Post-depositional structural changes in clay sediments

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POST-DEPOSITIONAL STRUCTURAL CHANGES IN CLAY SEDIMENTS

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"Possibly many may think that the deposition and consolidation of fine-grained mud must be a very simple matter and the results of little interest. However, when carefully studied experimentally, it is soon found to be so complex a question and the results dependent on so many variables that one might feel inclined to abandon the enquiry were it not that so much of the history of our rocks appears to be written in this language."

(Sorby, 1908)

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ABSTRACT (C W HURST)

Post-depositional structural changes in clay sediments

This thesis examines the changes in the microstructure of clays and mudrocks due to the influence of the environment of deposition and subsequent compaction. A sedimentary model is proposed in which the development of fissility in shales and the formation of hydrocarbon source rocks are attributed to the chemical conditions at the time of deposition. The geotechnical properties of fresh sediments are considered and the ability of the depositional environment to affect these properties is assessed.

From a SEM analysis of mudrocks it is shown that there is a correlation between microstructure and the conditions at the time of deposition. Organic-rich shales formed in anoxic environments are characterized by a preferred orientation of microstructure. Mudrocks formed under oxic conditions show a random orientation. This characteristic fabric of shales results from the peptizing capability of certain organic compounds in the environment of deposition and is considered to be a contributing factor to the development of fissility.

A laboratory simulation of the depositional environment is described. Test results show that pure clays sedimented with organic compounds in a marine environment exhibit an increased parallelism of particles compared with those without organic compounds. A mechanism is described whereby the organic compounds are adsorbed onto the clay particle surfaces and promote peptization.

The fabric of sediments obtained under laboratory conditions is found to control the rate of consolidation. Clays with a preferred particle orientation have slower rates of water loss and this is suggested to be an important factor with respect to the formation of under-compacted mudrocks.

Statistical methods applied to the analysis of directional data obtained from SEM micrographs are reviewed and a measure for the anisotropy index of mudrock microstructure is proposed.

CHAPTER 1

INTRODUCTION

1.1 The nature of the problem

The research undertaken in this study is concerned with the microstructure of clays and mudrocks and how this changes from initial deposition to full induration. The geotechnical properties of natural sediments and those prepared artificially are also discussed. In particular a model for the development of fissility is proposed which has a bearing upon the formation of petroleum. The microstructure of clay sediments and mudrocks has been studied by numerous workers, particularly within the last twenty years. The introduction of the electron microscope, with its ability to provide a detailed image of microstructure, did much to encourage this work. One avenue of study which has raised much discussion concerns the phenomenon of fissility, as displayed by those mudrocks called shales. Fissility is defined as the tendency of a rock to split along relatively smooth surfaces parallel to bedding (Pettijohn 1975 p263). The primary cause of fissility has been attributed to a variety of factors and its origin is one fundamental question which has remained unresolved.

There are basically two conflicting ideas that have been put forward to explain the reason why shales are fissile. Early work by Ingram (1953) and supported later by other workers (Gipson 1965, Odom 1967, O'Brien 1970) points to the fact that fissility is dependent on the parallel orientation of platy mineral grains. In contrast to this are those who suggest that the importance of parallel orientation is overestimated and that laminations are the controlling mechanism of fissility in shales (Spears 1976, Curtis et al 1980).

There is no disputing that some mudrocks tend to part along laminations but there are instances where this is not the case. Lundegard and Samuels (1980) pointed out that while lamination generally occurs in fissile mudrocks it is not common for laminated mudrocks to be fissile. They concluded that fissility is not a necessary product of lamination. In some cases where lamination is thought to be the controlling mechanism, it is to be noted that organic material, concentrated in bands, imparts the fissile structure, thus suggesting an organic influence on fissility (Odom 1967). The role that organic matter plays in the formation of fissile shale is central to the model that the author presents in this study.

If one accepts that platy minerals need to be aligned in a preferred direction in order that fissility can occur, then the question arises as to how and when this fabric is produced. There are a number of theories to explain the inducement of parallel orientation or conversely the lack of it. Re-orientation during compaction has been suggested as one means by which the aligned fabric is produced (Hedberg 1936, Meade 1966). This is supported by the work of Clark (1970) who has shown that an orientation of clay mineral particles can arise from uni-directional compaction. However, as White (1961) points out, if compaction was the sole cause of preferred orientation, and hence fissility, one would expect fissility to increase with increasing depth of burial. He concludes that from the evidence of drill cores this is clearly not the case. Compaction does have a role to play but it is more than likely one of bringing closer together the particles of an already established fabric.

It is probable then, that some additional process is at work to assist a preferred alignment to take place prior to any major compaction. Byers (1974) suggested a biogenic origin to fissility in shales. His view was that although fissility is related to the parallel orientation of mineral grains in the rock, this original depositional fabric will only be preserved in the absence of bioturbation. This he suggested is due to abiotic conditions in the original environment of deposition. It is accepted that some disturbance of fabric will arise from benthic activity in the sediment. However it cannot be mere coincidence that the conditions restricting burrowing infauna are those which have a dramatic effect on sedimenting clay minerals.

Clay minerals have various properties which induce a behaviour that is unique amongst clastic sedimentary particles. These properties are responsible for the flocculation or dispersion of clay minerals as determined by the chemistry of the depositional environment. It has been suggested by various authors (Ingram 1953, White 1961, O'Brien 1970, Moon 1972) that clay particles sedimented in a dispersed state will attain the preferred orientation which is evident in some mudrocks. Flocculated clay particles, on the other hand, would finally give rise to a randomly orientated structure. If this is the case then it is conceivable that there is a connection between the environment of deposition and fissility. The chemistry of the depositional environment is thus considered to be an important factor in the development of preferred orientation in shales and random orientation in mudstones. The detailed chemistry involved is not clear but a number of variables such as electrolyte concentration, pH and Eh are thought to be involved.

The suggestion is that the dispersing agents which will allow particles to settle individually and produce a parallel orientated fabric are present in organic-rich marine (or non-marine) environments. Various authors have registered a connection between organic content and shales (Ingram 1953, O'Brien 1968, Leventhal and Shaw 1980), noting that the organic material imparts a dark colour to the rock. This link is well illustrated by the black shales which are regarded as distinct facies in their own right. It is worth emphasizing that the black shales also tend to be highly fissile. Organic compounds capable of causing dispersion are common in a variety of depositional environments. Examples of which are fjords (Grasshof 1975), fjord like basins (Brown et al 1972), ocean deeps (Degens et al 1961, Deuser 1975) and barred basins as typified by the Black Sea (Degens and Stoffers 1980).

An examination of these environments reveals that an organic-rich anoxic zone exists at depths where the bottom waters are stagnant. The usual reason for this is that aerobic decomposition of planktonic organisms depletes the level of dissolved oxygen and in the absence of further oxidisation organic material is free to settle. The organic matter is not consumed then in the usual manner, by oxidation, but is decomposed by the action of anaerobic bacteria in the reducing environment that now exists. It is suggested that clay sediments on entering the anoxic zone have the charges on their particle surface neutralized by the organic material present. The exact mechanism of neutralisation is not yet fully understood but clearly it is dependent on the complex electrochemistry of clay particles and their immediate surroundings. These neutralised particles are deposited in a dispersed state which on subsequent induration produces the parallel orientated structure conducive to the development of fissility. In 'normal' marine conditions no dispersion occurs and the sediment remains flocculated. This gives rise to a random structure which produces a non fissile rock on induration. It is not assumed that dispersion by organic compounds is the only means by which fissility is produced in shales but it is considered to be one mechanism of great importance.

The types of organic compound thought responsible for dispersion may also be the source of material for petroleum. There is general agreement that petroleum has a biological origin, although the nature of the source material and the chemistry of its formation is the subject of argument (Colombo 1967). The usual approach to the problem of identifying the source material is to study the chemical similarities between petroleum and the biogenic compounds in sediments. Erdman (1961) points to fatty acids (lipids) and amino acids (protein) as being main sources for hydrocarbons. A high concentration of free amino acids at the sediment-water inferface has been noted in the reducing environment by Degens (1965) and it is in the reducing environment, under anaerobic conditions, that petroleum is said to be formed. Amine salts are known to cause a dispersion of flocculated clay (Van Olphen 1963) and so it is reasonable to infer that amino acids may contribute to both the fissility of shales and petroleum formation. Other organic compounds (sugars, fatty acids, alcohols) are also possible candidates. The evidence thus suggests that a unique relationship exists between the chemical environment which produces a fissile shale and the generation of petroleum. Part of the aim of this study is to explore that relationship.

It is generally recognised that the geochemical environment at the time of deposition also has some bearing on the geotechnical properties of a sediment. The geochemical conditions influence the microstructure adopted by clay particles and this fabric will affect, in particular, the consolidation and shear properties (Rowe 1972). The term consolidation is used in the engineering sense to mean the reduction in volume of a saturated soil due to the expulsion of water. With a flocculated clay the open, random structure would give a higher void ratio that its dispersed counterpart under the same overburden stress. In the latter case, due to the preferred orientation, the clay particles have a better chance of coming closer together when water is expelled.

In a comparison of samples from deep sea cores with those of the Mississippi Delta, Bennett et al (1977) looked at the fabric of sediments obtained at the same depth of overburden. They found that a randomly orientated structure had almost twice the void ratio of the preferentially orientated one. With regard to strength properties the Delta cores showed a ten times increase in shear strength from the upper high void ratio sediments to the deeper low void ratio ones. They concluded that factors other than depth of burial and overburden stress influence the microstructure and hence consolidation of a particular sedimentary deposit. One of these factors is the geochemical conditions at the depositional interface.

It must be acknowledged at this point, that in many cases larger scale features are more important, particularly in their effect on mechanical anisotropy, than the microstructural features. In a comprehensive study of the geotechnical properties and parameters of British mudrocks compiled by Cripps and Taylor (1981) it was evident that no one factor is responsible for variations in these properties. Amongst their conclusions they suggested that the removal of overburden (uplift and erosion) has a significant influence on engineering behaviour. A response to the change in overburden stress is the creation of joints and fissures and it is likely that the microstructure together with other factors will influence the orientation of these features.

There are many uncertainties surrounding the nature of formation of clay sediments and mudrocks. The number of variables involved in the processes that occur from initial sedimentation to complete diagenesis are numerous and to model the event in its entirety is difficult, although with sufficient funds reasonable attempts have been made by other workers (Knill et al 1976). This study is aimed at reproducing in a simplified manner one of the important features of natural sediment accumulation - the environment of deposition. The simulation is not without faults but it illustrates what may be achieved in the confines of a laboratory using unsophisticated equipment.

1.2 Organisation of the report

The literature survey in Chapter 3 records the various fundamental models of clay microfabric. Recent studies of natural and laboratory prepared samples using the scanning electron microscope (SEM) are presented. Chapter 4 considers some of the environments of deposition in which clays and mudrocks are formed and examines the geochemistry of these environments. On the basis of the evidence presented in this first part of the study a tentative model is suggested for the origin of fissility in shales.

The second part of the study presents the observations and data to support the proposed model. Chapter 5 covers the SEM investigation of sampled mudrocks and tabulates the statistical data for each specimen. The environments of deposition of the various mudrocks are considered with respect to the proposed model. The experimental evidence is given in Chapter 6. The SEM investigation and statistical data for the laboratory prepared samples are presented together with their geotechnical properties.

Chapter 7 discusses the evidence obtained from both SEM investigations in relation to the model offered and reviews the possible implications for petroleum formation.

The conclusions of the study and a consideration of future research are presented in Chapter 8.

The experimental procedures, equipment details, statistical theory, computer programs and sample records are all presented in the Appendices.

A study of mudrocks would not be complete without some reference to the confused terminology which has evolved over the years. It is unlikely that terminology will ever be standardised since the contributions to clay and mudrock research come from a variety of disciplines each with their own interpretations. The classification of mudrock is similarly confusing and discussion on this and terminology is given first in Chapter 2.

CHAPTER 2

FINE-GRAINED SEDIMENTS: TERMINOLOGY AND CLASSIFICATION

2.1 Terminology

There is, without doubt, a great deal of confusion over the terminology used in the description of fine-grained sedimentary rocks. Much of this confusion arises because no widely accepted classification of these rocks exists. Attempts have been made to rectify this situation (Underwood 1967, Picard 1971, Lewan 1978, Spears 1980, Lundegard and Samuels 1980, Stow 1981) but since mudrock terminology is often not consistent in its meaning this has led to confusion. An example is the word 'shale' which may be used in a stratigraphic context, as a class name for a sequence of strata (e.g. Edale Shale), and in a more specific role to define a rock composed of predominantly fine-grained clastic particles. Since the class name of 'Shale' is firmly entrenched in the geological literature (Tourtelot 1960) then most workers accept its dual meaning. The use of the upper-case character helps to reduce any confusion.

The usual interpretation of shale is that the word refers to those sedimentary rocks which possess a characteristic fissile nature. However this definition is not always adhered to in the literature. Shale has been used as a synonym for all fine-grained sedimentary rocks, regardless of specific texture or structure. Hedberg (1936, p 283) defines clay and shale as "loose terms comprising an infinite variety of rocks". His interpretation of a shale must include all clay dominant rocks regardless of whether fissility is present. or not. Fertl (1977 p 294) defines shale as an "earthy, finegrained, sedimentary rock with a specific laminated character". Again, no reference is made to fissility. Pettijohn (1975 p 261) refers to shale as "a laminated or fissile rock" and does not specify that fissility must be present. This is the case with Potter et al (1980) whose classification of shale requires only that laminae <10 mm thick be present. They define fissile simply as a class of parting with thickness between 0.5 and 1.0 mm. Under their classification it is possible for a non-fissile shale to exist. In line with Hedberg, Blatt et al (1980 p 381) confirm that shale is often used as a word to describe the entire class of finegrained sedimentary rocks in which clay minerals predominate. To confound the issue further, Clark's (1954) definition of a shale includes rocks that are widely accepted as siltstones.

In an attempt to alleviate the confusion surrounding the use of the word 'shale' the term 'mudrock' has been proposed (Lundegard and Samuels 1980) although this itself has given rise to some debate (de Freitas 1981). It may be argued on two counts that as a term mudrock is ambiguous. Firstly because mud and rock define opposing physical states and secondly because although its name implies rocks of a siliciclastic composition it can also include rocks that are calcareous or siliceaous. In recognition of the latter, biogenic mudrock has been suggested as an appropriate title for carbonates etc (Stow and Piper, 1984).

For the purpose of this study mudrock is used as a name for those sedimentary rocks composed predominantly of clay and silt sized siliciclastic minerals, where the latter make up more than 50% of the rock. Its usage is somewhat synonymous with argillaceous rock. Under the heading of mudrock lie a range of unlithified and lithified sediments whose terminology is related to the particle size distribution of the sediment and whether or not lamination and/or fissility are present.

Silt-sized material lies between the upper boundary of clay-size which is 4 µm and the lower bound of sand-size which is 63 µm (Wentworth 1922). It is usual to refer to an unlithified sediment with predominantly silt-sized particles as a silt and one of predominantly clay-size as a clay. There is no reference to the mineralogical composition in this instance and a clay need not necessarily be composed of clay minerals. If the mix of silt and clay sizes is not easily definable then the sediment is often described as a mud.

The process of lithification transforms the loose sediment into a mudrock which will either be laminated and/or fissile or it will be massive. The laminated/fissile mudrocks are given the suffix -shale whilst the massive type adopt the suffix -stone. Thus, depending upon the proportions of silt and clay, the rock may be called a clayshale/mudshale/siltshale or a claystone/mudstone/siltstone respectively. This terminology appears logical and devoid of confusion. However it relies upon particle size determination and more often than not the system is simplified. All laminated/fissile mudrocks become shales and the massive mudrocks become mudstones or, if definitely silty, siltstones. This broad usage of the word 'shale' is probably responsible for much of the confusion in mudrock terminology.

2.2 Classification

There is a definite requirement for a unified, descriptive classification of mudrocks. Existing geological classifications are commonly based upon textural and compositional attributes and are biased towards laboratory analysis. Ideally a classification should rely upon parameters which are recognisable both in the field and in the laboratory.

The splitting character, or fissility, of the rock features predominantly in most classifications. However in the majority of cases fissility only becomes apparent at the surface outcrop and is not always evident in fresh core samples (Lundegard and Samuels 1980). Its manifestation would appear to be time dependent, arising as a response to both weathering and a release of confining stresses. On this basis it would not be appropriate to use fissility alone as a classifying parameter for mudrocks since its presence is temporal.

Lamination, on the other hand, is easily recognisable in core samples and at outcrop (Spears 1976, Lundegard and Samuels 1980). Where weathering makes lamination detection difficult at outcrop it is necessary to remove the weathered layer until fresh rock can be examined. By using lamination as the criteria for classification it overcomes the problems associated with applying fissility on its own. This is not to say that fissility is ignored, but rather it becomes a secondary feature as far as sedimentology is concerned, along with colour, mineral content and genetic origin.

The simplest and most appropriate field method of geological classification is that proposed by Lundegard and Samuels (1980) and is given in Table 1. The classification is based upon quantifying just two parameters - silt content and lamination. The determination of silt content need not be based upon precise laboratory methods. Blatt et al (1980 p 382) have suggested a simple method to define the size boundaries, which can be carried out in the field. With regard to lamination the recognition of a fabric-type lamination is not difficult, but grain size and colour lamination can pose a problem. The scheme of Lundegard and Samuels suggests 10% laminae as the minimum necessary to qualify as a shale. In the majority of cases this simple type of classification should suffice. Unless a more precise description is required there should be no need to resort to laboratory classifications like those of Picard (1971) and Lewan (1978).

An engineering classification of mudrocks requires a slightly different approach. Since strength properties feature prominantly in an engineering classification then a consideration of anisotropy is necessary. For example fissility has a significant influence on the bulk properties of mudrocks and is of major importance in geotechnical engineering (Taylor and Spears 1981).

One of the earliest attempts at classifying mudrocks in engineering terms was carried out by Terzaghi (1936). He presented a three-fold division comprising of soft intact clays, stiff intact clays (both free of joints and fissures) and stiff fissured clays. Only the weaker end of the mudrock spectrum was classified. In engineering parlance these mudrocks would most probably be regarded as soils. The reason being that to an engineer a rock is an "aggregate of minerals connected by strong and permament cohesive forces" (Terzaghi and Peck 1967) whereas the weaker mudrocks are easily crumbled in the hand like a soil.

This distinction is not appropriate to this study. To simplify matters the strength parameters of the Working Party Report on rock masses (Anon 1977) have been adopted. Mudrocks span the whole strength spectrum of soils and rocks and are only distinguished from 'clay sediments' by the fact that the latter may include freshly deposited material.

Underwood's (1967) classification takes into account strength properties and he divided shales (which to him were mudrocks possessing fissility) into 'soil-like' which are only compacted and 'rock-like' which are compacted and cemented. The diagenetic bonding of mudrocks is important in engineering classifications but has no significance geologically. A typical engineering classification of mudrocks is given in Table 2.

The mudrocks sampled for this study range in strength from strongly cemented to weakly consolidated. Details of the samples are given in Appendix V and analysis of their structure presented in

As a postscript to this chapter a comment of the prominance of mudrocks in the geological literature is warranted. The number of articles published specifically on mudrocks is paltry compared with the other sedimentary rocks. To quote a typical example, in their book Blatt et al (1980) devote about 100 pages to sandstones, 90 pages to carbonates but a mere 30 pages to mudrocks. This ratio is even worse in the general sedimentary journals. This is due, perhaps, to the inherent difficulties with their investigation, such as poor sample recovery from boreholes. Because of their importance both geologically, as petroleum source rocks, and in engineering terms, with respect to foundation and slope problems, it is to be hoped that more attention is directed to their understanding.

		SILT FRACTION		
		2/3	1/3	
INDURATED	non-laminated	SILTSTONE	MUDSTONE	CLAYSTONE
	laminated		MUDSHALE	CLAYSHALE

Table 1: Field classification of mudrock.
Classification is based on grain size
and stratification
(After Lundegard & Samuels (1980))

Siltstone/claystone + Fissility = Shale			
'Soil-like' shale (compaction or subshale)		'Rock-like' shale (cemented or bonded)	
Clayey shale (clay shale)	50% or more clay-sized particles which may or may not be true clay minerals	Clay-bonded shale	Welded by re-crystallization of clay minerals, or by other diagenetic bonds
Silty shale	25-45% silt sized particles. Silt may be in thin layers between clayey shale bands	Calcareous shale	20-35% $CaCO_3$ (Marls and shaley chalk 35-65% $CaCO_3$)
Sandy shale	25-45% sand sized particles. Sand may be in thin layers between clayey shale bands	Siliceous shale	70-85% amorphous silica often highly siliceous volcanic ash (quartzose shale-detrital quartz)
Black shale	Organic rich, splits into thin semi-flexible sheets	Ferruginous shale	(25-35% Fe_2O_3) (potassic shale - 5 - 10% potash)
		Carbonaceous shale	Carbonaceous matter (3-15%) tends to bond constituents together and imparts a certain degree of toughness

Table 2: Classification of shales (after Underwood (1967))

CHAPTER 3

PREVIOUS WORK

Fine-grained sediments have been estimated to account for 80% of the global rock column (Clarke 1924) and yet comparatively very little work has been devoted to analysing their microstructure. There are obvious reasons for this, namely their poor exposure, extremely fine texture and complex mineral composition. Interest in these sediments has fluctuated with the development of improved techniques to investigate their structure. One of the major improvements was the introduction of the electron microscope and this instrument has become an essential tool for use in the examination of clays and mudrocks. It is a credit to workers in the era before the application of the electron microscope to fabric studies that their models of clay microstructure are not widely dissimilar to present day concepts.

3.1 Fundamental models of clay microstructure

Evidence from natural and artificial clay sediments suggests that there are many different types of microstructure present in these soils. A number of fundamental models have been proposed and these are presented here.

The early models of clay microfabric were based upon the assumption that clay particles existed as single units. These models may be regarded as belonging to either the honeycomb structure, as proposed by Terzaghi (1925), or the cardhouse structure, adopted by Goldsmidt

(1926) and later modified by Lambe (1953). These structures are illustrated in Figs. 1 and 2. A subsequent report by Lambe (1958) revealed three modes of clay structure whose formation was primarily dependent upon electrolyte concentration (Fig. 3). The importance of electrolyte concentration in controlling the arrangement of clay particles was discussed at length by Van Olphen (1963). His hypothesis, based upon clay colloid chemistry, predicts a number of possible particle associations (Fig. 4). Von Englehardt and Gaida (1963) proposed a modified cardhouse model which they called an aggregate structure (Fig. 5). The aggregates are composed of many single clay particles having edge-to-face contacts.

A major development in microfabric studies was the introduction of the domain concept (Aylmore and Quirk 1959). A domain is a microscopic or submicroscopic region within which several clay particles are in parallel array. Domains have also been referred to as stacks, packets, tactoids or clusters by other workers. Aylmore and Quirk (1960) suggested the term turbostratic structure for a group of domains in a turbulent array (Fig. 6). A more open type of turbostratic arrangement was described by Sloane and Kell (1966) as a bookhouse structure (Fig. 7). Further evidence for the domain assembly was provided by Smalley and Cabrera (1969) who identified a stepped face-to-face particle association (Fig. 8) which was considered to be more stable than the book structure of Sloane and Kell.

