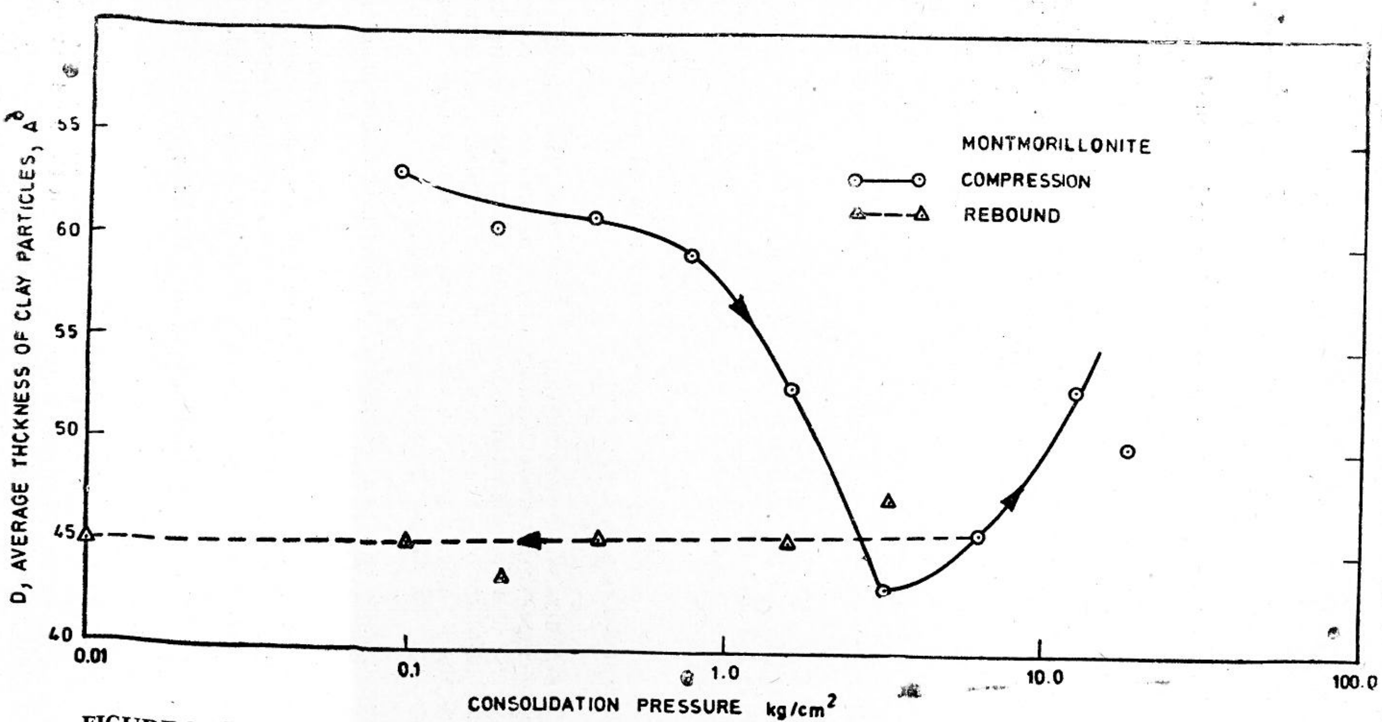

FIGURE 8 : Relation between the average thickness of clay particles (as per Scherrer Equation) and consolidation pressures

essentially the same. These observations are in accordance with the clay model and its mechanistic response of clays, hypothesised by Sankaran and Venkateshwar Rao (1974) and Venkateshwar Rao (1973). However, it must be pointed out that further research work is needed to clarify whether the estimation of the average particle thickness, as scanned by X-rays over the carbowax 6000 impregnated clay samples, represents the true average particle size.

Summary and Conclusions

An attempt is made in the present study to estimate *quantitatively* the average particle orientation of montmorillonite. From the X-ray diffraction analysis, the percentage of the average relative particle orientation, R, is expressed as a function of the orientation factors of *ideal parallel and ideal* random orientations. For ideal parallel particle orientation, the 'R' value is 100 and for ideal random orientation it is zero.

Samples of dry montmorillonite clay powder packed in the aluminium mount of X-ray diffractometer have yielded values of orientation factor closer to the theoretical value of unity for ideal random particle orientation than those obtained by carbowax 6000 impregnation method suggested by Martin (1966) and Quigley and Ogunbadejo (1972) for kaolinite and illitic clays respectively. Samples representing high degree of parallel orientation were prepared in the laboratory by modifying the procedure adopted by Gibbs (1965) and Spoljanic (1971), based upon the preliminary investigation. A *O. F.* value of 30 is obtained for ideal parallel orientation. The orientation ratios suggested by Morgenstern and Tchalenko (1967) for the particle orientation by the optical microscopic method have been made use of to represent the percentage of average particle orientation for comparative studies.

The montmorillonite clay was consolidated and rebounded at various pressures in oedometer. Samples were extruded from these consolidated specimens and were impregnated with carbowax 6000 to study the values of 'R' in terms of the applied external pressure using optical microscopic and X-ray diffraction techniques. They must be viewed with the limitations of the preparation of the samples and of the experimental techniques. The following general conclusions are drawn.

1. There is a consistent pattern of increase in the 'R' value with the increase of external pressure; on rebound, the change in the particle orientation is relatively insignificant irrespective of the method employed.

2. Estimation of 'R' by optical microscopic method yields relatively higher values than those obtained by X-ray diffraction analysis. The values obtained by the optical microscopic method based upon two dimensional theory are relatively closer to those obtained by the X-ray diffraction analysis.

3. Smearing effect is found to be predominant in the thin sections especially for the samples subjected to lower range of consolidation pressures.

4. X-ray diffraction analysis appears to be a better method than the

optical microscopic method for expressing the average particle orientation of clay particles quantitatively.

5. The new method of expressing the average particle orientation *quantitatively* using *X*-ray diffraction analysis appears to be satisfactory for montmorillonite clays. This approach may be useful to various other clay systems.

6. An attempt to estimate the average thickness of clay particles is made from the *X*-ray diffraction analysis and it appears to be satisfactory.

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