The foregoing concepts in structure are illustrated by two dimensional analogues since their natural three dimensional structures

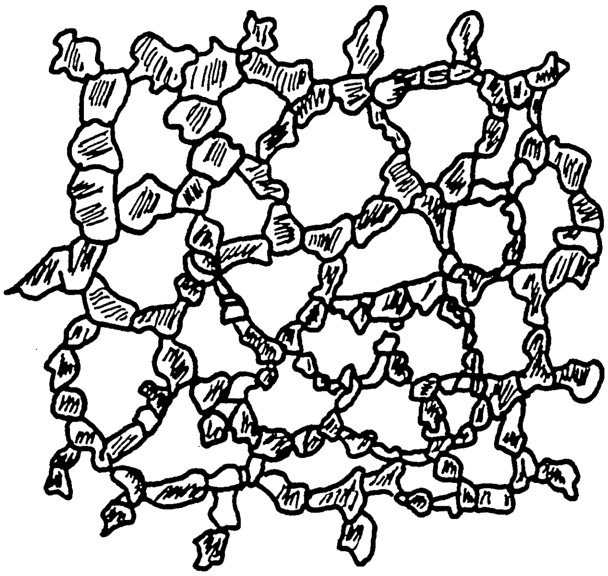


Figure 1 HONEYCOMB STRUCTURE
(Terzaghi-Casagrande 1925)

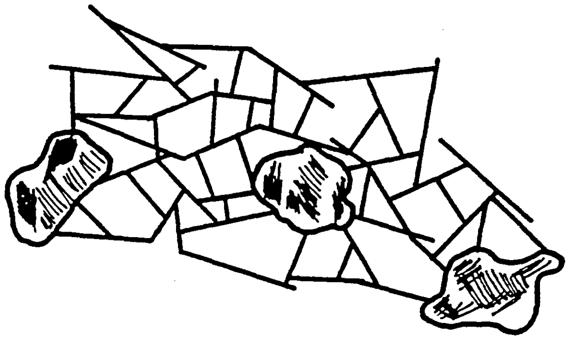
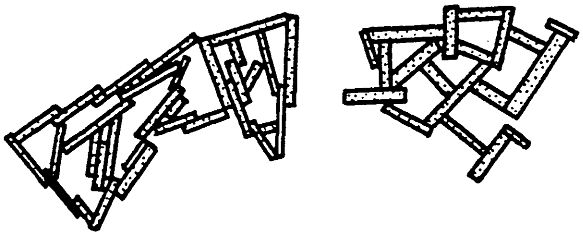


Figure 2 CARDHOUSE STRUCTURE, SALTWATER DEPOSIT
(Goldschmidt-Lambe 1953)



(a)salt flocculation (b)non-salt flocculation

Figure 3 SEDIMENT STRUCTURES
(Lambe 1958)

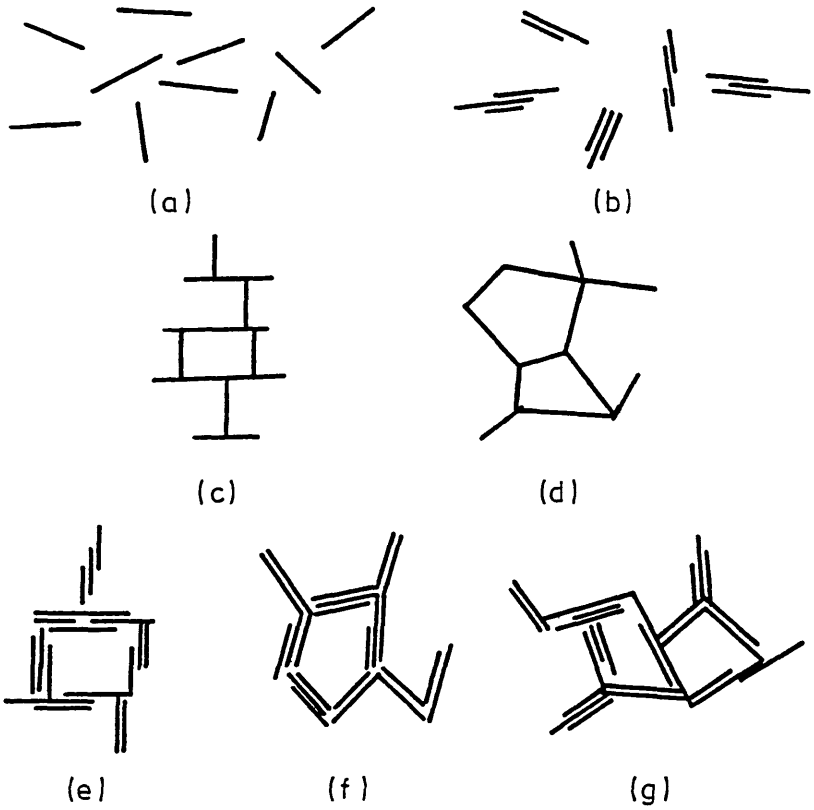


Figure 4 MODES OF PARTICLE ASSOCIATION IN CLAY SUSPENSIONS
(Van Olphen 1963)
(a) dispersed (b) aggregated (FF) (c) flocculated (EF)
(d) flocculated (EE) (e) flocculated (EF) and aggregated
(f) flocculated (EE) and aggregated
(g) flocculated (EF & EE) and aggregrated

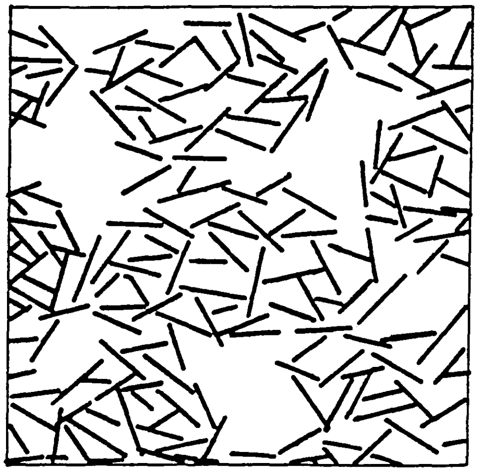


Figure 5 THE AGGREGATE STRUCTURE OF VON ENGELHARDT AND GAIDA (1963)

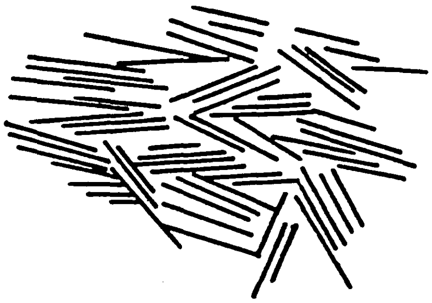


Figure 6 "TURBOSTRATIC" STRUCTURE
(Aylmore and Quirk 1960)

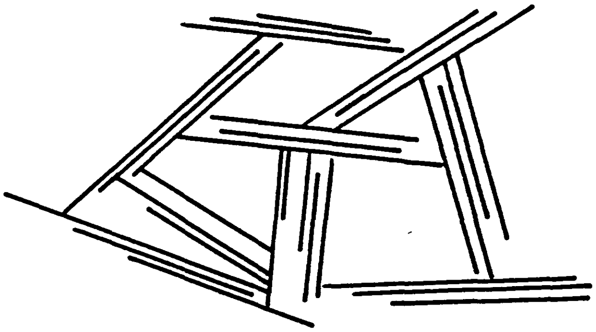


Figure 7 BOOKHOUSE OR BOOK STRUCTURE
(Sloan and Kell 1966)

Figure 8 DOMAIN STRUCTURES "STEPPED FACE-TO-FACE"
(Smalley and Cabrera 1969)

are quite complex. Tan (1959) and Pusch (1970) are two workers who have provided 3D impressions of clay particle networks and with the scanning electron microscope true 3D observations are now possible. The time consumimmg process of obtaining data from these 3D images has negated their usefulness to fabric studies. A semi-automatic stereoscopic analysis method has been examined by Tovey (1978) but little commercial development has been forthcoming to date (Tovey, personal communication).

The distinction between single plate structures of the cardhouse type and the domain structures has been recorded by Barden (1972). He has suggested that the cardhouse structure is only likely to exist in dilute colloidal suspensions (i.e. during particle settling) and its relevance to consolidated natural clays is questionable. Even at or very near the depositional interface (or mudline) particle clusters will take up the domain arrangement when the clay concentration is high enough (Moon 1972). During settling, pelletization by planktonic organisms (Ernissee and Abbott 1975, McCave 1975, McCall 1979) and agitation of the water will produce domains by the agglomoration of particles. Once domains have undergone some light compaction it is unlikely that they will be destroyed, although with excessive shear a dislocation and slippage between particles could occur. Diagenetic processes force domains very close together to the extent that they become difficult to recognise in highly compacted mudrocks. In laboratory consolidated samples of kaolin slurries McConnachie (1974) identified domains after pressure of 10^5 kPa, and they were in fact recognisable from the outset. It seems logical that domains should be present in mudrocks even though they are difficult to detect.

3.2 The microfabric of sedimenting clay

This section will deal with those structures which are believed to be characteristic of dilute clay suspensions, in which the clay particles are settling towards the depositional interface under the influence of gravity alone. Because of the obvious practical difficulties in direct observation and the problems of fabric disturbance when sampling, the concepts of microstructure at this stage are either theoretical or based upon indirect evidence. Consequently few specific findings have been reported.

The nature of clay particles and their reaction to the chemistry of the depositional environment suggests that certain particle associations are possible. As already pointed out, Van Olphen (1963) has contributed much to the understanding of clay particle interactions. He suggested that three different modes of particle association may occur on flocculation: face-to-face (FF), edge-to-face (EF) and edge-to-edge (EE). The FF association is the domain type structure which Van Olphen called an aggregate, whilst the EF and EE modes provide the basis of the cardhouse structure which he considers as flocculated.

Rieke and Chilirganian (1974) claim to have observed fabrics in clay-in-water suspensions ("thick mud") which seemingly bear resemblance to some of Van Olphen's associations. However, they do not quantify the amount of clay in the suspension and so a direct comparison with Van Olphen's colloid situation is not practical.

According to their model single clay particles, initially scattered thinly about the water medium, form a cardhouse structure of EF configuration in thickening suspensions. Neutralisation of repulsive surface forces on the faces allow plate to plate (cf FF) floccules to form and thence domain type structures begin to develop.

The flocculation process results in the formation of EE and EF contacts between either single particles or small domains and the unit built up by these contacts is called the floc. However, this process is not irreversible and flocs may dissociate themselves by dispersion if the environment changes. Whitehouse et al (1960) regarded the settling unit (or floc) as a thermodynamically reversible assembly which they termed a 'coacervate'. They indicated that the aggregation of clay minerals by saline waters is reversible. The 'break up' of flocs is more likely to occur above the mudline but there is no reason to suggest that it may not happen at or just below the mudline where compaction is minimal.

The flocculation characteristics and hence settling behaviour of a sediment have been attributed to its mineralogy (Whitehouse et al 1960) but this has been challenged by Kranck (1975). In a study of suspended sediments from coastal environments Kranck concluded that particle size rather than mineralogy is the controlling factor. This tends to agree with the work of Packham (1962).

An interesting study was made by Schweitzer and Jennings (1971) who indirectly observed montmorillonite associations, in dilute suspensions, using a light scattering technique. Their conclusion was that the EF cardhouse structure was the most likely clay particle arrangement.

From the limited available evidence it seems likely that both single-plate cardhouse type structures and small domain-like packets (say 2 to 3 particles), which may or may not form structured groups, are present in sedimenting clays. Their stability will depend upon the prevailing conditions of pH, Eh and salinity in the depositional environment. The structures themselves may have a dozen or less particles (or domains) arranged in some form of EF array forming a floccule, which is the settling unit. The floccule has a limiting size which is governed by the comparable settling velocity of the largest individual grains (Kranck 1975).

3.3 The microfabric of fresh clay sediments

Fresh clay sediments are considered to be those clays which are present at or just below the mudline and, as such, have not suffered significant compaction by superincumbent material. As with sedimenting clays it is very difficult to sample and prepare specimens for observation without disturbance to the fabric. In view of this, lightly compacted sediments are more often studied, and from these observations the fabric of the uncompacted sediment is inferred. The laboratory sedimentation of mono-mineralic clay suspensions has also yielded much data in spite of the situation being artificial.

The springboard for clay fabric research was the application of the transmission (TEM) and scanning (SEM) electron microscopes. One of the pioneers of TEM work was Rosenqvist whose observations of illitic clays from Scandinavia were to dominate clay microstructure thinking for years. In the marine clays which he examined, Rosenqvist (1959, 1962) was able to confirm (albeit not too convincingly) the existence of the cardhouse structure, whose formation he attributed to the flocculating effects of electrolytes in the water. Using the then more standard X-ray diffraction method, Raitburd (1960) reported complete random orientations of illite and kaolinite particles in a Quaternary marine clay from the Black Sea, which supported some of Rosenqvist's observations.

Mitchell (1956) had examined marine clays using optical microscopy and reported that silt particles appeared to float in a clay matrix. This fabric was confirmed by O'Brien and Harrison (1967) who used the TEM on illitic marine clays. Even where the silt content exceeded 75% Mitchell noted that the clay particles kept the silt grains apart. He also observed the particle orientation in marine clays to be rather patchy, whereas freshwater deposits tended to have well developed orientation aligned parallel or sub-parallel to the bedding. Freshwater deposits with weakly developed orientation were found to have high contents of fine sand or silt, suggesting a dependence of clay orientation on non clay-size particles.

TEM studies by Aylmore and Quirk (1960), Sloane and Kell (1966) and Smart (1967) provided evidence of the domain structure and this was supported by X-ray diffraction measurements (Blackmore and Miller 1961). Again using the TEM Bowles et al (1969) recognised packets (domains?) of parallel orientated particles in slightly consolidated (1 kPa) marine sediments and suggested that the packets existed from the time of deposition.

With the arrival of the SEM clay fabric studies progressed rapidly during the 1970s. Smart (1967a, 1969) paved the way for SEM research and produced a comprehensive account of soil structure. His observations showed that both single particle and domain type structures were possible in freshly sedimented clays. Laboratory sedimented samples of illite and kaolinite were examined in an uncompressed state by O'Brien (1970, 1971, 1972). His SEM studies revealed a fabric consisting of an open network of randomly orientated domains with stepped face-to-face flocculated flakes. He concluded that the gross fabric was similar to the honeycomb or cardhouse models but more face-to-face flocculation was found than had been expected.

Sides and Barden (1971) investigated the microstructure of kaolinite, illite and montmorillonite clays which were consolidated from slurries containing chemical dispersant and flocculating agents. The slurries were consolidated one-dimensionally under an effective stress of about 28 kPa which was thought low enough not to collapse and reorientate any flocculated structure. They found that the structures in the kaolinite samples tended to be turbostratic regardless of any flocculating effects. This confirms the work of Smart (1967) who also did not find any cardhouse or salt-flocculated structures in his kaolinite samples. On the other hand the illitic samples did show the effects of salt flocculation but there was a marked scarcity of the single particle EF contacts associated with a cardhouse structure. The dispersed samples of illite displayed a well orientated structure of closely packed particles. Domains could not be clearly distinguished in their micrographs. Sides and Barden demonstrated that the surface activity of a clay has a great influence on the resulting fabric.

An interesting study by McConnachie (1974) considered the fabric changes in remoulded kaolin during consolidation. The TEM was used since it proved more suitable for domain and pore size measurements. The original clay structure (pressure: 0.1 kPa) had a high void ratio (5.5) with predominant EE and EF contacts between domains. With increasing pressure void size reduced and some domain orientation was effective. At pressures of 100 kPa bonds between constituent particles of domains ruptured and there was a reduction in size and breadth. This is contrary to the findings of other investigators (e.g. Blackmore and Miller 1961) but McConnachie did not consider it significant. The important fact to note from his experiments was that domains were present from the outset which suggests their presence at the depositional interface.

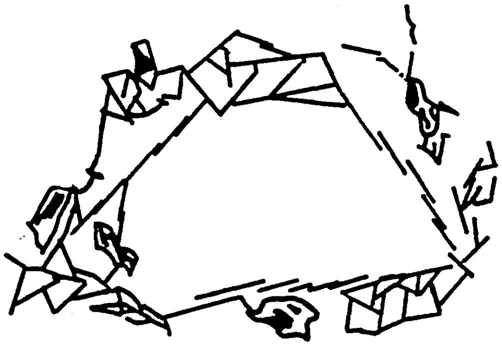
Kirkpatrick and Rennie (1972, 1973) investigated the structure and stress-strain behaviour of laboratory prepared kaolin slurries using isotropic and anisotropic consolidation. Their findings suggested that anisotropic structure develops at an early stage in K_o consolidation and is not altered by a change in stress to isotropic conditions. Their conclusions reflect the importance of the consolidation stress regime in controlling the clay microstructure. A similar study was carried out by Krizek et al (1975) who developed a method of preparing clay samples (Kaolinite) to a predetermined fabric by controlling the chemistry of the clay-water system and the magnitude and type of consolidation stress path. They confirmed that considerable preferred orientation of particles is induced at low K_o consolidation stress values (as low as 200 kPa). The effect of the initial chemistry was to cause higher degrees of orientation in the dispersed samples than the flocculated ones for a given consolidation stress path. It was found that anisotropically consolidated samples produced a preferred orientation whereas a random orientation arose from isotropic consolidation, and this was true regardless of the initial chemistry of the slurries. X-ray diffraction methods were used for the fabric assessment but SEM micrographs did reveal domain formation. In the isotropically consolidated samples domains were composed of face-to-face particle association and their orientation was random (cf turbostratic).

Koff et al (1973) studied Holocene clay shelf deposits from the Barents, Black and Caspian Seas. They observed that all of these marine deposits have a randomly orientated structure at shallow depth. At a certain stage in their consolidation, the orientation of domains and particles acquire an angle of 45^o to the bedding plane, becoming parallel to the bedding at greater depths. Similar sediments were investigated by Osipov and Sokolov (1978). Analysis of the microstructure of these uncompacted deposits revealed a honeycomb arrangement of clay microaggregates having a foliated shape with EF and FF contracts. Lacustrine silts which were also studied had similar microstructures, in spite of having been deposited in a freshwater environment. As a comparison, Osipov and Sokolov looked at artificially prepared non-compacted sediments obtained by precipitation from distilled and salt water. When precipitated in distilled water kaolinite formed a loosely aggregated microstructure similar to the bookhouse structure. The domains were produced exclusively by FF particle associations and had EF and FF contacts. In contrast, when precipitated in the electrolyte solution a characteristic honeycomb structure was formed of particles in a stepped face-to-face association. Illite was found to have a cardhouse microstructure of mostly single particles with EE and EF contacts.

In a series of reports, Bennett and his co-workers (1977, 1979, 1981) looked at the clay fabric of submarine sediments from the Mississippi Delta. At shallow burial depth (1.4 m) the sediments had a high porosity (max 71%) and the fabric was one of randomly arranged domains in EE and EF contact which closely resembled the turbostratic model of Aylmore and Quirk (1960). The domains were composed of FF clay platelets, forming a nearly perfect stack, and also varied in size. The major part of their clay fabric study was carried out using the TEM but some SEM micrographs have been produced (1979 and 1981). One such micrograph of sediment sampled at 0.3 m below the mudline depicts randomly orientated particles with large voids. The structure appears to be similar to the cardhouse type with few domains present. This contrasts with the TEM micrograph at 1.4 m depth and the SEM micrographs at 12.5 and 26.5 m depth which all show well developed domains even though the structure is very open. Bennett et al (1977) presented a tentative clay fabric model for submarine sediments and related it to the void ratio. Based upon their observations and those of Bowles (1968) and

Mathewson et al (1972) the model suggests that at very high void ratios (> 3.0) single plate-like particles are present (rather than domains) which develop long chains of EE and stepped FF contacts, connecting cardhouse type flocs (Fig. 9). At high void ratios (> 2.5) randomly arranged domains in near perfect stack are formed with EE and EF contacts. Beyond this stage lower void ratio sediment (< 1.2) is achieved by FF particle packing with a high degree of preferred orientation.

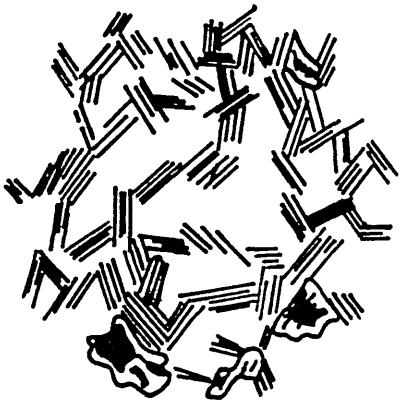
Although few of the clay sediments discussed in this section could be regarded as truly freshly deposited, their fabrics are generally considered as being representative of the fresh state. At shallow burial depths the evidence suggests that the microfabric of undisturbed clay sediments, particularly in the marine environment, consists of an open random arrangement of particles. The details of the particle arrangements are unclear. Rosenqvist (1959) and O'Brien and Harrison (1967) have observed arrangements conforming to the cardhouse structure. Bowles (1968) has concluded that the fabric more closely resembles the honeycomb structure. Substantial reviews by Barden (1972) and Collins and McGown (1974) of natural clay soils indicate that single clay platelet arrangements are rare. Natural soils have structures which vary between flocculated bookhouse and dispersed turbostratic arragements. It would appear that the single particle is a rare commodity in lightly consolidated sediments. Since the previous section indicates that single-plate structures are present in sedimenting clays then there must be a transition to the domain structure found in fresh sediments. From the limited evidence of SEM micrographs (Bennett et al 1981) the



single platelike particles and chains

very high void ratio

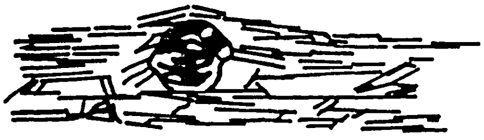
> 3·0



domain particles and chains

high void ratio

> 2·5



very low void ratio

< 1·2

Figure 9 PROPOSED TENTATIVE CLAY FABRIC MODELS FOR SUBMARINE SEDIMENT (Bennett et al 1977)

transition occurs at the depositional interface where there is a sudden change in clay mineral volume concentration. This could account for the reason why there is a considerable loss of water in the first few centimetres of fresh clay sediments (Moore 1932, Züllig 1956). It is unlikely that the increase in volume concentration is the only mechanism operative to cause domain formation. Other controls, such as the electrochemical conditions and benthic biological processes, should not be overlooked.

3.4 The microfabric of clays and mudrocks

The change from a fresh sediment to clay soil and subsequently a mudrock is not as clearly defined as the previous transition from a sedimenting clay. For this study the guidelines of the Working Party Report (Anon 1977) have been adopted, as explained in Chapter 2, giving a range of sediments from soft clays to very strong rocks. The change in microstructure within these limits is partly the result of compaction processes which force out the interstitial water and bring the domains closer together. In doing so, there is an increase in domain orientation and the likelihood of more particles being incorporated into each domain (Moon 1972). Interrelated with these mechanical changes are the normal chemical transformations associated with the diagenetic processes (Dunoyer de Segonzac 1970, Weaver and Beck 1971).

The general effects of compaction on clay sediments have been looked at by many workers. Early notable studies include those of Sorby (1908), Hedberg (1926, 1936) and Athy (1930), although none contains specific information on microstructure. The theoretical relations between compaction and depth of burial were explored by Weller (1959) who considered that the colloidal properties of clay are important with respect to compaction. Excellent reviews by Meade (1964, 1966), Müller (1967) and Rieke and Chilingarian (1974) discuss the many aspects of clay sediment diagenesis with some references to microstructural change. Skempton (1970) presented a valuable study on the gravitational compaction of a variety of clays, relating this to certain geotechnical properties, but did not include details of microstructure.

A major contribution to clay fabric studies has come from the field of geotechnics, which has long recognised the profound effect of particle arrangements upon the engineering properties of clay soils. Terzaghi (1925) and Casagrande (1932) provided the early impetus to this work which was followed up by Hvorslev (1938) and Lambe (1953). Several workers have attempted to correlate geotechnical properties with microstructure (Mitchell 1956, Trollope and Chan 1960, Cheetham 1971, Bennett et al 1977). However, as other factors (e.g. fissures) more often have an overriding influence on the in-situ strength of soils and rocks, the interest in microstructure from the engineer's point of view has diminished. This is not the case with some sensitive clays, though.

Laboratory experiments on clay-water systems have established that with increasing pressure, the effect on sediment microstructure is to force the constituent particles or domains closer together into a preferred orientation, which is normal to the compressive stress (Mitchell 1956, Von Englehardt and Gaida 1963, Sloane and Kell 1966, Smart 1967). It is logical that the amount of water present will influence the ability of particles to reorientate. Martin (1966) has shown in consolidation experiments with kaolinite that high initial water contents produced an increase in the degree of preferred orientation at low stress levels (150 kPa). Meade (1966) reviewed the work of many authors on the role of compaction in producing preferred orientation and concluded that most of the orientation develops very early under the first few metres of overburden.

McConnachie (1974) found that the preferred orientation of kaolinite domains increased with pressure up to 10 kPa and thereafter remained essentially constant. This low value agrees closely with that of Morgenstern and Tchalenko (1967), who found no improvement in orientation above a pressure of 10.8 kPa. As consolidation proceeds, McConnachie observed that domains were able to move freely and become more closely packed. Rotation rather than rupture at points of contact accounted for the increase in preferred orientation. It was found that EF contacts decreased in number at the expense of FF contacts. At pressures of 10^5 kPa inter-domain contacts were mostly EE and EF, although the degree of preferred orientation was still less than perfect. In contrast to other workers (Blackmore and Miller 1961) McConnachie did not find that domains increased in size as consolidation progressed.

In a comparable study Bowles et al (1969) consolidated natural sediment samples, obtained from cores taken in the Gulf of Mexico, over a stress range of about 50 to 6300 kPa. TEM micrographs showed a progressive change in particle orientation during the laboratory consolidation tests. A distinct alteration in microstructure was found to occur between 50 and 400 kPa. At the upper stress level there was a dense packing of particles as opposed to the previous open arrangement. A significant feature was that in spite of the large decrease in void ratio (2.2 to 1.3) there was no noticeable development of preferred orientation. Micrographs of samples subjected to the maximum stresses (3100 and 6300 kPa) showed a strong development of preferred particle orientation, although localised areas of random orientation existed. On the basis of their limited observations Bowles et al concluded that random orientation is likely to be retained over the greater part of the reduction in void ratio.

The microstructure of a variety of clays has been studied by Barden (1972) in an attempt to relate soil structure to engineering behaviour. He concluded that the most important single parameter governing microstructure was the electro-chemical environment during deposition. Lightly overconsolidated marine clays were found to have a flocculated open structure which was modified by the nature of the silt content. Brackish and freshwater clays, which again were only lightly overconsolidated, displayed both flocculated and dispersed structures. Heavily overconsolidated clays, from marine environments, had a turbostratic arrangement with a degree of horizontal orientation which was thought to be derived from the collapse of a flocculated structure. Tchalenko (1968) reported that heavily overconsolidated London clay exhibited a strong preferred particle orientation except for zones of severe weathering.

Bennett et al (1977) noted that the void geometry of deeply buried clays is quite different from that of the overlying high-porosity sediment. The deeply buried low-porosity clays were characterised by a strong preferred particle orientation of domains and continuous chains with associated thin voids, although significant numbers of domains did deviate from the preferred direction. Some of these obliquely orientated domains were in association with larger voids, which could possibly be holes left by dislodged silt particles. Sediments with high silt and fine sand contents do have less tendency for parallel orientation (Mitchell 1956).

The overwhelming evidence is that compaction produces an increase in the parallelism of particles. The most efficient way of producing a low-porosity sediment is by FF particle packing which naturally results in a high degree of preferred orientation. The domain structure has been repeatedly observed in compacted clay but the nature of domain interaction and orientation is very complex. Collins and McGown (1974) encountered different particle arrangements existing side by side in the same soil. Although in general, they found that bookhouse and stepped flocculated arrangements were more common in marine soils, whilst brackish water soils tended to have dispersed or turbostratic arrangements.

Fabric studies of mudrocks have usually addressed themselves to the question of fissility in shales. Comparatively few studies have looked at mudstones, particularly using the SEM. One of the first authors to investigate the occurrence of fissility in mudrocks in some detail was Ingram (1953). He associated fissility with a parallel arrangement of clay particles which was increased by the presence of organic matter. In his study Ingram identified three distinct breaking characteristics: massive, flaky-fissile and flaggy fissile. The difference between a flaky or flaggy shale was attributed to the nature of the cementing agent. Ingram used a light microscope for his observations and utilised the optical properties of clays, such as birefringence, to determine particle orientation. White (1961) confirmed that fissility in shales is dependent on preferentially orientated clay particles and suggested that its origin lies within the chemical environment at the time of sedimentation. Odom (1967) also concluded that a high degree of preferred orientation of clay particles is evident in shales, which gives rise to the fissile structure. The work of White and Odom was undertaken using X-ray diffraction techniques.

Early electron microscope studies of shale fabric with the TEM were carried out by Grim et al (1957), Gipson (1965) and O'Brien (1968). In an attempt to substantiate his findings Gipson applied statistical analysis methods to the data obtained. SEM investigations of shale fabric have been carried out by Gillott (1969), O'Brien (1970) and Barrows (1980). Gillott noted that the preferred orientation observed at low magnification (approx 300 x) was not so clearly distinct at higher magnifications (approx 3,600 x), with some particles even arranged at large angles to the fissile plane. The influence of magnification power on 'observed' particle orientation is important in fabric studies since the degree of preferred orientation could vary for the same sample at different magnifications.

O'Brien (1970) concluded that fissility arises from the deposition of clay in a dispersed state or from the collapse of clay floccules under the weight of overburden. The former view is endorsed by Moon (1972) and Moon and Hurst (1984). O'Brien noted that fissility was best developed in illitic black shales in which nearly perfect parallelism of clay flakes occurred. He suggested that the presence of adsorbed organic molecules promoted sedimentation in a dispersed state by their ability to neutralise the surface charges on clay particles. Non-fissile mudrocks were found to have a random orientation of clay flakes.

Not all workers agree with the suggestion that particle orientation controls fissility. Spears (1976) investigated weathered and unweathered mudrocks of the Upper Carboniferous and concluded that their fissility is related to laminations. This was supported by Curtis et al (1980) who quantified mudrock fabric using X-ray goniometry and found that fissility is not simply related to clay orientation, but rather mineral segregation into fine laminae is more important. Byers (1974) and Barrows (1980) both stress the role of bioturbation in disrupting lamination and particle orientation, a fact which is also noted by Blatt et al (1980) in their section on fissility (p 398-400). They concluded that due to ingestion, and hence disturbance to fabric, by bottom dwelling animals then fissility must be a property produced during diagenesis rather than at the time of deposition.

There is sufficient evidence to substantiate the important role played by benthic organisms in altering sediment fabric. Cluff (1980) showed that the variation in bulk fabric of the New Albany Shale Group was governed primarily by burrowing benthic organisms. Rhoads and Boyer (1982) presented a review of the effects of marine benthos on the physical properties of sediments. In a recent paper by O'Brien (1987) the effects of bioturbation on shale fabric were discussed. He found that when viewed on the SEM, bioturbated fabric was seen to be very similar to the fabric of mudstones formed from flocculated clay. The only distinguishing feature was the lack of stepped domains or stepped card-house structure in the bioturbated clay. Organic rich shales formed under anoxic conditions were unbioturbated and their fine laminations and fabric of parallel clay flakes were preserved. That a link exists between preservation of fabric and bioturbation is unquestionable, but how important that link is in the formation of shales remains to be proven. In the absence of total reworking by bioturbation, whether this is a consequence of anoxic conditions or not, the clay particles of a sediment take on a structure which may be classified as random or preferred. This primary fabric develops at, or soon after, the time of deposition in response to the environmental conditions present. Bioturbation is a secondary event which unfortunately clouds the shale formation issue.

CHAPTER 4

DEVELOPMENT OF MICROSTRUCTURE

Soils and most sedimentary rocks are particulate in nature consisting of an assortment of mineral grains. The manner in which these grains are spatially arranged determines the overall microstructure. In clastic sedimentary rocks the microstructuve owes its development to both the genetic origin of the rock and also the composition of its constituent minerals. The environment within which the particles are deposited is the first stage to a number of processes that will help to determine microstructure. The important aspects of the environment are the physical, chemical and biological conditions. These three mutually dependent parameters are reviewed. The response of the particles to these conditions will depend upon the type of particle forming the sediment. In the environments considered in this study the most influential particles are the clay minerals. How the clay minerals behave and their role in the development of mudrock microstructure is explained in the model presented here.

4.1. Environments of deposition

There is a recognised relationship between the environment of deposition and the nature of the sediment deposited. The area within which deposition takes place is defined by the physical, chemical and biological processes operating there. Variations in the speed and intensity of these processes usually have sufficient effect so as to produce a characteristic deposit. An important consideration is the duration of time in which sediment particles are transported and remain in the depositional environment since this governs the period during which important physico-chemical processes are allowed to act.

There are three major groups of mudrocks (Tucker 1981) but only those formed through the normal sedimentary processes of erosion, transportation and deposition will be considered. These are known as detrital mudrocks. The majority of the particles which form the mudrock are generated at the earth's surface by the erosion of landmasses. These clastic particles are usually transported in hydraulic suspension from their source to the depositional area which is typically a low energy environment. This allows the fine-grained particles to settle out of suspension. The environment may be classed as either marine or non-marine but other categories such as brackish and hypersaline also exist.

4.1.1 Deposition in marine environments

Marine deposition is by far the most common origin of detrital mudrocks. The three distinct sites of deposition for marine mudrocks are:

1. shelves and shallow marine platforms

2. continental slopes

3. abyssal plains and deep basins.

Mud accumulation on the inner shelf area can do so at quite high rates of progradation, as much as 10 m per year (le Fournier and

Friedman 1974). Typical of these inner shelf deposits are the Amazon muds that are forming along the Surinam Coast (Wells and Colemen 1981; Rine and Ginsburg 1985). These mudbanks are characteristically massive and homogeneous with a general absence of burrow traces and faecal pellets. Parallel and sub-parallel laminations are apparent. The lack of benthic evidence suggests that the massive nature of these beds, which are up to 2.2 m thick, have a depositional rather than bioturbational origin. Mud belts are also found in mid shelf and shelf-edge areas and will shift position in response to sediment supply and dispersal. Mid shelf and outer shelf mudbelts have been described by Baker (1976) and Gorsline and Grant (1972) respectively. In all cases the balance between sediment supply and the capacity of shelf processes to disperse the detritus is critical to the accumulation of mud on the shelf itself.

Sediments removed from the shelf area are typically deposited on the adjacent slopes to form muds of turbidite origin. These will be interbedded with hemipelagic clastic deposits which have settled through the water column from suspended sediment advected off the shelf. Biological pelletization will assist in encouraging the settlement of the suspended sediment (McCave 1975). A typical turbidite deposit will contain predominantly silt and clay size particles in a fining-up sequence to form graded bedding characteristic of a Bouma cycle (Bouma 1962). Incomplete sequences, where base truncation has occurred can lead to thick-bedded mud turbidites if the depositional process is repeated. In this way layers several metres thick can be deposited. Turbidity currents are thought to be responsible for much of the muds which were originally deposited in ancient geosynclines and now form the mudrocks of contemporary landmasses (Potter et al 1981).

Not all the sediment carried by turbidity currents is deposited on the slopes. Turbidity currents that are confined by the walls of submarine canyons will deposit their sediment in submarine fans spreading out onto the abyssal plain. These fans are regions of major sediment accumulation both presently and in the past (Blatt et al 1980). A model for submarine fan growth has been presented by Normark (1970).

The floors of abyssal plains and marginal basins represent the ultimate sediment sink in the oceans. The sediment deposited at depth consists of pelagic and hemi-pelagic muds which have settled slowly through the water column and accumulated with the minimum of disturbance from bottom currents. They may be interbedded with turbidite muds. True pelagic sediments consists of the skeletal parts of marine plankton intermixed with very fine terrigenous silt and clay that has reached the open ocean by aeolian transport. Where the biogenic component is reduced, perhaps by dissolution, the resulting sediment is known as pelagic or red clay. The sedimentation rate of pelagic clays are commonly less than 1-10 mm per 1000 years (Menard 1964). Even though the clays are deposited in the deepest and most remote parts of the ocean basins they are often thoroughly bioturbated, indicative of well-oxygenated bottom waters.

Hemi-pelagic sediments are typical of marginal oceanic settings not far removed from terrigenous sediment sources. They are commonly homogeneous, with an absence of primary structures due to extensive bioturbation. Descriptions of modern hemipelagites have been given by Rupke (1975).

4.1.2 Deposition in non-marine environments

The two principal environments of deposition for non-marine mudrocks are:

1. lacustrine
2. fluvial.

Mudrocks originating from lakes vary considerably depending on the chemistry of the water, biological productivity and climate. Compared with oceans, lakes have a short geological life and become completely filled with sediment in a few million years. As a lake becomes shallower biological productivity increases and organic matter becomes more abundant. The accumulation of the organic matter can lead to the formation of oil shales. The chemistry of lake waters indicate a carbonate rich environment rather than surphate or chloride rich, especially in humid climates. The absence of saline waters reduces the potential flocculation of clay particles and these remain in suspension to be deposited in the quiet central parts of the lake.

A seasonal variation in water temperature causes alternating periods of stratification and mixing which affects the sedimentation process. These seasonal fluctuations hinder the development of a biological diversity and increase the chance of sediment lamination.

In deep lakes a lack of significant water circulation can lead to anoxic conditions and help to preserve the abundant organic matter (Swain 1970). Such environments are characteristic 'black-shale' sources.

Fluvial mudrocks are not so widespread because of the infrequent opportunity of clay particles to be released from suspension. The main sources are overbank muds that occur on floodplains and more importantly deltaic sediments. Deltas have been singled out as fluvial but in fact the environment overlaps into the lacustrine and marine situation. Much work has been written about marine deltaic sediments primarily because they have significant economic importance with respect to petroleum and coal resources. The main location for mudrock forming sediments is in the prodelta area where salt water flocculation promotes the deposition of clay particles (Hyne et al 1979). Seaward migration of a delta buries the prodelta clays and a coarsening-upward cycle of sediments is produced. A typical cycle begins with a fissile dark shale which becomes interbedded with graded siltstone turbidites towards its top, gradually passing upwards into sandstones (de Raaf et al 1965).

4.2. Geochemistry of environments

In general sediments are deposited under either oxidizing or reducing conditions. These environments of deposition are often referred to as aerobic (or oxic) and anaerobic respectively. The terminology describing levels of oxygenation is not standard (Tyson 1987). For the purpose of this study an aerobic environment is taken as one in which there is sufficient dissolved oxygen to cause an almost total absence of organic matter in the sediment. The anaerobic environment, by virtue of its extremely low oxygen content, permits the preservation of organic matter and the resulting accumulation of plant and animal debris causes reducing conditions to form. Rhoads and Morse (1971) quantify the anaerobic environment as having an upper limit of 0.1 ml/litre of dissolved oxygen. The term 'anoxic' is restricted to an environment totally devoid of oxygen and containing free hydrogen sulphide. Bottom feeding animals cannot survive in these anoxic conditions; it is the realm of the sulphate reducing bacteria like Desulfuvibrio.

4.2.1 Chemical factors in sedimentation (pH, Eh and electrolyte concentration)

Chemical reactions accompanying sedimentary processes are primarily dependent on the hydrogen-ion concentration (pH) oxidation-reduction potential (Eh) and electrolyte (salinity) concentration of the natural waters.

The pH of the environment is particularly significant in controlling the precipitation of minerals from solution. Observed pH values in most natural environments lie between 4 and 9 (Krauskopf 1967), with marine waters normally showing a range of 8.1 to 8.3. The neutral pH value is 7 which means that marine waters are slightly alkaline. The lowest recorded pHs in nature are found in solutions in contact with oxidizing pyrite, indicating a strongly acidic environment. The importance of pH is that it effects the surface charge of clay minerals as noted in section 4.4.2.

The oxidizing capability of an environment is given by its Eh value, which is a measure of the energy change involved in adding and removing electrons from an element. The oxidation potential varies with changes in concentration of the reacting elements and is also influenced by the pH value. The range of oxidation potentials of the environment will determine the reactions that may take place. In the sedimentary environment Eh is controlled by the balance of dissolved oxygen and the amount of organic matter to be oxidized. Iron minerals are particularly sensitive to oxidation-reduction potentials and the type of mineral present in the sediment is indicative of the former depositional state. Generally speaking, oxidizing environments promote haematite and reducing environments promote pyrite.

Neither Eh nor pH is an independent variable; both are determined by reactions occurring in the environment. The resulting combination of Eh and pH provides the control that determines the nature of the sedimentary products. A careful study of the mineralogy of a sediment will thus elucidate the physicochemical conditions under which it developed. A classification of sediments in terms of Eh and pH has been given by Krumbein and Garrels (1952). The approximate position of some natural environments as characterized by Eh and pH values is discussed by Garrels and Christ (1965).

Clay particles are surrounded by a diffuse electric double-layer as defined by the Gouy-Chapman theory (Chapman 1913). When two particles approach each other their double-layers interact to cause a repulsion. The effect of an increase in electrolyte concentration is to compress the double layer and the distance over which repulsion acts is considerably reduced. This allows for attractive

Van der Waals forces to take effect and a flocculation of particles arises. The electrolyte concentration in water increases with increasing salinity and as a consequence clay particles tend to flocculate readily in marine environments.

Adsorption of organic molecules by clay particles is affected by the salinity and pH of the water. Rashid et al (1972) found that adsorption of humic acid increases with increased salinity and decreased pH but also that the cation exchange capacity of the clay particle played its part. It is thought that adsorption can increase the resistance of some organic materials to bacterial decomposition.

4.2.2 Chemical reactions of aerobic and anaerobic environments

In the aerobic environment a plentiful supply of dissolved oxygen allows aerobic microbes to decompose any organic matter in suspension prior to its burial. With the removal of organic matter from the sediment ferric oxide minerals are not reduced and after burial and diagenesis are able to transform to haematite. This is one cause of the predominant red colouration in sediments formed under oxic conditions at the time of deposition.

In situations where the replenishment of oxygen cannot match the demand then the consumption of the oxygen by the aerobic microbes leads to an oxygen deficiency and eventually anaerobic conditions. Further bacterial reduction of the organic matter by other microbial communities takes place and in a chain of events hydrogen suphide is produced which renders the anaerobic environment anoxic. The sequence of oxidation is controlled by the physiology of the particular organisms (McCarty 1972).

Not all anaerobic environments become sulphidic. It is possible that in low concentrations organic matter may use up all the dissolved oxygen but not be in sufficient quantities to enable sulphate reduction to occur. This leads to a weak reducing environment, referred to as post-oxic by Berner (1981). Alternatively, if enough organic matter is deposited to promote suphate reduction and reducible iron is also present, then the hydrogen sulphide produced will subsequently be precipitated to form iron sulphides and pyrite (Goldhaber and Kaplan 1974, Fisher 1986). After cessation of sulphate reduction further decomposition of the organic matter results in the formation of dissolved methane to give a strongly reducing environment. Methane production is accomplished with greater ease in fresh rather than marine waters since the initial sulphate content is lower.

4.3 Accumulation of organic matter in sediments

The principal source of organic matter in sediments is the autochthonous population of surface waters, particularly phytoplankton (Bordovskiy 1965). A secondary source is transported terrestrial organic matter from rivers, streams and offshore winds. In spite of the vast quantity of organic matter available in the oceans only a small quantity ever reaches the sediment floor. Trask (1932) estimated that only 2% reached the sea floor in shallow waters and 0.02% in the open sea. However even these small amounts may strongly influence the post-depositional behaviour of a sediment.

The accumulation of organic matter is controlled by both biologic activity and physical factors. Biological activity relates to the primary productivity of the organic matter as well as its biochemical degradation by metazoan and microbial scavangers. Physical factors include mode of transport prior to deposition, particle size and sedimentation rates. The principal setting for the accumulation of organic matter is regarded as the anoxic environment (Demaison and Moore 1980) in which various chemical and physical factors interplay to produce oxygen deficient bottom waters conducive to organic matter preservation.

4.3.1 Biological factors

Primary productivity determines how much organic matter is available for deposition. The biochemical process which controls organic matter production is called photosynthesis. This process is responsible for the reducing ability of organic compounds (Krauskopf 1967). The importance of productivity in controlling the organic matter accumulation in a sediment has been questioned by Demaison and Moore (1980) and Tyson (1987). They both concur that there is no consistent general correlation between the organic carbon content of bottom sediments and the productivity of the overlying waters. However high productivity rates have been quoted as directly responsible for the formation of some organic-rich deposits (Morris 1987).

Bacteria play a critical role in decomposing dead plantonic organisms and they perform their task most efficiently in aerobic waters. In present day marine environments the rapid transport of organic matter to the sea floor by biological mechanisms means that the residence time within the water column is short. The organic matter arrives at the sediment-water interface and within the upper few centimetres of the sediment column it is significantly altered by the intense activity of bacteria and benthic organisms. Due to eventual exhaustion of the oxygen supply by this activity, bottom muds commonly become anaerobic a few centimetres below the sediment-water interface, even under an oxygen-rich water column. At this level anaerobic bacteria take over from aerobic.

Before the advent of biological interference the settling velocities of organic matter and fine particles must have been much slower, allowing time for intense particle-seawater interactions. Oxidation of organic matter in the water column would create anaerobic conditions more readily. Slight changes in the organic matter budget would cause fluctuations between anaerobic and oxic conditions as noted by Degens and Stoffers (1976).

4.3.2 Physical factors

Sediment accumulation rates have a noticeable effect on the organic carbon content of marine sediments (Müller and Suess 1979). The relationship is attributed to the residence time of organic matter in the region of the sediment-water interface. As already stated, it is at this zone that bacterial degradation is most rapid. Fast burial rates should promote organic matter preservation (Calvert 1987) but at the same time a dilution effect by clastic material is experienced (Ibach 1982).

Under oxic water conditions fluctuations in organic carbon content are related to sedimentation rate until dilution effects become significant. Surface productivity rate has little influence. It would appear that any increase in organic matter production is matched by the voracity of the organisms in the water column. With anaerobic to anoxic water conditions the influence of sedimentation rate is unclear and both this and surface productivity are thought to contribute to organic carbon preservation (Demaison and Moore 1980).

4.3.3 Anoxic environments

The general concensus is that organic matter is preferentially preserved in marine sediments which accumulate under anaerobic to anoxic conditions. A classification of anoxic environments has been reported by Demaison and Moore (1980). It should be noted that their upper bound for anoxia is 0.5 ml/litre of dissolved oxygen which they define as the threshold of arrested bioturbation. Thus it includes true anoxic conditions (sensu Rhoads and Morse, 1971) and part anaerobic conditions. In this section the word anoxic is used to indicate both anaerobic and true anoxic conditions.

The main causes of anoxia in bodies of water are excessive oxygen demand and deficient oxygen supply. This leads to a reduced oxygen concentration in the bottom waters. The major environments where reduced oxygen concentration is likely to occur and where organic rich sediments can accumulate are illustrated in Fig. 10.

Excessive oxygen demand is reported in regions of coastal upwelling (Calvert 1987) where plentiful organic matter lowers the oxygen content of the water to create an oxygen minimum zone. If the zone contains an anoxic core which impinges upon the continental slope or

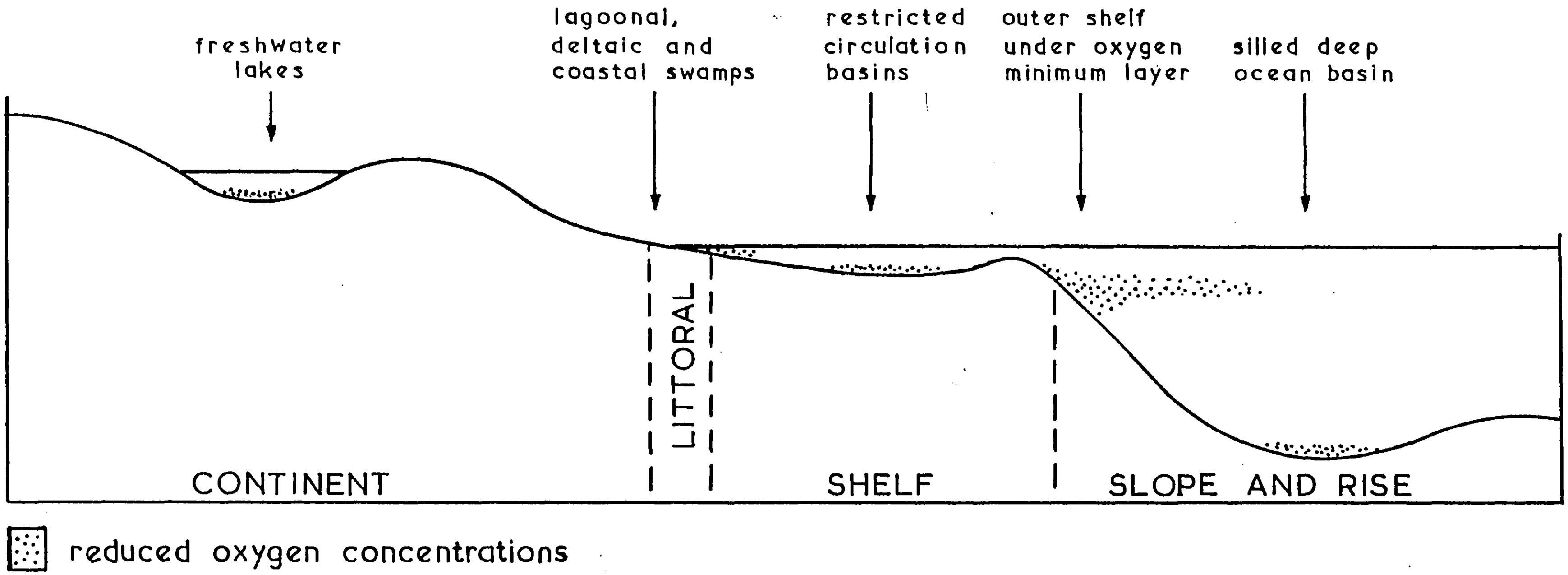


Figure 10 MAJOR ENVIRONMENTS OF ORGANIC-RICH SEDIMENT ACCUMULATION (Brooks et al 1987)

shelf then organic rich sediment can accumulate there. Such an environment is called an anoxic open ocean and has been suggested as the model for some intermediate-depth organic-rich black shales (Arthur et al 1984).

Oxygen depletion occurs in silled basins and some large inland lakes due to the demand exceeding supply. Anoxic conditions occur when the bottom water becomes stagnant because a permanent density stratification prevents good circulation. The most quoted example of a silled basin model is the Black Sea (Caspers 1957, Degens and Ross 1974). The presence of a sill alone is not sufficient to create anoxic bottom conditions; it is dependent on patterns of water circulation. Silled basins with a 'negative water balance' (e.g. the Red Sea) remain oxic because a large inflow of water from the oxygenated ocean is needed to replenish the evaporating waters. An example of a large anoxic lake is Lake Tanganyika (Degens et al 1971). The main factor producing anoxic lakes is a warm climate with little seasonal change. This promotes a permanent water column stratification. The seasonal overturning of water evident in temperate lakes prevents anoxic conditions from forming.

4.4 The influence of clay minerals in microstructure development

The microstructure of a sediment will be governed by the relative positions of its constituent particles. In the case of fine-grained sediments these are predominantly the clay minerals, although other minerals are present in various percentages (Shaw and Weaver, 1965). In order to understand why different microstructures are generated from the same constituent particles it is appropriate to consider the influence that clay minerals have on microstructure development.

The physical and chemical behaviour of clay minerals is largely determined by surface electrical charges which arise from ionic imbalances in the mineral's molecular structure. The forces associated with these charges act between all adjacent clay particles and because the mass of each particle is very small the attraction and repulsion of the charges become significant. When the electrical forces dominate the mass forces then the particles behave according to the rules of colloid activity.

The structure, properties and colloid activity of clay minerals are all interrelated and together they contribute to the unique role that these minerals play in determining mudrock microstructure.

4.4.1 The structure of clay minerals

The first studies of clay mineral structure required the application of X-ray diffraction techniques (Ross 1927, Pauling 1930). This approach revealed that clay minerals consist of a stack of parallel unit layers where each unit layer is a combination of two dimensional arrays of silicon-oxygen tetrahedra and aluminium - or magnesium-oxygen-hydroxyl octahedra (Fig 11a, b). The variety of clay minerals arise from differences in the number of unit layers, how these are stacked, the substitution of other elements in the units and the nature of any exchangeable cations present in the interlayer zone.

The three principal groups of clay mineral are montmorillonites, illites and kaolinites, taken in order of ascending size. The latter two have been chosen for the sedimentation experiments in

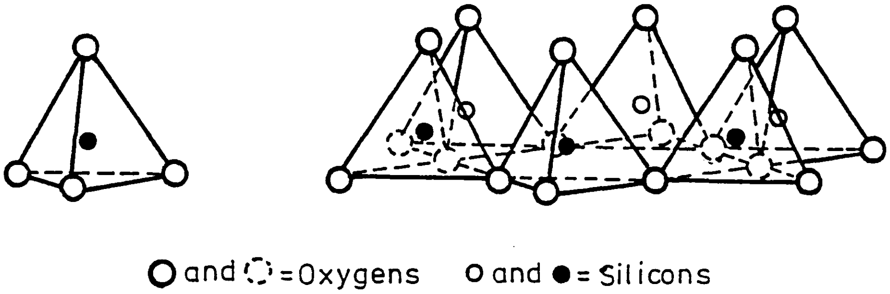


Figure 11a DIAGRAMMATIC SKETCH SHOWING A SINGLE SILICA TETRAHEDRON AND THE SHEET STRUCTURE ARRANGED IN A HEXAGONAL NETWORK (Grim 1968)

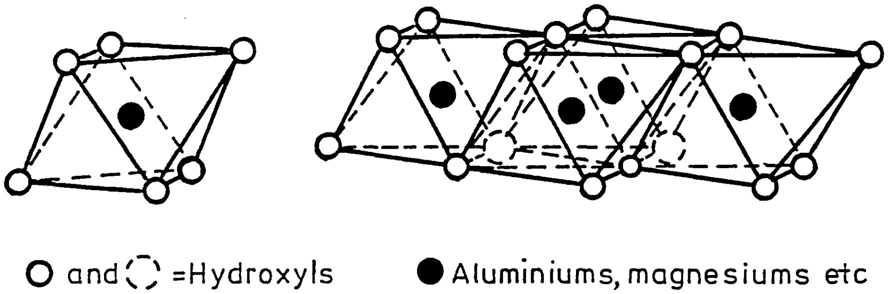


Figure 11b DIAGRAMMATIC SKETCH SHOWING A SINGLE SILICA OCTAHEDRAL UNIT AND THE SHEET STRUCTURE ARRANGED IN A OCTAHEDRAL NETWORK (Grim 1968)

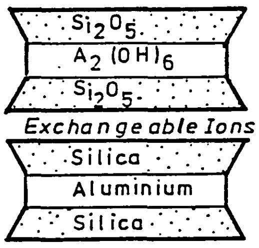


Figure 12 ILLITE

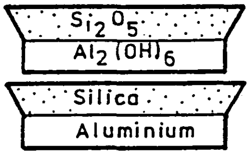


Figure 13 KAOLINITE

Simplified sketches depicting arrangements of unit layers (Potter et al 1980) this study. The illite group is the most abundant of the clay minerals. It is a three layer or 2:1 type clay mineral and is classed as non expanding since it does not swell with the addition of water. Illite is built up of unit layers composed of tetrahedral sheets with one octahedral sheet in between (Fig 12). Substitution of atoms in the unit layers results in a charge deficiency which in the case of illite is compensated by potassium cations (K^+) in the interlayers. Lenses of water may also be present in the interlayer sites. The kaolinite group is composed of clay minerals which have an almost perfect two layer or 1:1 type lattice. They consist of unit layers of one tetrahedral and one octahedral sheet (Fig 13). Polarisation of the Si-O and O-H groups causes the surface oxygen and proton planes to become negatively and positively charged respectively. These charges assist the stacking ability of unit layers one above the other by electrostatic-type attraction forces (Giese, 1973) in addition to van der Waals bonding (Cruz et al 1972).

Within unit layers the atoms are held together by primary valence bonding but the unit layers themselves are linked only by secondary forces. These weaker bonds allow comparatively easy splitting in a plane parallel with the unit layers. In the case of kaolinite large particles are able to form due to strong hydrogen bonding between the oxygens and hydroxyls in adjacent units.

4.4.2 The properties of clay minerals

The various properties of clay minerals induce a behaviour which makes them unique among clastic sedimentary particles. This behaviour arises from their small size, platy crystal habit, large surface area per unit weight, cation exchange capacity and an affinity for water.

The size of clay particles has an upper limit of 2 μm equivalent spherical diameter (e.s.d.) which is regarded as the diameter of a sphere with an identical settling velocity. The size of clay minerals varies, as does their shape. Kaolinite crystals are typically pseudo-hexagonal plates, 0.3 - 3 μm in diameter and about a tenth of that in thickness. The inequant shape of clay minerals may influence the manner in which they are sedimented, although the final settling pattern is complex and other factors come into play.

The surface area per unit weight (specific surface) of clay minerals is very high ranging from 10 m^2/g for kaolinite to 800 m^2/g for montmorillonite. Particles whose specific surface is of this magnitude are able to form colloids if mixed with a liquid medium. In such a colloid system gravitational forces become less of an influence on the behaviour of the particles than the forces associated with electrical charges on the mineral surfaces.

Surface charges on clay mineral faces are invariably negative and may typically result from either an isomorphous substitution of certain cations in the crystal lattice by cations of a lower valency or, alternatively, from broken bonds in the silica-alumina units. In the former case the negative charge is 'permanent' and does not depend on the pH of the environment, whereas the charge derived from broken bonds is variable and increases with increasing pH. In both cases the presence of the negative charge allows soluble cations to be attracted or adsorbed onto the mineral surface without alteration to the basic structure. These cations may also be replaced by other cations if the ionic environment alters. The capacity for cation exchange (c.e.c.) varies widely in clay minerals depending upon whether the attractive forces arise mainly from broken bonds (low) or from lattice substitution (high). Ferris and Jepson (1975) found that lattice substitution is rare in kaolinites which accounts for the small exchange capacities of these minerals.

At the edges of clay minerals broken bonds arising from disruption of the silica and alumina sheets usually contribute a positive charge to the edge surface area. The charge varies becoming more positive with decreasing pH, but its sign may be reversed with increasing pH. Interaction between face and edge charges gives rise to a variety of particle arrangements (see Fig 4.).

The affinity of a clay mineral for water is directly related to its cation exchange capacity. The high c.e.c. value of montmorillonite (80-150 me/100 g) is reflected in its ability to adsorb vast quantities of water molecules within the interlayer sites, as well as on the mineral outer surfaces. Kaolinite (c.e.c. of 3-15 me/100 g) only adsorbs small amounts of water molecules.

The aqueous phase adjacent to clay particles is divided into three regions of distinct dielectric behaviour (Bell and Levine 1972). The region closest to the particle surface consists of a layer of preferentially orientated water molecules; it is called the inner Helmholtz layer. Beyond this is the outer Helmholtz and the Gouy-Chapman diffuse layers.

4.4.3 Clay Colloid activity

Colloid systems are defined as systems containing at least two mutually dependent components, these being a continuous dispersing medium and a disperse phase (Yariv and Cross 1979). In sedimentation terms the dispersing medium is a liquid and the disperse phase is a solid. Colloids may either be stable (lyophilic) or metastable (lyophobic). Where water is the dispersing medium these colloids are called hydrophilic and hydrophobic, respectively.

The characteristic behaviour of clay-sized particles when dispersed in water results from the surface activity at the solid-liquid boundary. Molecules and ions at the interface have different properties and energy characteristics from those in the internal structure. At the interface a molecule will interact simultaneously with adjacent identical molecules and with molecules that form the other phase of the colloid system. These interactions are very rapid and provide the ability of an almost instantaneous response to any change in the balance of a colloid system.

The dispersion of clay particles in water is classed as a hydrophobic colloid whose properties depend largely on the extensive interface between particles and liquid. Hydrophobic colloids are characterised by having excess free energy which is dependent on the high interfacial area. Any increase in the size of particles leads to a decrease in the total surface area and the excess free energy decreases. The hydrophobic colloid will display a variety of properties which are affected by changes in either the solid or liquid phase, but the overall reaction of the system is governed primarily by the large area of particle-water interface.

Clay particles in suspension are affected by attractive and repulsive forces which are dependent upon the nature of the system. Interparticle attraction and repulsion operate simultaneously, but the repulsive force is more responsive to changes in the system characteristics. The repulsive force is reduced by, amongst other things, increasing the electrolyte concentration or reducing the pH of the system. Thus an increase in salt concentration will lead to a nett attraction between clay particles. Within colloidal systems the particles are in constant collision due to the effects of Brownian motion. Hence, collisions of clay particles in saltwater solutions result in particle adhesion or flocculation. If the nett force is sufficiently repulsive then the clay particles remain dispersed.

One means by which dispersion can be achieved is by neutralising the attractive forces on the clay particles. The addition of small amounts of certain chemicals can have a dramatic effect on the stability of a colloid. Van Olphen (1963) reports that the most efficient method of deflocculating (peptizing) clay suspensions is to change the positive edge charge into a negative one. Phosphates, oxalates and other salts disperse clay flocs in dilute suspensions, the anions of these salts reacting with the particle edge cations and becoming adsorbed onto the edge surfaces. Since the edge surface area is small only relatively small amounts of deflocculating chemicals are needed.

Similar mechanisms are evident in the dispersive ability of organic anions such as tannates. The tannate anions are adsorbed onto the edge surfaces of the clay particles, reversing the edge charge, and creating a negative double layer which prevents EF and EE associations.

In contrast to this mechanism, organic cations, such as amino acids, are readily adsorbed onto the negative face surfaces, giving the clay particle an overall positive charge. Many polar organic compounds behave in a similar manner.

4.4.4 Interaction between clay minerals and organic compounds

Clay minerals have an ability to adsorb polar organic compounds and ions because their mineral surfaces contain both electron donor sites and electron acceptor sites. The unit layers are also electrically charged. The organic compounds become adsorbed within the interlayer spaces of the minerals, on the broken bond surface and on the outer planes.

Yariv (1976) has presented a model in which the charged head of a hydrocarbon chain is able to displace water molecules surrounding the clay surface. The non-charged end is forced away from clay surface due to repulsion forces in the diffuse double-layer. It is therefore possible for a clay particle to become surrounded by organic ions which effectively neutralise the surface. The repulsive forces that occur between two similar double layers of adjacent particles are decreased and the saturated organo-clay complexes are able to adopt a face to face stacking.

The adsorption of organic polar molecules depends on the nature of the exchangeable inorganic cation present in the interlayer sites and on the hydration state of the clay. It would appear that when the clay surface has a low charge density hydrated cations may easily displace water in the inner Helmholtz layer and hence 'coat' the clay surface (Yariv and Cross 1979). Surface charges arising from broken bonds are least under low pH conditions (see Section 4.4.2).

In Chapter 1 attention was drawn to the potential influence that organic compounds may have in causing fissility in mudrocks. The important compounds suggested were amino acids, fatty acids and sugars. Theng (1974) has reviewed the chemistry of clay-organic reactions and describes the association of clay minerals with these compounds.

The adsorption of amino acids by clay systems is said to be extremely sensitive to pH variations. In order to substitute all inorganic cations for amino acids a low pH is required. The combination of amino acids, clay minerals and a low pH comes together in reducing environments.

Fatty acids and clay minerals have been extensively studied because of their association with forming petroleum hydrocarbons (Jurg and Eisma 1964; Shimoyama and Johns 1971). The adsorption of fatty acids is principally due to their interaction with broken-bond surfaces and hence is pH dependent. Meyers and Quinn (1971) found that the amount of fatty acid removed from saline solution was related to the organic concentration.

Bloomfield (1956) has demonstrated the dispersing properties of aqueous leaf extracts and identified the active constituents as a sugar polymer (polysaccharides). Free sugars have been recorded in reducing environments (Degens 1965). The adsorption of sugars by montmorillonite has been reported by Greenland (1956) who noted that a maximum uptake occurred in the range of pH 4 to 5.

From the evidence above it would appear that in reducing environments, where the pH is low, clay minerals have an increased chance of adsorbing those organic compounds suggested as being responsible for dispersing the clay particles. These same compounds also have a role to play as precursors to petroleum hydrocarbons.

4.5 Fissility in mudrocks - a suggested model for its development

It has been noted in Chapter 1 that several authors have observed a parallel fabric in fissile mudrocks whereas in non-fissile mudrocks a random domain structure is usually apparent. A preferred orientation of platy particles produces an anisotropic fabric which is easily recognised in other rocks, for example slates (Davies 1980). In these rocks the anisotropy manifests itself by causing a fracture in the plane of the parallel particles. It is the author's contention that this phenomena may just as easily apply to mudrocks. It is not assumed that this is the sole reason for fissility in mudrocks, but it is considered to be one of great importance.

It remains therefore to find a mechanism by which preferred orientation is developed in some mudrocks and not in others. It is suggested that the mechanism is embodied in the physico-chemical conditions of the environment of deposition, especially the chemical conditions. The detailed chemistry is obscure but the variables concerned include Eh, pH and electrolyte concentrations and these have been described in Section 4.2.1. The control of pH on the way that clay particles interact has been demonstrated by Flegmann et al (1969) and Goodwin (1971). Colloid chemistry has highlighted the equally important role of electrolyte concentration (Van Olphen 1963).

The basic premise of the model is that a dispersal of clay particles will allow individual settlement and, because of particle shape, produce a parallel orientated microstructure. This in turn will result in the anisotropy responsible for fissility. The agents which will cause the dispersal are to be found in the organic-rich conditions prevalent in most of the anaerobic or anoxic environments referred to in Section 4.3.3. It is well documented that organic compounds are capable of dispersing clay minerals (Van Olphen 1963) and a number of these compounds are common in various depositional environments, the most notable being anoxic basins.

From a consideration of the geochemistry of these environments given in Section 4.3 it is evident that an anoxic zone exists at a depth which is rich in organic compounds derived from the decomposition of organic matter by anaerobic microbes. It is proposed in the model that clay particles have their surface charges neutralised by the organic compounds when they enter the anoxic zone (defined by a negative Eh). The compounds responsible are thought to be amino acids, fatty acids and sugars. The mechanism for the neutralisation is presented in Section 4.4.4.

The position of the zero oxidation potential relative to the sediment-water interface is critical, since it will determine whether neutralisation takes place before, at or after deposition. Where the change to anoxic conditions occurs within the water column then the falling flocculated clay particles are dispersed and the individual particles are likely to attain a preferred orientation parallel to the sediment-water interface. This will produce the strongest preferred orientation and is considered to be the situation in which the highly fissile mudrocks are deposited. When coincidence between zero oxidation potential and sediment-water interface occurs the clay particles have just begun to form a coherent sediment and although the same dispersive action takes place there is less likelihood of strong preferred orientation. With the bond between particles broken a collapse of flocs is expected but the close interaction of many particles will prevent a complete orientation with the sediment-water interface. Where the anoxic conditions lie well down in the sediment the fabric formed at the interface by the flocculated particles will be retained. The dispersive effects will then have less of an influence but it is anticipated that a weak preferred orientation could arise in places where the water content is unnaturally high. Such a situation would arise due to rapid deposition.

Following compaction and induration the degree of preferred orientation in the rock will be dependent on the initial fabric. A strong preferred orientation will be conducive to the development of fissility. When no dispersion occurs the sediment remains flocculated producing a random structure and hence induration results in a non-fissile rock. An idealized scheme of the proposed model is shown in Fig. 14.

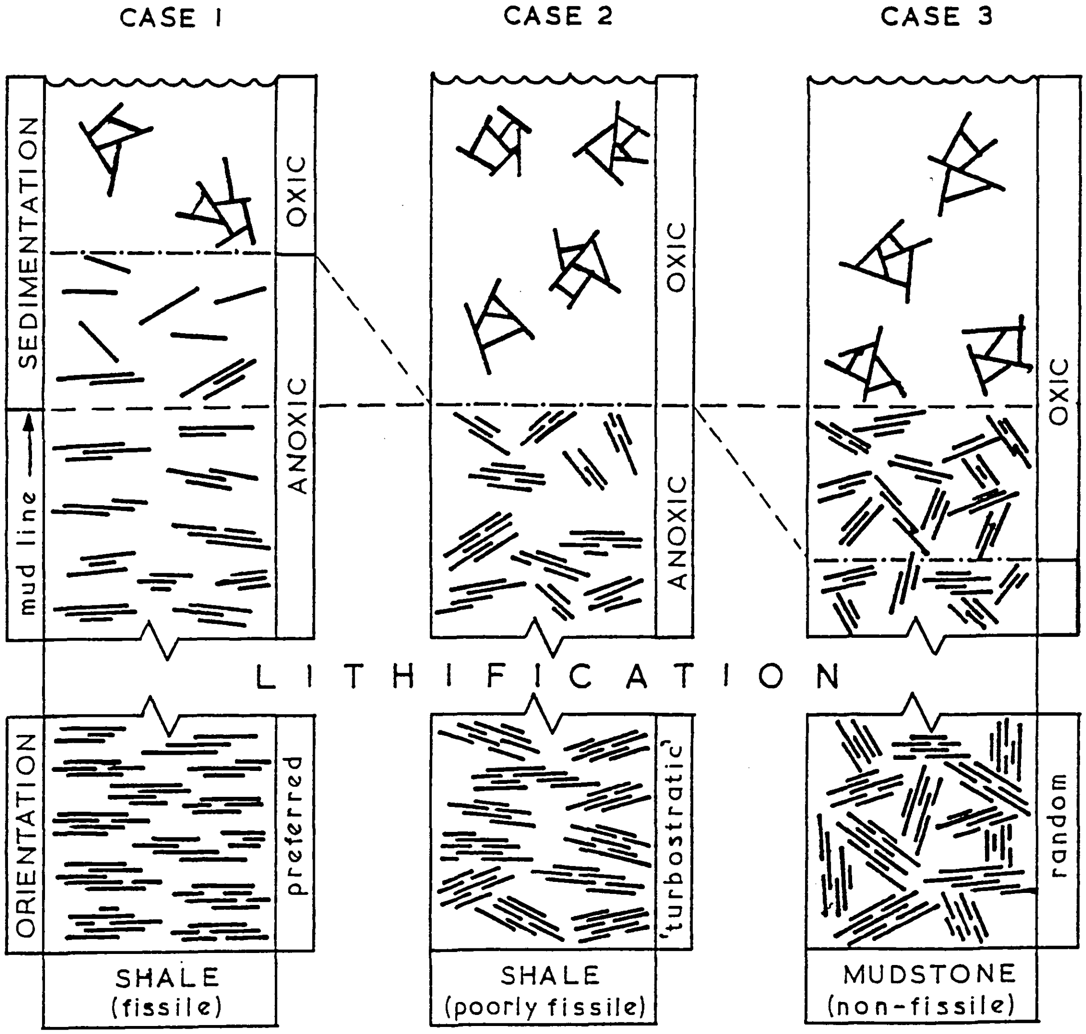


Figure 14 SUGGESTED MODEL OF SHALE AND MUDSTONE FORMATION

CHAPTER 5

MICROSTRUCTURAL OBSERVATIONS IN MUDROCKS

5.1 Introduction

There have been few SEM studies of lithified mudrock microstructure in comparison to the softer clay sediments. This imbalance may be due to the belief that the properties of rocks are more influenced by their macrostructure than their microstructure. Most of the mudrock observations have been of shales rather than mudstones.

One aspect of this study is concerned with the observation of microstructure in mudrocks derived from different depositional environments. The following section contains selected SEM micrographs of various mudrocks sampled for the study. Sample locations are given in Appendix V. Each sample is illustrated by two micrographs and these display the sample's characteristic microstructure. In addition the environment of deposition is considered for each sample with respect to the models presented in Chapter 4. The mudrocks are discussed in chronological order.

A statistical analysis of all micrographs has been carried out using the procedure described in Appendix III. A summary of the analysis is presented in Section 5.3.

5.2 Mudrocks: depositional information

5.2.1 Red Shale - Lower Cambrian : Caerfai Series

The so-called red shale of the Caerfai Series is in fact a red cleaved mudstone deposited during a shallow marine transgression of early Cambrian times. The mudstones are uniformly fine-grained and contain several discrete horizons of feldspathic tuffs. They are locally extensively bioturbated. Their red colouration is attributed to diagnetic alteration of iron silicates and volcanic ash under oxidizing conditions in which the dissolution of detrital grains provided the requisite ions for the authigenic phases (Turner 1979). The physical environment corresponds with a continental shelf area. SEM micrographs of the mudstone are shown in Figs. 15&16.

In agreement with the model the oxidizing environment has produced a random microstructure in the sample and this is confirmed by the statistical analysis of the micrographs.

5.2.2 Dolwyddelan Shale - Ordovician : Caradoc Series

The Dolwyddelan Shale is referred to in many texts as a slate, however the presence of graptolites on the 'cleavage' planes suggests that these are in fact bedding planes. Williams and Bulman (1931) have interpreted their origin as being deposited in shallow water because they conformably overlie a series of shallow water pyroclastic rocks. Deposition in an epicontinental sea is suggested.

The presence of pelagic graptolites indicates partial water circulation in the area of deposition but the restricted fauna, occurrence of pyrite in the rock and its black colour suggests a lack of oxygen and possible stagnation where the muds were deposited. According to the model, such conditions would lead to a strong preferred orientation and this is supported by the

Fig 15

RED SHALE

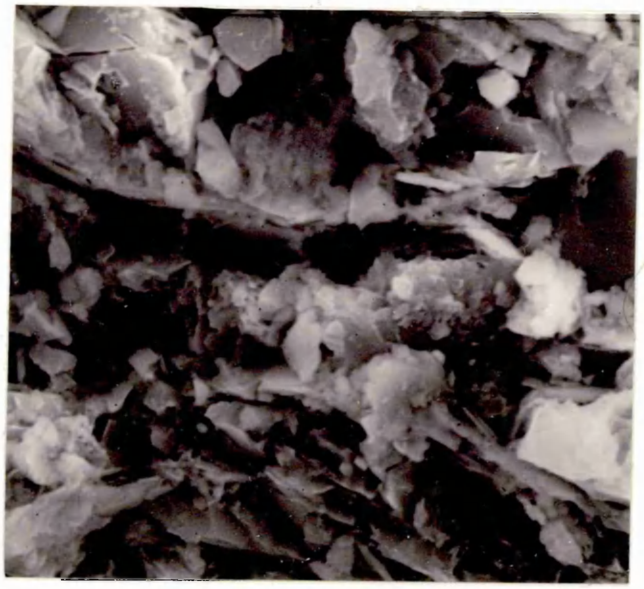


Fig 16

Fig 17

DOLWYDDELAN SHALE

Fig 18

statistical and visual evidence (Figs. 17 and 18). Williams and Bulman (1931) consider that the likely physical environment was one of shallow troughs with free egress and ingress of marine waters, but controlled by submarine barriers.

5.2.3 Cautley Mudstone - Ordovician : Ashgillian

These monotonous bluish-grey calcareous mudstones contain impure limestone nodules of diagenetic origin. The consistency of the strata and its richness in shelly fossils suggests deposition in a widespread, relatively quiet muddy shelf-sea capable of supporting considerable benthos. The environmental waters were well oxygenated in order to support the abundant trilobite and brachiopod fauna.

The results of the statistical analysis verify the apparent random structure observed in the SEM micrographs (Figs. 19 and 20).

5.2.4 Stockdale Shale - Silurian : Llandoverian

The development of the early Silurian rocks in the Lake District is best considered as a gradually deepending sequence following on from the shallow-water mudstones of the late Ordovician (Moseley 1978). The dark graptolitic Stockdale Shales were deposited under quiescent marine conditions in a trough-like anaerobic environment some distance from the shoreline (Marr 1925). Benthonic fossils are practically absent suggesting poisonous bottom waters. This is also indicated by the presence of pyrite spheroids (Fig.44) in the rock.

Cave (1979) concluded that similar black graptolitic mudrocks deposited in the Silurian basins of Wales were inter-turbidity

Fig 19 10 µm

CAUTLEY MUDSTONE

10 µm Fig 20

Fig 21

40 µm

STOCKDALE SHALE

Fig 22

sediments. He suggested that the sediments were mainly pelagic fallout through marine waters onto a sea bottom where conditions were sufficiently anoxic to allow organic matter preservation. This mechanism for yielding black shales has been referred to by Degens et al (1986).

The high degree of preferred orientation in these shales in evident from Figs. 21 and 22.

5.2.5 Yoredale Shale - Lower Carboniferous : Visean

The Yoredale Series is composed of an alternating series of limestone and shale deposits of deltaic origin (Moore 1958). The environment in which these sediments were deposited has been likened to that of the present day Mississippi delta. The series of sediments have resulted from intermittent deltaic invasions of a shallow limestone sea.

The shales of the Yoredale series are dark grey, calcareous and poorly fissile. They are interbedded with crinoidal calcite mudstones. In terms of a modern delta they may be correlated with the pro-delta facies, where mixed faunal plankton is entombed within a slowly depositing clay.

The lack of strong preferred orientation in the shales (Fig. 23 and 24) is expressed by their poorly fissile nature.

5.2.6 Coal Measure Shales - Upper Carboniferous : Westphalian A & B

Mudrocks of the Westphalian vary from black to dark grey and usually

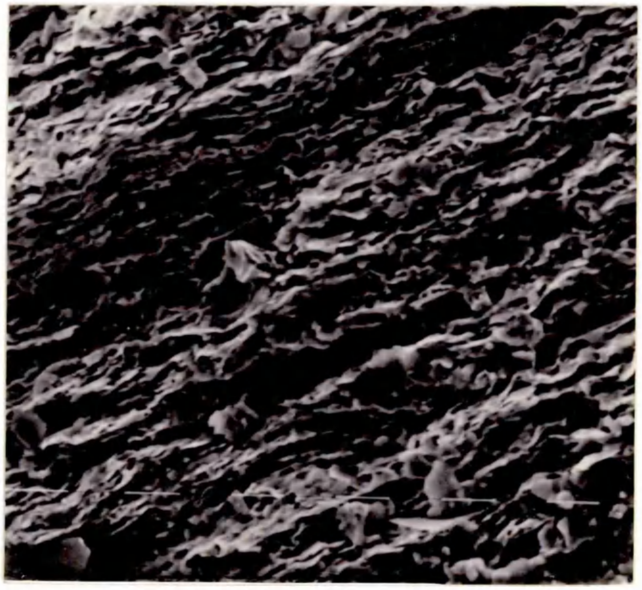
10 µm

Fig 23

YOREDALE SHALE

Fig 24

10 µm



20 µm

Fig 25

COAL MEASURE SHALE I

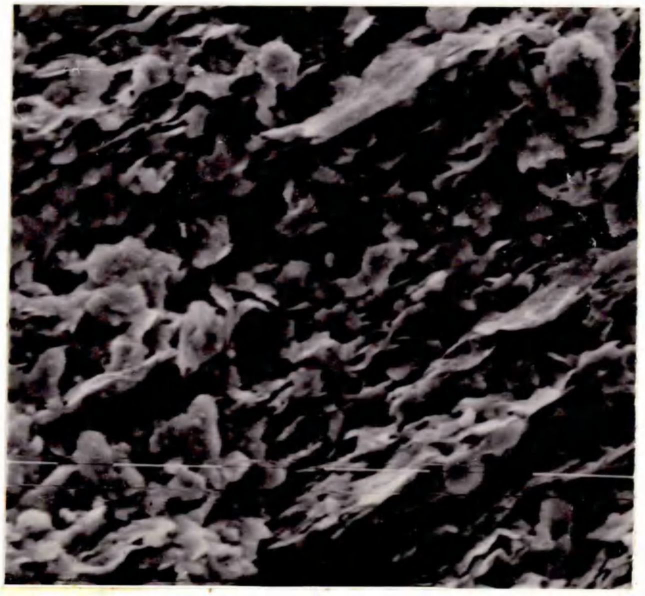


Fig 26

10 µm

pale upwards in each cycle as the silt content increases. The environment of deposition is generally accepted as being paludal, associated with the large deltas which developed during the Namurian. The deltas grew to form low lying paralic plains containing swamps and brackish and freshwater lagoons in which the sediments accumulted. Because of the low relief marine invasions were frequent giving rise to more saline waters hosting marine molluscs.

The samples chosen are a black, fissile non-marine shale obtained from the lenisulcata zone (Figs. 25 and 26) and a dark grey marine shale obtained from the similis-pulchra zone (Figs. 27 and 28). Both exhibit noticeable preferred orientation of particles. The cause of particle dispersion is considered to be due to the high organic content of the depositional waters supplied from the densely vegetated hinterland.

5.2.7 Red lutite - Permo-Triassic : Littleham Mudstone Formation

These largely structureless sediments are considered to have been deposited as overbank muds in an ephemeral lake environment under a semi-arid climate (Henson 1973). The red colouration of the beds is due to the presence of crystalline haematite which is either present as a coating to the larger sediment grains or as a fine material within the clay mineral matrix. It is believed that the main reddening of the strata occurred much later than the time of deposition (Durrance et al 1978).

The SEM micrographs of the sample (Fig. 29 and 30) reveal an open

Fig 27

COAL MEASURE SHALE 2

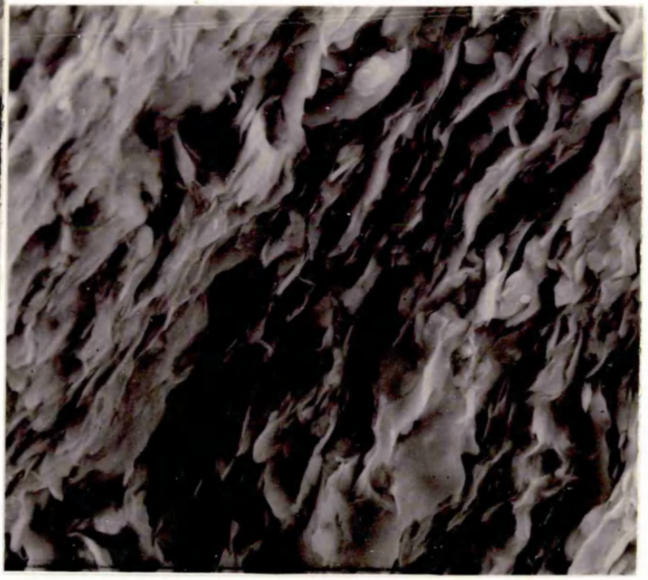


Fig 28

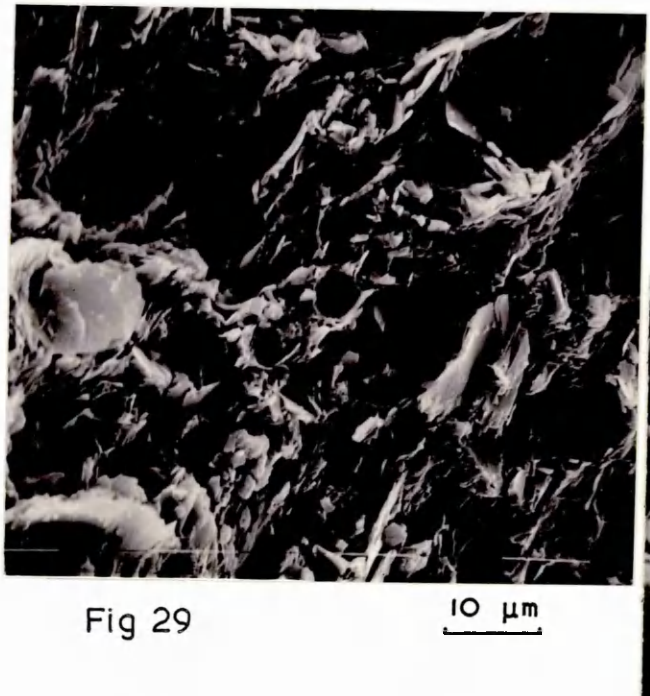


Fig 29

LITTLEHAM MUDSTONE

Fig 30

structure with an indistinct random orientation. The randomness is confirmed by the statistical assessment.

5.2.8 Bituminous Shales - Lower Jurassic : Toarcian

These black-brown shales were deposited in the shallow marine waters of an extensive sea-shelf environment in which the circulation of water was restricted by its shallowness. Hallam (1967) considered the water depth to be about 20 m, which is much shallower than alternative suggestions (Hemmingway 1974).

The sediments are characterised by a low diversity of benthic fauna and these are mainly epifaunal suspension feeders. Bioturbation is rare and the abundance of pyrite and large amounts of organic material (>4% organic carbon) testify to reducing conditions being present at the time of deposition. The oxic/anoxic boundary probably coincided with the sediment-water interface for most of the Lower Toarcian, although occasional rises of the boundary were experienced (Morris 1980).

The microstructure of the shales is shown in Figs. 31 and 32 to have a general preferred orientation which is supported by the statistical analysis.

5.2.9 Oxford Clay - Upper Jurassic : Callovian

The silty clay sediments of the Middle Callovian are considered to have been deposited relatively continuously during a transgressive marine phase. Duff (1975) suggests that the sequence was laid down fairly near to shore in quiet embayments.

Fig 31

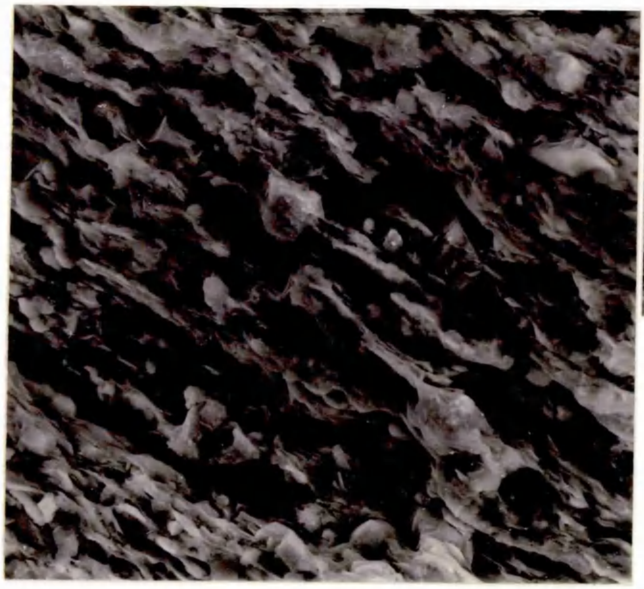


Fig 32

BITUMINOUS SHALE

20 µm

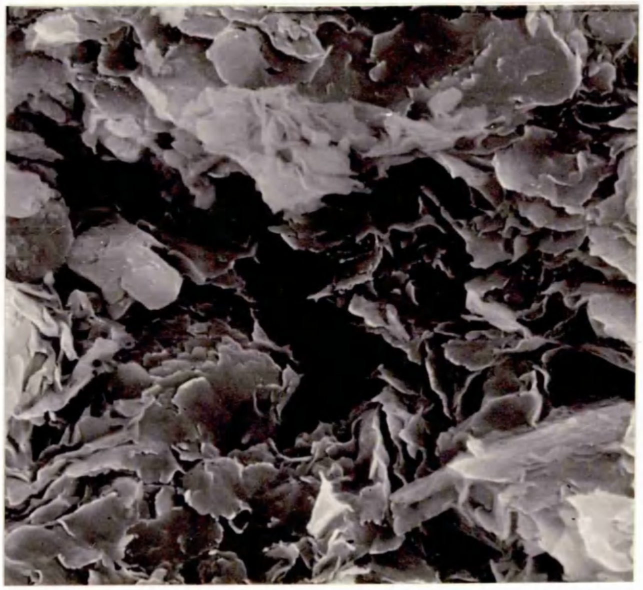


Fig 33

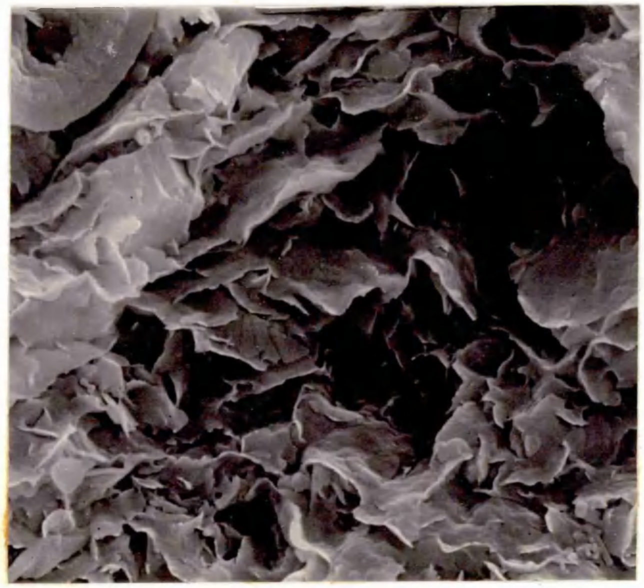


Fig 34

OXFORD CLAY

5 µm

Benthonic fossils are common, with deposit feeders being the major assemblage. This indicates oxic conditions in the bottom waters and the sediment surface. However because of abundant pyrite within the sediment Morris (1980) concluded that this oxic layer was very thin and anoxic conditions were present only a short distance below the sediment surface.

The analysis of the SEM micrographs of the Oxford Clay confirms a random orientation of particles and this is quite distinctly revealed in Figs. 33 and 34. Support for the model is again demonstrated.

5.2.10 Kimmeridge Clay - Upper Jurassic : Kimmeridgian

The Kimmeridge clay formation consists of alternating beds of organic-rich and organic-poor mudrocks which were deposited rapidly under fluctuating bottom conditions (Morris 1980). The rapid burial has aided the preservation of organic material in these sediments. The cause of the fluctuating bottom conditions is not certain. Gallois (1976) suggests a scenario in which the high productivity of algae (algal blooms) caused temporary deoxygenation, producing anaerobic bottom waters. Tyson et al (1979) believe that algal blooms occur as a result of anaerobic conditions (rather than the cause) and invoked a stratified water column model with a shifting oxic/anoxic interface to account for the variation in the beds.

What is clear,is that deposition occured below the wave base in an oxygen deficient environment (probably shallow marine basins <200 m

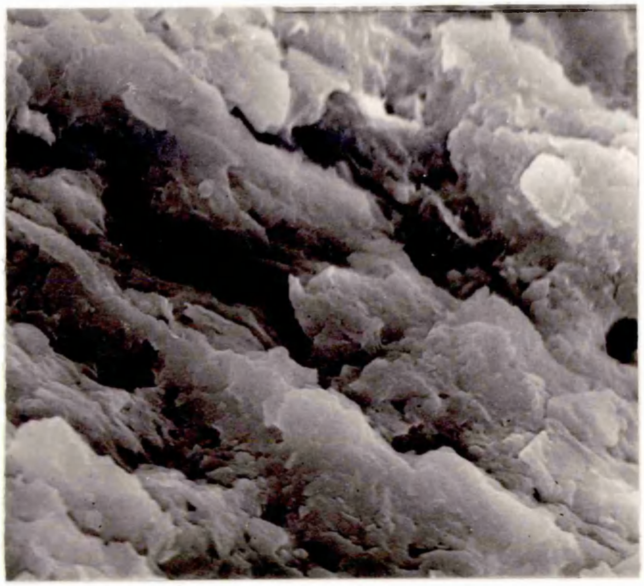


Fig 35

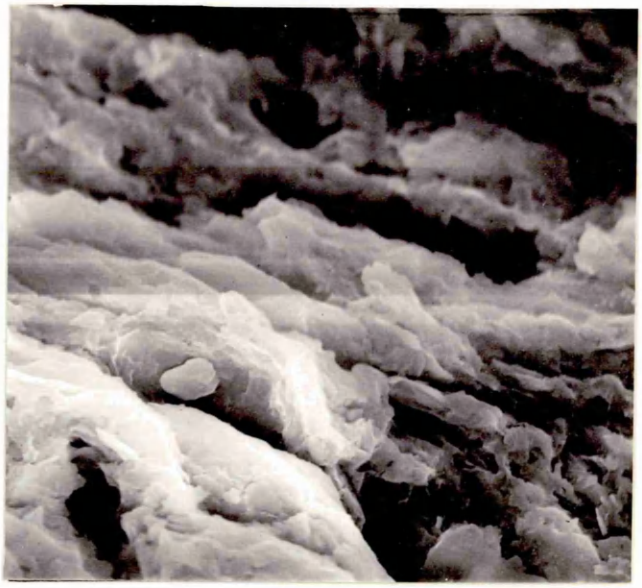


Fig 36

KIMMERIDGE CLAY

5 µm

Fig 37

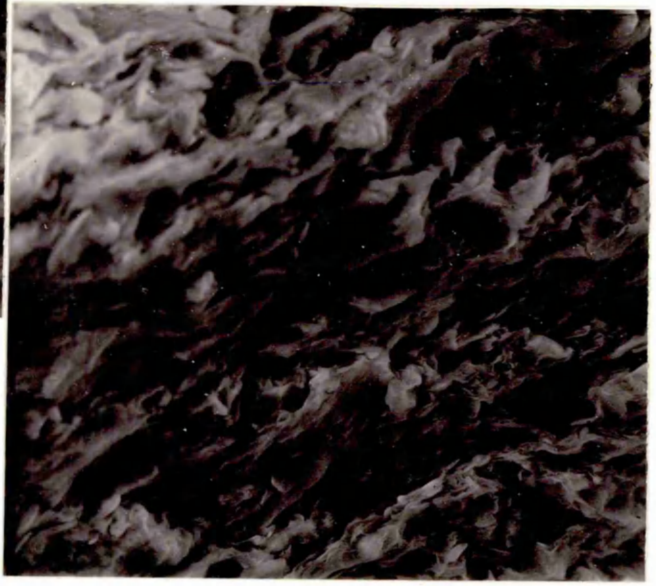


Fig 38

SPEETON CLAY (non-fissile)

20 µm

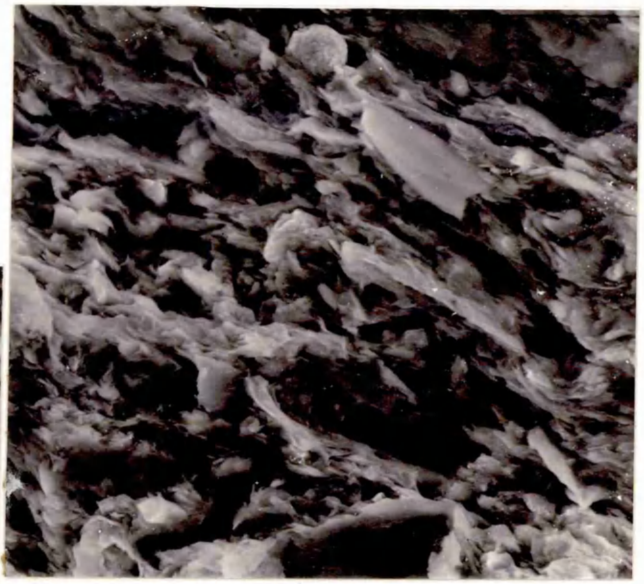


Fig 39

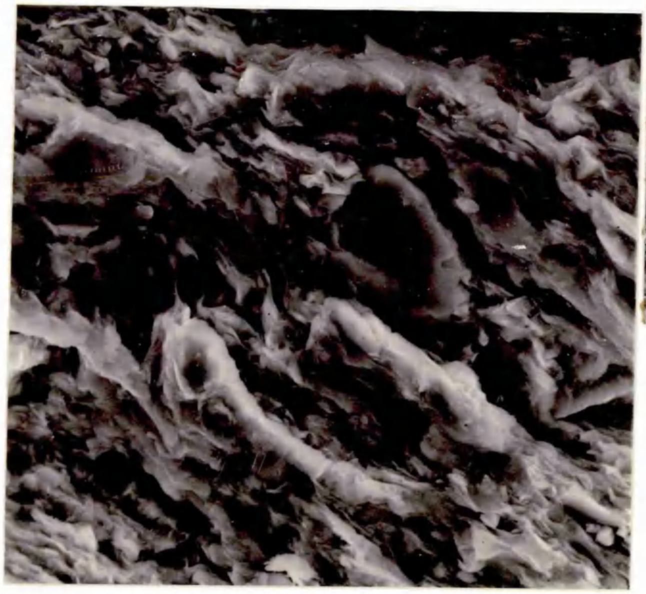


Fig 40

SPEETON CLAY (fissile)

20 µm

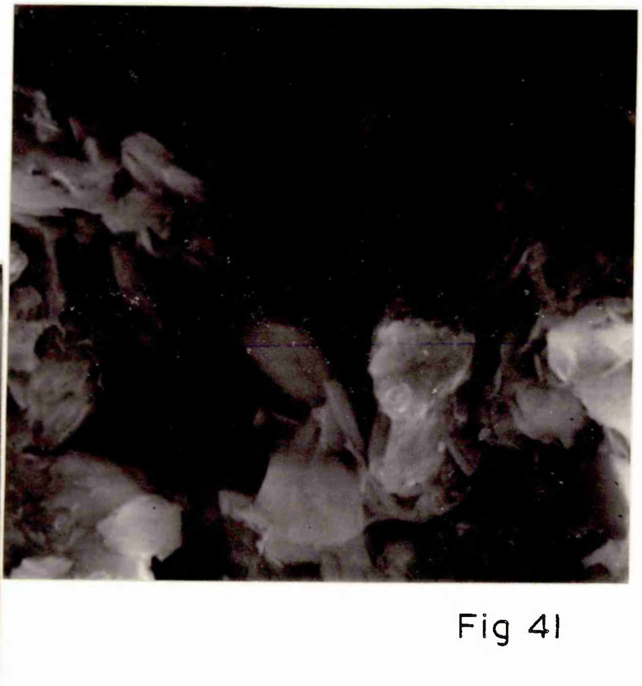


Fig 41

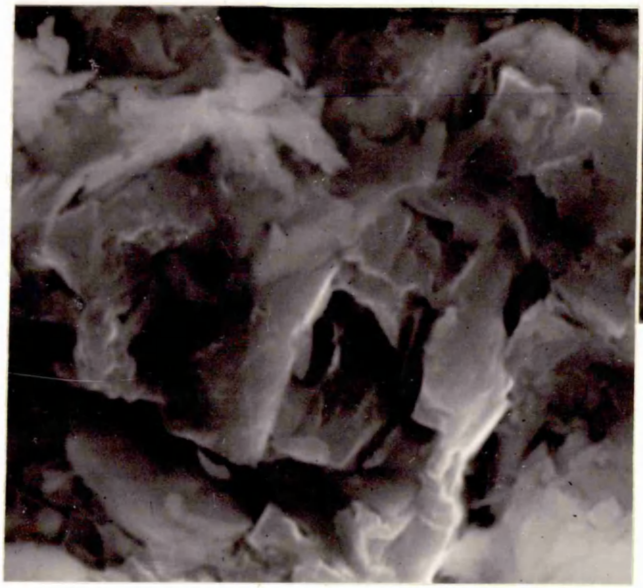


Fig 42

GREEK MUDSTONE

5 µm

deep) with high planktonic productivity. The dispersion of the clay particles, as they entered the anoxic zone in the water column, has produced a strong preferred orientation (Figs. 35 and 36).

5.2.11 Speeton Clay - Lower Cretaceous : Hauterivian

The mudrocks of the Speeton area were deposited during the early stages of a marine transgression which eventually submerged much of north west Europe. The character and amount of sedimentation was influenced by the emergence of the Market Weighton anticline.

These Lower Cretaceous sediments are predominantly soft, dark-grey to black clays containing authigenic pyrite and glauconite. Both minerals are indicative of anoxic conditions but the presence of glauconite suggests a low concentration of organic matter at the time of deposition (Berner 1981). This could account for the poor fissility displayed by certain beds within the Speeton Clay sequence.

Although anoxic conditions were present, the paucity of organic matter prevented a complete dispersion of clay particles at times. Variability in the concentration of organic matter changed the conditions of deposition repeatedly and both fissile and non-fissile strata are noted (Neale 1974). An example of the non-fissile beds are shown in Figs. 37 and 38. For comparison the fissile beds are illustrated in Figs. 39 and 40.

5.2.12 Un-named mudstones - Tertiary : Neogene?

The majority of Tertiary sediments of Europe are shallow water in origin, laid down during the withdrawal of the chalk seas of the Cretaceous. The depositional history pertaining to this particular mudstone sample from Greece is uncertain. Monopolis and Bruneton (1982) and Got (1984) provide brief details of sediments to be found in western Greece.

Both marine and freshwater facies are represented. Miocene beds are typically flysch deposits, whereas the Pliocene beds are of freshwater origin, laid down in shallow basinal lakes.

The soft mudstones of Zakynthos have a random particle orientation (Figs. 41 and 42) possibly associated with flocculation in a marine environment. According to the model the waters would be well oxygenated. Bioturbation is not evident in the sample.

5.3 Mudrocks : statistical analysis

An analysis of the particle orientations in Figs. 15 to 42 has been carried out and the results are presented in Table 3. Values for mean direction and circular variance are given on the range 0 to 180°. The values for mean resultant length are given on the ranges 0 to 180° and 0 to 360°. The statistical analysis of the data is discussed in the next section.

5.3.1 Chi-squared goodness of fit test

This showed that data from the following figures are distributed according to the Von Mises distribution and can be analysed by parametric tests.

FIG NO.	MEAN DIRECTION	CIRCULAR VARIANCE	MEAN RESULTANT LENGTH		CHI-SQUARED	ν
			$\bar{R}'$	$\bar{R}$		
15		.309	.691	.228	2.73	2
16		.214	.786	.382	4.60	2
17	33	.054	.946	.800	1.31	1
18	24.5	.043	.957	.832	2.64	1
19		.265	.735	.292	3.26	2
20		.253	.747	.311	5.11	2
21	175.5	.020	.980	.922	*	
22	126.5	.023	.977	.911	*	
23		.286	.714	.260	0.72	1
24		.194	.816	.423	0.28	1
25	24	.014	.986	.945	*	
26	32	.046	.954	.828	*	
27	66	.042	.958	.843	0.56	1
28	59.5	.082	.918	.709	2.65	1
29		.273	.727	.279	0.21	1
30		.255	.745	.309	2.54	1
31	147	.065	.935	.765	1.95	1
32	145	.030	.970	.885	3.17	1
33		.255	.745	.309	1.29	1
34		.288	.712	.257	1.10	2
35	145	.021	.989	.917	3.27	1
36	152	.067	.933	.759	4.67	2
37		.283	.717	.265	7.58	3
38		.203	.797	.403	4.81	2
39	149	.043	.957	.838	1.10	1
40	145	.048	.952	.821	3.10	1
41		.378	.622	.149	5.63	3
42		.466	.534	.081	4.23	3

* insufficient data in some cells

TABLE 3: ORIENTATION ANALYSIS OF SEM MICROGRAPHS

15, 16, 17, 18, 19, 20, 23, 24, 27, 28, 29, 30,
31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42.

The following figures provided data which did not fit the Von Mises distribution and in these cases non-parametric tests are carried out.

21, 22, 25, 26.

5.3.2 Initial data analysis

From Table 3 the values of circular variance indicate that some data sets (e.g. Fig. 21) are clustered tightly about the mean direction, whilst others (e.g. Fig. 15) are distributed around the circle. This confirms that the particles within certain mudrocks do have a preferred orientation.

None of the sets shows perfect randomness, where S_o approaches unity, but a distinction between various sets of data is obvious. This is most noticeable when the mean resultant lengths are calculated over the range 0 to 360^o.

To emphasise this distinction the results from the orientation analysis of many SEM micrographs are presented in Fig. 43. From this histogram it is clear that the particle arrangements in the mudrocks examined define two discrete categories of orientation. Mudrocks with a visually strong preferred orientation give mean resultant length values in excess of 0.70. More randomly orientated microstructures generally have mean resultant lengths of 0.45 and less.

The initial data analysis indicates that mudrock particles may adopt a preferred or random orientation. By comparing the value of the mean resultant length to the conditions of deposition for the mudrock, a pattern emerges in which the strongly orientated microstructure correlates with deposition in an anaerobic/reducing environment.

5.3.3 Results from the test of uniformity

Table 4 gives the results of the Rayleigh test carried out on the Von Mises distributed data. For a data sample of 50 particles the critical value of $\bar{R}$ at the 0.1% level is 0.367. From Table 4 it is noted that a group of micrographs have $\bar{R}$ values in excess of 0.367 and it may be concluded with confidence that these show preferred particle orientation. $\bar{R}$ values for Figs. 16 and 38 fall just outside the critical value but their particles are not strongly orientated. The micrographs with $\bar{R}$ less than the critical are considered to have random particle orientation.

Table 5 gives the results of Watson's U^{*2} test for data which was not Von Mises distributed. At the 1% level the critical value of U^{*2} is 0.267. Hence it is concluded that the arrangement of particles in Figs. 21, 22, 25 and 26 are not randomly orientated and they have a preferred direction.

5.3.4 Confidence interval results

Table 6 lists the 95% confidence intervals for the population means

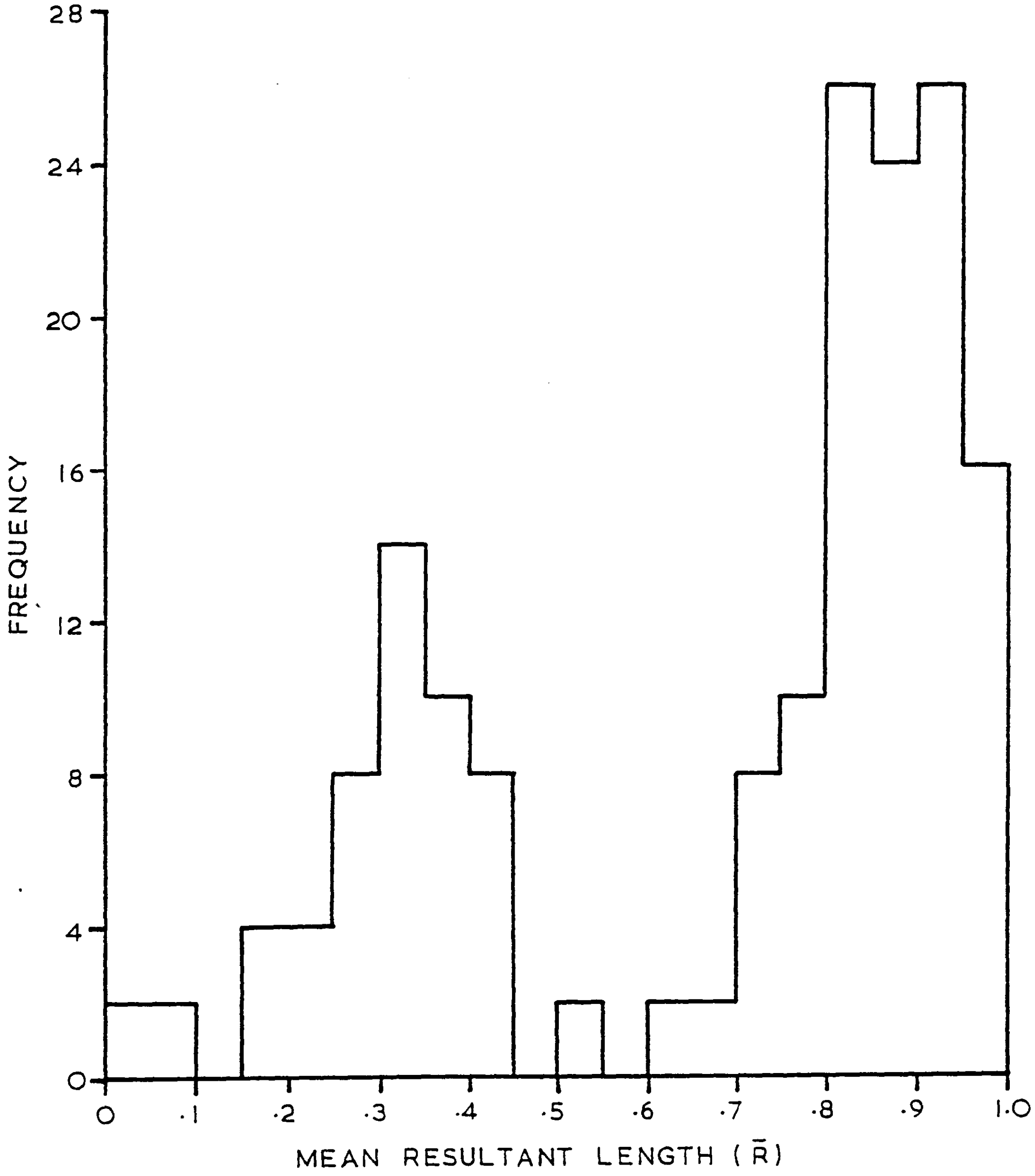


Figure 43 ORIENTATION ANALYSIS RESULTS OF MUDROCK SAMPLES

PARTICLE ORIENTATION		
PREFERRED $\bar{R} \gg 0.367$	WEAKLY PREFERRED $0.367 < \bar{R} < 0.5$	RANDOM $\bar{R} < 0.367$
17, 18	16	15
27, 28		19, 20
31, 32	24	23
35, 36		29, 30
39, 40		33, 34
	38	37
		41, 42

TABLE 4. RESULT OF RALEIGH TEST OF UNIFORMITY ON SAMPLE PAIRS OF VON MISES DISTRIBUTED DATA

FIG NO	U^{*2}
21	0.355
22	0.369
25	0.325
26	0.338

TABLE 5. RESULTS OF WATSON'S U^{*2} TEST OF UNIFORMITY ON NON VON MISES DISTRIBUTED DATA

FIG NO	SAMPLE MEAN DIRECTION	ALLOWED DEVIATION	POPULATION MEAN DIRN.	
17	33	± 11	20	*
18	25	± 9	20	
27	66	± 9	68	
28	60	± 13	68	
31	147	± 12	155	
32	145	± 7	155	*
35	145	± 6	149	
36	152	± 12	149	
39	148	± 10	154	
40	153	± 11	154	

*sample mean does not fall within population mean deviation

TABLE 6. CONFIDENCE INTERVAL TEST ON VON MISES DISTRIBUTED DATA SHOWING PREFERRED PARTICLE ORIENTATION

(i.e. fissile planes) of the Von Mises distributed data. In most cases the population mean falls within the confidence limits. This indicates that the preferred orientation of particles within these samples does not differ significantly from the measured angle of the fissile plane in each case. It is therefore confirmed that, for the mudrock samples analysed, there is a definite correlation between the direction of the fissile plane and the orientation of microstructure.

5.3.5 Comparison of data from the same specimen or sample

It is established in Appendix III, Section 4.0 that a change in the magnification used to observe particles in a particular specimen of Stockdale Shale does not alter the derived degree of orientation significantly. This test was undertaken on other mudrock samples and the same conclusion was reached in each case. Thus it is assumed that any magnification may be chosen which is suitable for data aquisition. Measurements taken at that magnification will be minimally biased.

Since only two example micrographs have been presented for each sample a comparison of data is made using two sample testing methods only. The results are given in Tables 7 and 8. It should be noted that where data is derived from the same specimen the tests outlined in Appendix II (Sections 4.3 and 5.2) can be used. For Figs. 21 and 22 the data did not come from the same specimen and hence the different orientations in the SEM precludes the use of the above tests. In this case a comparison is made using the assumption that the value of $\bar{R}_1{}^2/\bar{R}_2{}^2$ for the two sets of data approaches unity if

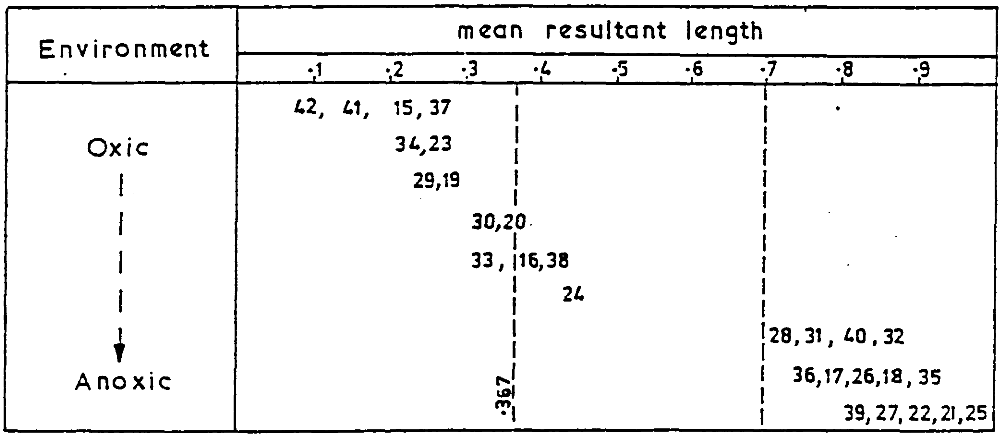
FIG NOS	R1 + R2	R	K	$2K(R_1 + R_2 - R)$	
15, 16	30.52	29.66	0.61	1.05	
17, 18	81.89	80.99	2.95	5.26	*
19, 20	30.19	24.11	0.51	6.17	*
23, 24	34.14	34.02	0.75	0.19	
27, 28	77.57	77.07	2.60	2.65	
29, 30	29.38	29.33	0.63	0.06	
31, 32	82.51	82.44	3.15	0.43	
33, 34	28.29	26.08	0.54	2.40	
35, 36	83.78	83.19	3.38	4.05	*
37, 38	33.37	33.15	0.70	0.31	
39, 40	82.93	82.69	3.30	1.57	
41, 42	11.53	11.37	0.23	0.07	

*test value exceeds critical

TABLE 7. WATSON AND WILLIAMS TWO SAMPLE TEST ON VON MISES DISTRIBUTED DATA

FIG NOS	$\bar{R}_1$	$\bar{R}_2$	$\bar{R}_1^{\,2}/\bar{R}_2^{\,2}$	$2nR^2/n_1n_2$
21, 22	.980	.977	1.006	-
25, 26	.986	.954	-	0.062

TABLE 8. NON-PARAMETRIC TESTS FOR TWO SAMPLES



numbers refer to figures 15-42

TABLE 9. COMPARISON BETWEEN ENVIRONMENT OF DEPOSITION AND PARTICLE ORIENTATION

the degree of preferred orientation is similar within each sample. This value is checked against the Fisher statistic which has a critical value of 1.41 at the 5% level of significance.

The results in Tables 7 and 8 confirm that in most cases the orientation of particles within each specimen does not alter significantly if sampled from different parts of the specimen, or in the case of the Stockdale Shale from different specimens.

5.3.6 Statistical conclusions

On the evidence of the analysis presented in Tables 3 to 8 it is concluded that fissile mudrocks display a greater degree of particle orientation than non-fissile mudrocks. A comparison of particle orientation with depositional environment is summarised in Table 9. The correlation suggests that mudrocks with preferred particle orientation are deposited under reducing conditions.

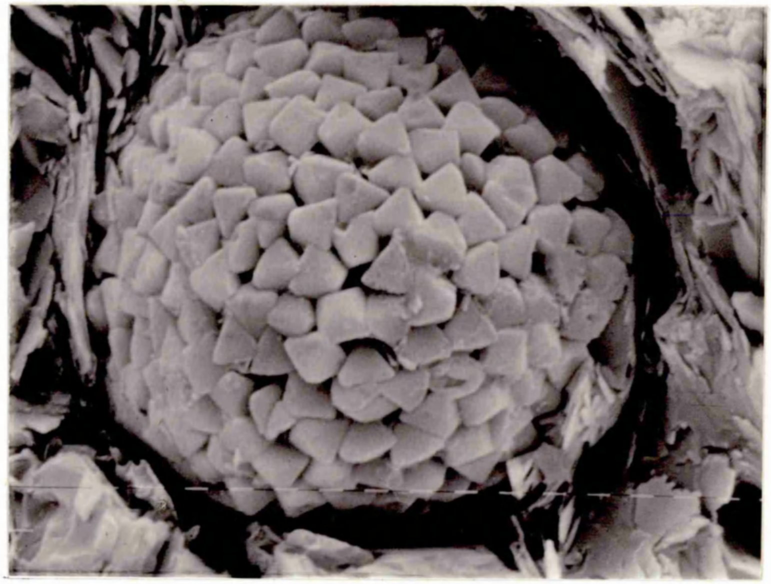


Figure 44 SEM micrograph of a pyrite framboid

CHAPTER 6

EXPERIMENTAL MODEL AND OBSERVATIONS

6.1 Previous work

In most other studies of the microstructure of laboratory prepared clay specimens, the normal practice has been to prepare a slurry by mixing the clay soil or monomineralic powder with water to moisture contents in the order of 100-250 per cent (say 1½-4 times the liquid limit). The prepared slurry has then been consolidated by various means and samples obtained for examination by the standard methods (Tovey and Wong 1973). In order to achieve the different clay fabrics artificial flocculating and deflocculating agents have been added to the clay slurry prior to consolidation (Krizek et al 1975).

Kirkpatrick and Rennie (1972) mixed quantities of kaolin powder with an equal mass of de-aired water, using a paddle-mixer, to achieve a slurry water content of about 100%. The slurry was consolidated in an extended Rowe cell under one-dimensional conditions to maximum pressures of 690 kPa. They obtained moisture contents of around 45% at this high pressure indicating a reduction in void ratio from about 2.5 to 1.2. Small specimens (8 mm diameter) were cut from the main sample blocks and air dried prior to fracturing. The fractured surfaces were cleaned of debris by peeling with adhesive tape before preparation for viewing in the SEM.

McConnachie (1971) used a purified kaolin mixed with distilled water at a moisture content of 250% to form a slurry which was consolidated initially in a specially designed cell. A substitution drying method, using acetone and subsequently a polyester resin, was employed to remove the pore water in the samples. Ultra thin sections were then prepared for study in the TEM.

An unusual approach was adopted by O'Brien (1972) in which one gram of illite, initially dispersed in a dilute solution of NH_4OH, was flocculated with a NaCl solution to produce an illite suspension at about 1000% moisture content. This was allowed to drip slowly into glass sedimentation tubes and the supernatant liquid siphoned off after a period of time. The process was repeated and a naturally consolidated sample of about 17 cm height was produced in the sedimentation tube. On air drying the 1 gm per litre sample decreased in height to 10 cm. O'Brien considered this method to approximate natural sedimentary conditions in a marine environment.

In an attempt to simulate the geological history of clay by artificial means Knill et al (1976) developed a comprehensive package of equipment which controlled not only the overburden pressure but also the pore fluid pressure and geothermal gradient. They recognised the importance of minimal disturbance to the sample prior to consolidating and used an extension column in order to sediment the clay slurry directly into the specimen cup.

Tchalenko (1968) has reported of sedimenting London clay, at an initial moisture content of 750%, directly into an oedometer pot fixed at the base of a settling column. A salt water medium of 36 g/l NaCl in distilled water was used. He recognised the limitations of comparing artificially sedimented samples with natural clays, but as far as structure is concerned his results showed strong compatibility.

A notable study by Owen (1970) looked at the properties of consolidating muds with respect to particle concentration, salinity and bed thickness. Mud suspensions were pumped into a 10 m high perspex settling column and left to settle. Owen observed that at certain particle concentrations the settling velocity was reduced by the onset of hindered settling. At this stage the normal settling process is replaced by a consolidation process in which water passes through the pores between particles. Owen found that, depending upon the salinity, hindered settling occurred with particle concentrations of between 4 and 20 g/l.

Settling columns were also adopted by Been and Sills (1981) in their geotechnical investigation of soft soil consolidation. They used an estuarine mud mixed with tap water to form a slurry of the appropriate density (about 10 kN/m^3). Particle concentration was high (300-350 g/l) which indicates that the sediment was undergoing consolidation rather than settling.

6.2 A model for sediment accumulation

The objective of modelling sediment accumulation is to produce a laboratory sediment by a method similar to how a natural sediment accumulates. The model adopted will depend on the type of sediment involved and the processes which are assumed to act upon it in the natural state. The predominant influence apparent in the accumulation of sediment is that of gravitational force. Sediment is transported in suspension as long as the flow velocity is high or turbulence exists. Once the velocity or turbulence reduces sufficiently to allow gravitational forces to be dominant then the particles begin to settle and form a bed. The particles, or flocs, are be brought closer together as the sediment concentration increases, until they finally make physical contact. With additional material settling on top the degree of contact increases, and this causes an expulsion of pore water from within the sediment. At some juncture the sediment ceases to behave as isolated particles and takes on the properties of a soil which may be described by traditional geotechnical parameters.

6.2.1 Physical aspects

It was considered important to achieve a model in which sediment settled out of suspension and formed a bed by a process of growth from the base upwards, since this reflected natural accumulation. In this respect the initial concentration of solids becomes important. When slurries are used with moisture contents in the order of 250% (400 g/l) the sediment settles from the upper surface and no actual bed forms at the base. This produces hindered settling (Owen 1970) where water is being forced out of the sediment in a manner similar to the consolidation process, as opposed to the normal settling process of particles moving through water. Since the initial fabric will depend upon the physical and chemical processes that have acted on the particles prior to their arrival at the sediment bed it is necessary to allow for these processes to act. It is evident that when clay slurries produce hindered settling the physical characteristics of sediment accumulation are not met. The levels of sediment concentration and method of deposition in this study were chosen carefully to avoid hindered settling.

6.2.2 Chemical aspects

The natural chemistry of the depositional environment may be modelled more easily. A marine environment was achieved by dissolving 'instant ocean' crystals in water at the specified concentration. By varying the concentration, salt water conditions from mildly brackish to hypersaline could be modelled. This novel method of producing the flocculating environment was considered more appropriate than using artificial flocculating agents, and to the author's knowledge had not been used previously. The role that organic matter plays in influencing clay microstructure was effected by adding the required compounds to the water mixture before or during the sedimentation phase.

6.2.3 Biological aspects

It is recognised that biological activity affects the microstructure of clay sediments and can be a significant factor in the process of sedimentation. This aspect was outside the scope of the study and not considered appropriate for the model. However, if the correct biological conditions to support life are provided then a variety of suspension feeding and bottom feeding organisms may be introduced into the model.

6.2.4 Consolidation aspects

A natural sediment undergoes a dewatering phase in response to the increasing overburden pressure. For practical reasons was not possible to continue deposition and accumulate a sufficient depth of sediment to provide that pressure. Thus to model the effects of overburden it was necessary at some stage to resort to laboratory consolidation methods. The change from the deposition phase to sediment consolidation should occur with a minimum of disturbance to the sample. For this reason sediment was deposited directly into a consolidation cell and then subjected to a vertical pressure to simulate an increasing overburden thickness.

6.2.5 Laboratory procedure

Details of the experimental set-up and procedure may be found in Appendix I. The method involved mixing dry clay powder with 'prepared' water in a preparation tank. The mixture was then pumped into a settling column where it was allowed to settle into a consolidation cell clamped at the base of the column. When all the sediment had settled the cell was removed and the clay slurry consolidated in the usual manner (Vickers 1983). The maximum consolidation pressure applied was 10^3 kPa, which is equivalent to about 100 m of overburden sediment. Although mudrocks are compacted to far greater pressures than this their microstructure is often determined at low pressures and does not alter significantly upon further consolidation (Martin 1965).

6.3 Experimental programme

This phase of the study consisted of producing clay sediments by controlled deposition in various chemical environments under similar physical conditions. The main objective was to determine to what extent the chemistry of the deposition waters influences the microstructure of the clay sediments. A monomineralic sediment was chosen in order to reduce the variables.

The consolidation characteristics of each sediment were determined by standard soil mechanics methods. These characteristics were related to the microfabric produced in the sediment.

The microfabric was observed by electron microscopy and the degree of preferred particle orientation obtained using statistical methods. This enabled a comparison to be made between the microfabric of laboratory sedimented clays and that of natural clay sediments and mudrocks.

6.3.1 Clay sedimentation

The procedure for sedimentation is recorded in Appendix I. A total of eleven laboratory sedimentation tests were carried out including five with the addition of organic chemicals. The different tests are given in Table 10.

The non-organic tests were carried out to provide evidence of the basic microstructure obtainable under freshwater and saltwater conditions. The aim of the organic tests was to determine if naturally occurring organic compounds could encourage a preferred orientation under laboratory conditions.

The organic chemistry of the environment of deposition is obviously very complex. Since the aim was to consider the influence of individual organic compounds, mixtures of compounds were not used. The choice of organic compound to be tested was governed by its relative abundance and its link with hydrocarbon formation. Principal sources for this information included Degens (1965), Rittenberg et al (1963), various contributions in Eglinton and Murphy (1969) and Bordovskiy (1965). According to these sources the

TEST NO	CLAY MINERAL	WATER CONDITIONS	ORGANIC COMPOUND	FIGURES
1	K	Fresh		45, 46
2	K	Brackish		47, 48
3	K	Salt		49, 50
4	I	Fresh		51, 52
5	I	Brackish		- -
6	I	Salt		53, 54
7	K	Salt	aspartic acid	55, 56
8	K	Salt	palmitic acid	57, 58
9	K	Salt	glucose	59, 60
10	I	Salt	palmitic acid	61, 62
11	I	Salt	aspartic acid	63, 64

K - kaolinite I - illite

Table 10. CLAY SEDIMENTATION TESTS

TEST NO	CONSOLIDATION c_v (mm^2/min)	PARAMETERS C_c	SHEAR STRENGTH kN/m^2	VOID RATIO	$\bar{R}$
1	2.36	0.43	13.7	1.01	0.851
2	7.52	0.51	6.0	1.14	0.241
3	9.21	0.56	7.4	1.16	0.131
4	3.09	0.80	13.0	1.11	0.805
5	3.29	0.92	11.5	1.62	0.652
6	3.82	0.95	10.1	1.67	0.681
7	2.57	0.45	9.4	1.03	0.713
8	2.66	0.44	10.7	1.04	0.757
9	6.77	0.48	9.6	1.20	0.451
10	1.45	0.83	14.8	1.08	0.818
11	1.81	0.85	16.1	1.13	0.809

Table 11. GEOTECHNICAL PROPERTIES OF SEDIMENTED CLAYS

organic compounds which appear to satisfy both requirements are amino acids, fatty acids and sugars.

6.3.2 Geotechnical data

Following the sedimentation phase each clay sample was subjected to standard laboratory consolidation in which porewater pressure variation and settlement were recorded. From this data graphs of settlement against time and void ratio against effective stress were plotted in order to determine the consolidation characteristics. Prior to sampling, the shear strength of the consolidated clay was measured. The consolidation and shear strength data are summarised in Table 11.

Consolidation characteristics

The geotechnical properties of a clay sediment often depended upon its geological history. An important aspect of the geological history is the manner in which the sediment wass deposited and the variation of stresses due to subsequent burial and erosion. This history becomes 'locked' into the sediment and is usually revealed in the measurement of its geotechnical properties. With regard to clay sediments foremost amongst these properties are the consolidation characteristics.

Under one dimensional consolidation conditions Terzaghi (1943) showed that the time taken for a layer of clay soil to settle under a given increase in load is related to its coefficient of consolidation (c_v). The value of c_v is obtained from the settlement-time graph for a particular load and is approximately constant over a large range of pressure. The reason for this is because c_v is directly related to the permeability (k) and inversely related to the compressibility (m_v) of a clay . Since under normally consolidated conditions both permeability and compressibility decrease with increasing pressure, changes in their values are self cancelling and the ratio k/m_v is fairly constant. Average values of the coefficient of consolidation for each sediment, as determined by Taylor's square root time fitting method (Vickers 1983 : chapter 5), are given in Table 11. The value of c_v is dependent on the sediment structure and this may be compared to the degree of preferred particle orientation.

For a sediment that has undergone the natural process of consolidation the relationship between its void ratio (e) and corresponding overburden pressure (p) is approximately logarithmic. If the values are plotted to a semi-log scale (e to a natural scale, p to a logarithmic scale) the result is a straight line whose slope is called the compression index (C_c). This index is a measure of the compressibility of a sediment and will vary depending upon the type of clay and the various physico-chemical effects associated with electrolyte concentration, cation valency, dielectric constant etc (see Chapter 4).

Shear strength characteristics

The shear strength of a sediment increases with natural consolidation as a result of a reduction in moisture content due to the closer packing of particles. Hence shear strength is inversely proportional to void ratio. The effect of the chemistry of depositional waters is to vary the microstructure of a clay sediment. Normally the more flocculated the structure the greater the void ratio and this should be evident from the shear strength value if the same type of sediment has been deposited under different chemical conditions.

Shear strength measurements were obtained using the falling cone penetrometer method proposed by Hansbo (1957). The values of shear strength for the various sediments are given in Table 11.

6.4 Statistical analysis

A selection of SEM micrographs of specimens taken from the various sedimented clays are illustrated in Figs. 45-64. The microstructure in each case was defined by measuring the angular orientation of its clay particles. The statistical methods outlined in Appendix II were used to evaluate the orientation and assess its significance. The data obtained from each micrograph is presented in Table 12. Values for mean direction and circular variance are quoted on the range 0 to 180° whereas the mean resultant length is on the ranges 0 to 180° and 0 to 360°.

The angular values for each micrograph were checked against a Von Mises distribution using a chi-squared goodness of fit test. This revealed that all data matched a Von Mises distribution and thus parametric testing was carried out.

The initial results from Table 12 show that in the majority of cases the value of $\bar{R}$ exceeds the critical value of 0.367 at the 0.1% level of confidence. Thus a preferred orientation is indicated. For the brackish and saltwater sedimented kaolinite the clay particles are shown to be randomly orientated. A comparison of the

FIG NO	MEAN DIRECTION	CIRCULAR VARIANCE	MEAN RESULTANT LENGTH		CHI-SQUARED (ν = 1)
			$\bar{R}'$	$\bar{R}$	
45	117	.037	.963	.859	0.68
46	112	.040	.960	.850	1.84
47	—	.323	.687	.211	5.35*
48	—	.256	.744	.307	0.64
49	—	.390	.610	.139	0.37
50	—	.424	.576	.110	0.74
51	168	.052	.948	.808	2.22
52	165	.053	.947	.806	0.15
53	167	.127	.873	.580	0.35
54	168	.118	.882	.605	0.48
55	165	.071	.929	.744	1.12
56	161	.090	.910	.685	0.51
57	165	.055	.945	.800	0.41
58	169	.080	.920	.717	2.25
59	171	.156	.844	.509	3.63
60	160	.199	.801	.412	0.96
61	172	.053	.947	.806	0.23
62	169	.037	.963	.859	0.87
63	164	.042	.958	.842	2.93
64	163	.050	.950	.815	0.16

* ν = 2

TABLE 12 ORIENTATION ANALYSIS OF SEM MICROGRAPHS OF LABORATORY SEDIMENTS

Fig 45

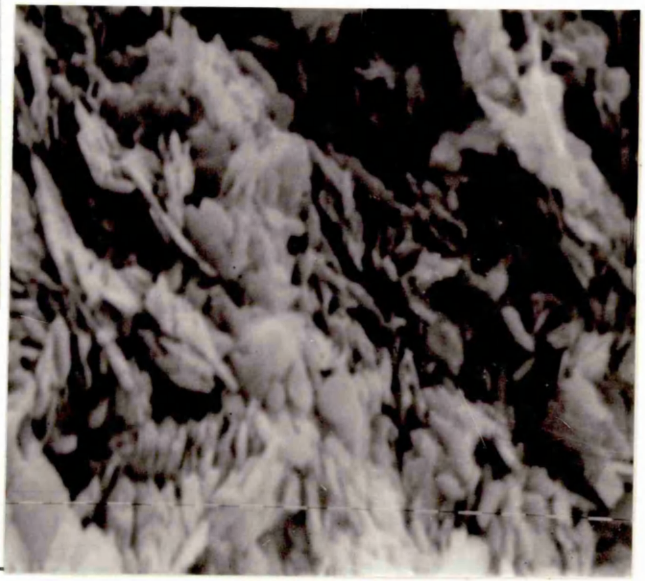


Fig 46

KAOLINITE :- freshwater

5 µm

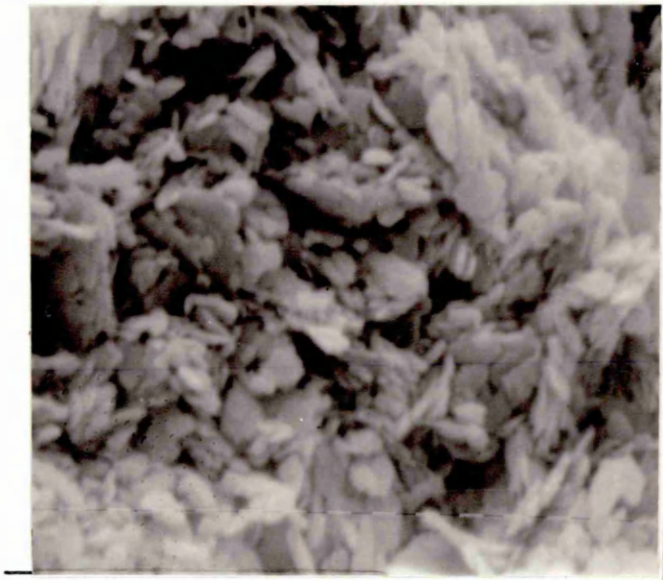


Fig 47

Fig 48

KAOLINITE :- brackish-water

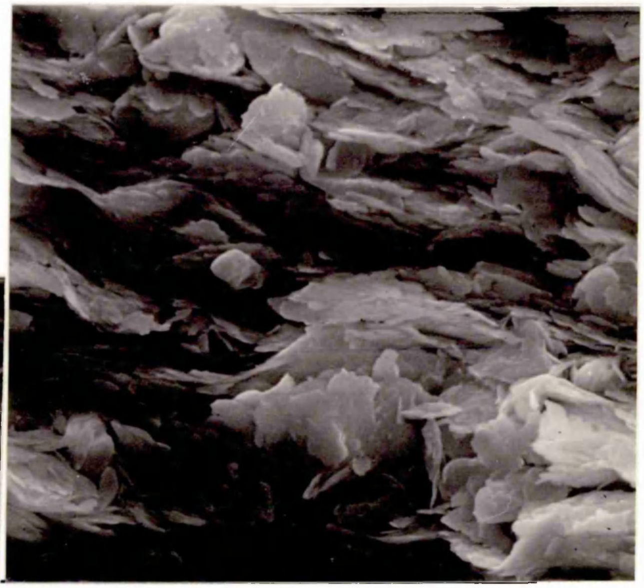
5 µm

Fig 49

KAOLINITE — saltwater

3 μm

Fig 50



2 μm

Fig 51

ILLITE — freshwater

Fig 52

2 μm

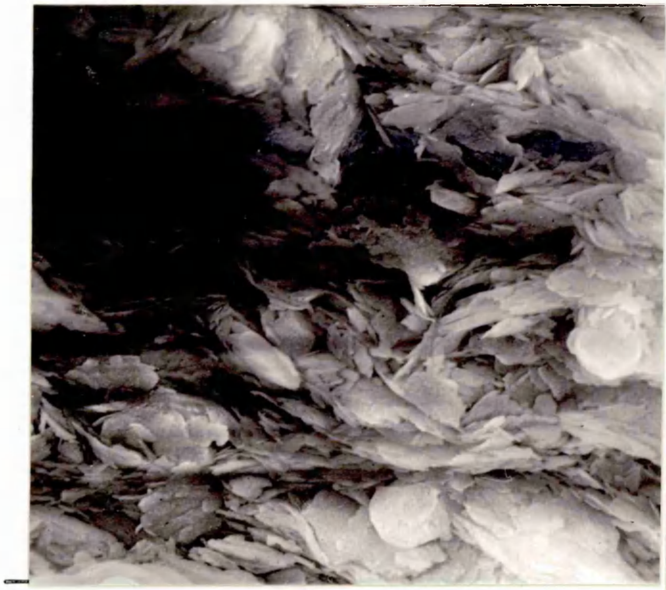


Fig 53

ILLITE — saltwater

2 µm

Fig 54

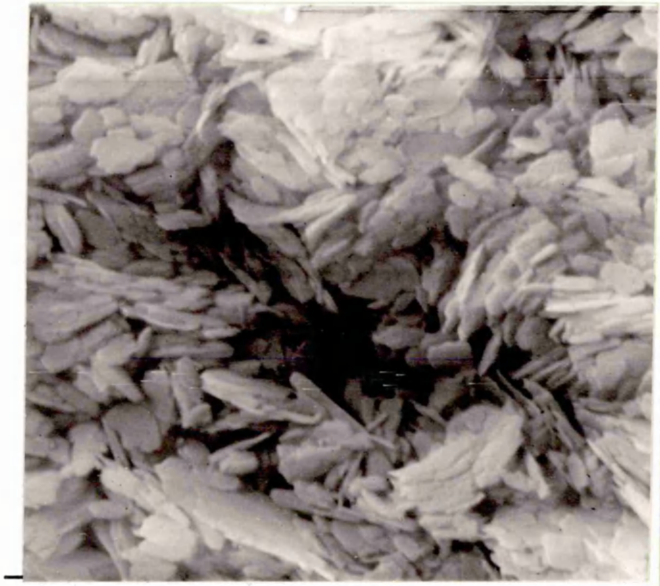


Fig 55

KAOLINITE + aspartic acid

2 µm

Fig 56

Fig 57

KAOLINITE + palmitic acid

2 µm

Fig 58

Fig 59

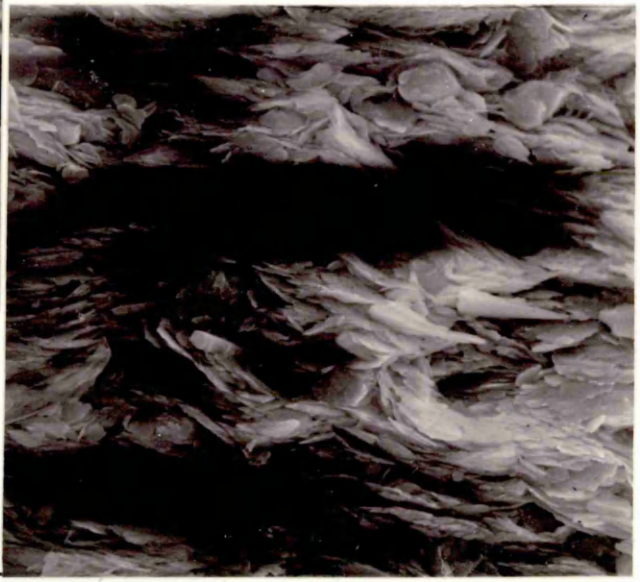
KAOLINITE + glucose

2 µm

Fig 60

Fig 61 2 μm

ILLITE + palmitic acid



2 μm Fig 62

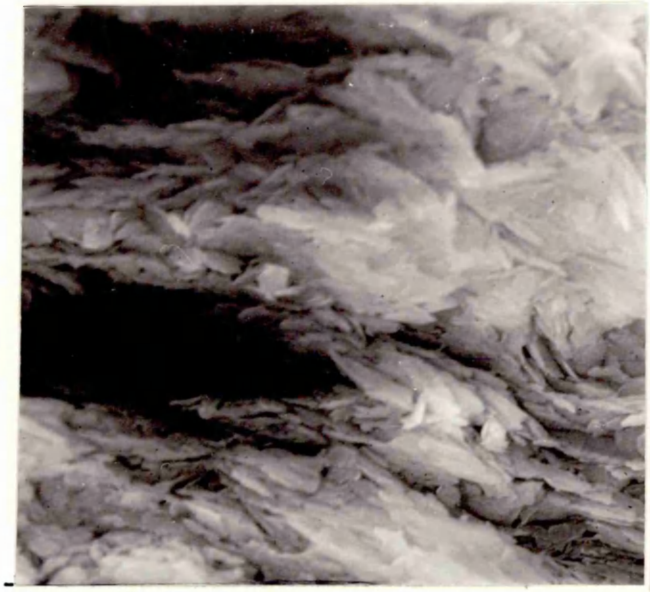


Fig 63

ILLITE + aspartic acid

2 μm

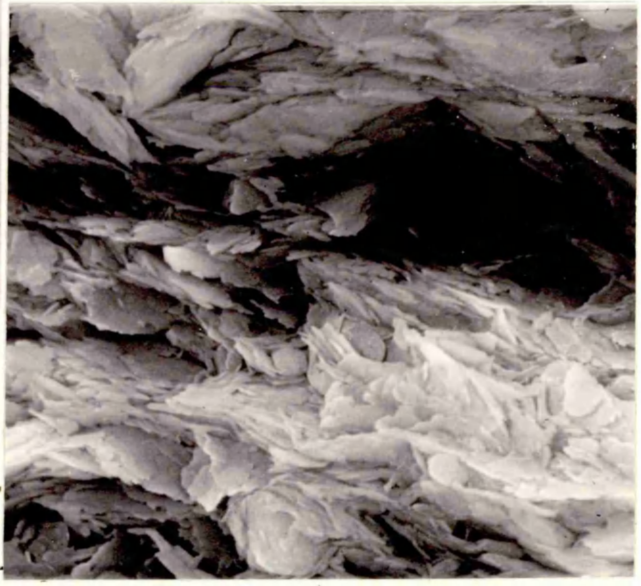


Fig 64

FIG NO	SAMPLE MEAN DIRECTION	ALLOWED DEVIATION	POPULATION MEAN DIRECTION
45	117	± 8	118
46	112	± 8	118
51	168	± 10	175
52	165	± 10	175
53	167	± 18	179
54	168	± 17	179
55	165	± 12	172
56	161	± 14	172
57	165	± 10	170
58	169	± 13	170
59	171	± 21	155
60	160	± 27	155
61	172	± 10	180
62	169	± 8	180*
63	164	± 8	165
64	163	± 9	165

*Sample mean falls outside allowed deviation

TABLE 13 CONFIDENCE INTERVALS TEST ON DATA SHOWING PREFERRED PARTICLE ORIENTATION

FIG NOs	R	K	R1 + R2	2K(R1 + R2 - R)
45, 46	85.08	3.77	85.41	2.47
47, 48	24.07	0.50	25.88	1.82
49, 50	9.22	0.18	12.44	1.19
51, 52	80.54	2.98	80.65	0.68
53, 54	68.05	1.91	68.05	0.00
55, 54	71.33	2.12	71.46	0.56
57, 58	75.65	2.39	75.81	0.73
59, 60	45.11	1.02	46.02	1.84
61, 62	81.77	3.08	81.78	0.03
63, 64	80.92	2.87	80.99	0.38

TABLE 14 WATSON AND WILLIAMS TWO SAMPLE TEST FOR LABORATORY CLAYS

organic tests for kaolinite suggests that the degree of preferred orientation is influenced by the organic compound used.

With the illite sediments there appares to be no substantial difference between the particle orientations of the various specimens. The saltwater sediment shows a lower degree of orientation but the particles are not significantly random.

Although the specimens did not have a fissile character it was assumed that the plane normal to the direction of loading was equivalent to the fissile plane. Table 13 gives the results of the confidence interval test for each specimen. Except in one instance the preferred orientation of particles lies within the 95% confidence limits of the 'fissile' plane.

A check for similarity of orientation between micrograph pairs of the same specimen was carried out using Watson's two-sample test. The results are presented in Table 14 and they confirm that orientation was indeed consistent within specimens.

6.4.1 Statistical conclusions

A histogram of $\bar{R}$ values from SEM micrographs of the laboratory sediments is plotted in Fig. 65. The histogram shows that the data falls into two distinct groupings. A random orientation of particles is suggested for samples whose $\bar{R}$ value is less than 0.4, and this group corresponds to sediments deposited in the marine (flocculating) environment.

The freshwater sediments and those sediments for which organic compounds were added to the saltwater give values of $\bar{R}$ in excess of 0.5, indicating a preferred orientation is present. It may be concluded that, in the absence of a flocculating agent (i.e. in freshwater) or where the potential for flocculation is neutralized (i.e. organic matter present), there is an increased chance of clay particles in the sediment assuming a preferred orientation.

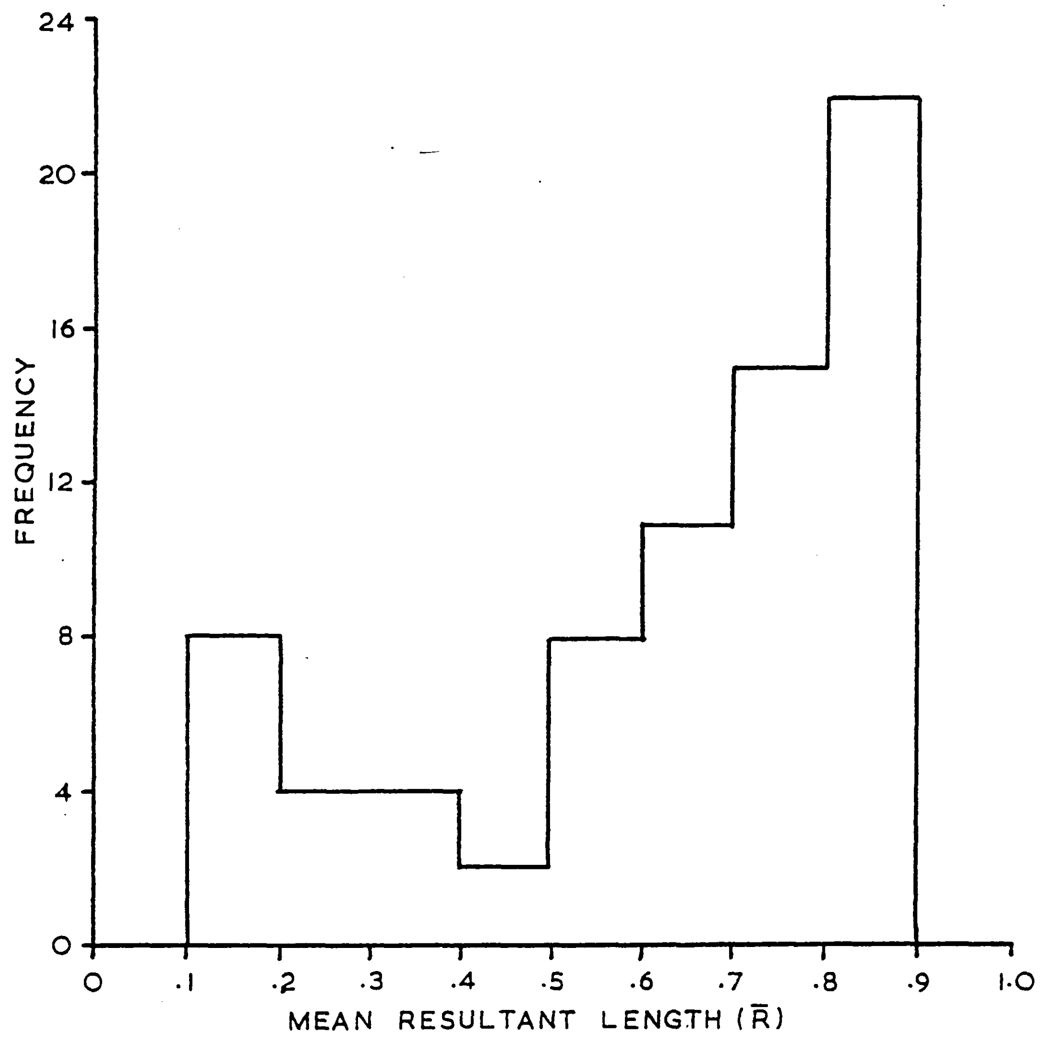


Figure 65 ORIENTATION ANALYSIS RESULTS OF LABORATORY SEDIMENTS

CHAPTER 7

DISCUSSION

7.1 Introduction

The variation in the microstructure of clay-water systems, from the initial colloidal state to the final compacted sediment, has been reviewed in Chapter 3. The evidence presented demonstrates that significant changes occur as the water content of the system decreases. Three phases are recognised. Within each phase there is a characteristic microstructure which imparts distinctive geotechnical properties.

During the primary sedimenting phase the extreme high water content enables the system to behave as a viscous fluid. In a marine environment the diffuse double layers on the clay mineral surfaces are depressed and this allows particles to flocculate. Floccule formation and size are enhanced by high particle concentration and agitation in turbulent waters. In the absence of natural flocculation clay particles may be aggregated into fecal pellets by free swimming organisms. This stage is characterised by single-plate cardhouse type structures and small domain-like clusters. The latter are possibly caused by pelletization.

The sedimenting phase terminates at the mud-line where there is a sudden increase in particle concentration and hence floccule formation. The water content remains sufficiently high to provide layers of water which become sandwiched between the solids. Domains are increasingly formed during this next phase and single particles become a rarity. The overall structure is that of an open random arrangement in which the void spaces are still the major proportion of the total sediment volume. However at this stage the system loses its fluid nature and behaves more as a plastic material.

Further water loss results in a slow progression towards a semi-solid system in which the volume of sediment decreases in proportion to the volume of water expelled. As a consequence more interparticle linkages are formed and the system takes on a rigidity that is able to resist external disturbance. Under sufficient pressure a partial collapse of the open structure is inevitable and an increase in the parrallelism of particles is produced. Plasticity remains a distinctive feature of the sediment during most of this stage but increased burial eventually produces a more brittle material as the sediment alters from a semi-solid to a solid. Concurrent with these physical changes are the chemical alterations associated with diagenetic processes.

During the stages outlined above the microstructure of the clay sediment continuously changes as more domains are formed and particle reorientation occurs. These phenomena are accompanied by water migration, reduction in sediment volume and porosity, and an increase in sediment density, which all contribute to give a variation in the geotechnical properties.

7.2 Measurement and analysis of microstructure

Assessment of mudrock microstructure has in the main been of a qualitative rather than a quantitative nature, for example Barden (1972). However the ability to quantify microstructure is essential if a comparison between sediments is to be established. With optical microscopy this was achieved by taking advantage of the birefrigent properties of clay minerals (Ingram 1953). X-ray diffraction methods have also been used (Odom 1967). The main drawback of both techniques lies with the inability to observe the individual particles on which the measurement is based. There is no means of knowing if the results are due to the orientation of a few large particles or many small ones. The electron microscope overcomes this problem since what is 'seen' can be measured. Unfortunately, depending upon the angle that the specimen makes with the electron beam, there is some distortion of the image due to a foreshortening effect. This has been discussed in detail by Tovey (1970). Specimens in this study were all viewed at similar angles and hence the effect of foreshortening was reasonably consistent and could be ignored.

There are several methods of quantifying the soil structure revealed by SEM micrographs. Tovey (1971) describes two optical techniques - one utilising an optical diffractometer, the other a convolution square camera. Both methods derive a pattern or image, from the micrograph, which is measured to give a value of preferred orientation. Koff et al (1973) developed an electronic device to automatically scan a micrograph and provide a measure of the orientation directions of the fabric elements. Whilst these methods are able to give a rapid assessment of orientation they require specialist equipment which is not always available.

The writer has adopted a simple approach which utilizes the capabilities of a computer aided design package called DOGS.

Although the system has been designed for drawing office use it has several facilities useful for the extraction of data from photographs. Other than the angular measurement of particles, as required for this study, the system can also provide linear and areal dimensions convenient for volumetric determination. The major drawback of the method is that it is manual and therefore time consuming. Each particle measured has to be randomly chosen to eliminate sampling bias. However, once a micrograph has been 'mapped' in this way the extraction of data is rapid. The convenience of using the method for angular measurement relies on the ability to determine the shape of a particle. The measurement of elongated particles presents no problem but when the sample is contaminated with directionless particles the task becomes more difficult.

The measurement of particle orientation from single SEM micrographs is of course a two-dimensional approach to what is realistically a three-dimensional problem. Although the sense of direction in the plane of the micrograph can be determined, the dip of a particle cannot be measured. This requires stereo interpretation and an application of photogrammetric techniques to quantify the information. Such a rigorous approach is mainly applicable to micrographs showing very open structures and it was felt that the additional work involved was not warranted when the majority of the specimens presented a compact microstructure.

The statistical analysis of orientation first involves finding the direction of preferred orientation and the method is discussed above and reported in full in Appendix III. The next step entails measuring the strength and significance of the preference. A summary of various statistical methods is given by Smart (1973). Two such methods - the Gap Test and the Chi-Square Test - have been applied by Gipson (1965). In all cases an appropriate mathematical model is found to describe the observations. Where data is in the form of angles then circular and spherical distributions are applicable. The mathematics of such distributions are discussed at length by Mardia (1972). For this study the recommended circular model is the Von Mises distribution.

7.3 Mudrock microstructure

There have been few attempts to correlate mudrock microstructure with the environment of deposition despite of a longtime awareness that a relationship exists between these two variables. Early hints at the importance of the environment were reported by Keller (1946) and Krumbein (1947). A link between the breaking characteristics of mudrocks and the arrangement of its clay particles was established by Ingram (1953). He also noted the connection between organic matter and the increased parallelism of clay particles. It may be inferred from Ingram's work that the arrangement of clay particles is influenced by the organic matter in the depositional environment. White (1961) overlooked the importance of organic matter but was able to establish a link between the chemistry of the environment at the time of deposition and a preferred orientation of clay minerals which caused fissility. White (loc cit) used an X-ray diffraction method to determine the orientation but did not report any quantitative data.

Early accounts of quantifying particle orientation are given by Gipson (1965, 1966) and he established relationships between orientation, depth of burial, porosity, clay mineralogy and grain size. Although Gipson considered that the physico-chemical conditions in the depositional environment influenced the original orientation of particles within a sediment, he failed to take this into account in his correlation exercise.

Odom (1967) reflected upon the affinity between particle orientation and environment in more detail. A positive correlation was established between clay particle orientation and organic carbon content of the sediment. He concluded that variations in fabric, not attributable to benthic disturbance or tectonic deformation, were due to the physico-chemical conditions in the depositional environment. These environments were not specifically identified. The sediments investigated were of Pennsylvanian age from the Illinois Basin, which closely resemble the UK Coal Measures and thus the types of environment can be inferred. Odom (loc cit) found that in general the degree of clay particle orientation increased with increasing organic content. If it is assumed that the organic content measured in the rock is in proportion to that in the water at the time deposition then a correlation between mudrock microstructure and the environment of deposition is established. Environments with a high organic content are more likely to produce a strong preferred orientation.

The microstructure of various mudrocks has been observed during the course of this study. The mudrocks were chosen partly because of the convenience in obtaining a sample but more importantly with the intent that they were to be representative of a wide spectrum of depositional environments. Although more mudrocks were collected and analysed than is presented in Chapter 5, those that are shown were chosen as typical for each environment. Because the physical and chemical conditions possible at the time of deposition for each mudrock are complex the true nature of the environment is not known for certain. The writer has thus used associated relevant literature when required in order to provide the necessary information. However, where possible the samples were chosen from 'type areas' and the assessment of the depositional environment is likely to be more valid.

The environments have been divided into three major types, oxygen rich (aerobic), oxygen poor (anaerobic) and absence of oxygen (anoxic). Mudrock samples from each of these environments (and also spanning geological time) have been investigated and the observations show that the preferred orientation of particles within the samples improves with a decrease in the presumed oxygen content of the environment of deposition. The writer suggests that as a result of the decrease in oxygen there is more chance of organic matter being preserved and this leads to an increase in the organic content at the sediment-water boundary. The dissolution of the organic matter provides the environment in which the clay particles are dispersed and the probability of there being a preferred particle orientation in the sediment increases.

It would appear from the correlation of depositional environment and clay particle orientation given in Chapter 5 that there is some justification for accepting the model proposed in Chapter 4. The model is not presented as being the only cause of preferred particle orientation in compacted mudrocks since the process of compaction itself can induce a certain degree of orientation. For fresh sediments, where compaction is minimal, the model is certainly valid. Microstructure considered as primary fabric (O'Brien 1987) most probably owes its origin to the characteristics of the environment of deposition in the manner described by the model.

7.4 Clay microstructure

In this section the microstructure of the laboratory sediments are considered. The limitations of the laboratory set-up have not allowed the writer to simulate all conditions within a depositional environment. The experimental programme was devised so that the model proposed in Chapter 4 could be tested. The model suggests that organic compounds present in the water at the time of deposition would cause a dispersion of the clay particles settling within the water column. The probable organic compounds are amino acids, fatty acids and sugars since these are present in both the overlying waters and the upper layers of sediment. To test the model the following compounds were chosen - aspartic acid, palmitic acid and glucose. These are representative of amino acids, fatty acids and sugars respectively.

Monomineralic powders of kaolinite and illite were used as the source clay sediment and these were mixed with water and then allowed to settle under different chemical conditions. The use of pure clays rather than naturally occurring sediments is questionable but the writer considered that it was important to test the model without the influence of too many variables.

From the limited number of tests carried out it is evident that organic compounds are able to influence the orientation of particles. The typical value of mean resultant length ($\bar{R}$) of kaolinite particles sedimented under marine conditions is 0.1 which shows the particles to be randomly orientated. When organic compounds are added to the same mix the value of $\bar{R}$ rises to about 0.8, indicating that the particles have taken on a preferred orientation. It is noted that glucose does not have as much effect as the other two organic compounds. The reason for this may be due to less adsorption of the glucose by the clay mineral. Without the neutralizing effect on the mineral surface, flocculation would still occur and the ability to produce a preferred orientation reduces.

The tests using illite were not as conclusive since under flocculating conditions the resulting particle orientation in the samples was not significantly random. This is somewhat surprising since it is expected that the smaller a clay mineral the more readily it reacts to changes in its environment. These findings appear to contradict the work of others (Sides and Barden, 1971). It may be that the sampling or preparation technique affected the results but since all samples of the flocculated illite gave similar orientation values this is considered unlikely. A more plausible explanation relates to the thickness of the flocculated sediment after consolidation. The thickness was unexpectedly low (15 mm) in test no. 6 and it is probable that the drainage effects reported by Knill et al (1976) encroached within the central zone where the sample was taken from. Tovey (1973a) has shown that in a consolidation test to only 64 kPa the drainage effect penetrated at least 4 mm from the drainage surface. At the higher pressures used in this study it is feasible that the 'disturbed' zone extends deeper.

Notwithstanding the anomaly of the salt flocculated illite, the tests (10 and 11) using palmitic and aspartic acid as the dispersive compounds produced clays with a well orientated microstructure as opposed to the expected random one from salt flocculation. This strengthens the conclusion reached for the kaolinite samples that these organic compounds do influence the orientation of clay particles within a sediment.

7.5 Comparison of microstructure in the natural and laboratory sediments

The analysis of clay particle orientation in both the natural and laboratory sediments has revealed that in each case the orientation displays a measurable random and preferred direction. For the natural sediments the measured value of preferred orientation correlates with the assumed conditions at the time of deposition. In the laboratory tests certain aspects of these conditions were duplicated and the measurements confirm that particle orientation is modified when organic compounds are introduced into the clay-water system.

A measure of the degree of preferred particle orientation in a specimen is shown to be given by the mean resultant length ($\bar{R}$) of its angular data. Histograms of the frequency of particular $\bar{R}$ values for both sediments have been presented. These histograms are combined in Fig. 66. It is noted that the frequency peaks are shifted in favour of higher values of $\bar{R}$ being recorded for the natural sediments. It is the writer's opinion that this shift is attributed to an increase in parallelism caused by a greater consolidation pressure being experienced in the natural environment. Since the displacement of the peaks shown in Fig. 66 is relatively minor it is concluded that reorientation of particles is not significant at pressures greater than the maximum laboratory pressures of 1000 kPa. If reorientation above this pressure was significant it is believed that a greater displacement would have been observed.

7.6 Geotechnical properties of clay sediments

It is generally accepted that the geotechnical properties of clay sediments are a reflection of their structure, mineral composition and geological history. The structural control may be due to the gross structure, in the form of fissures for example, or the microstructure, as related to the spatial arrangement of the sediment particles. Because the structure of a sediment is affected by many variables the geotechnical properties are likely to be as varied. For this reason there is not always a unique relationship between structure and geotechnical properties. In the following sections on natural sediments the values quoted are typical for the type of sediment in question.

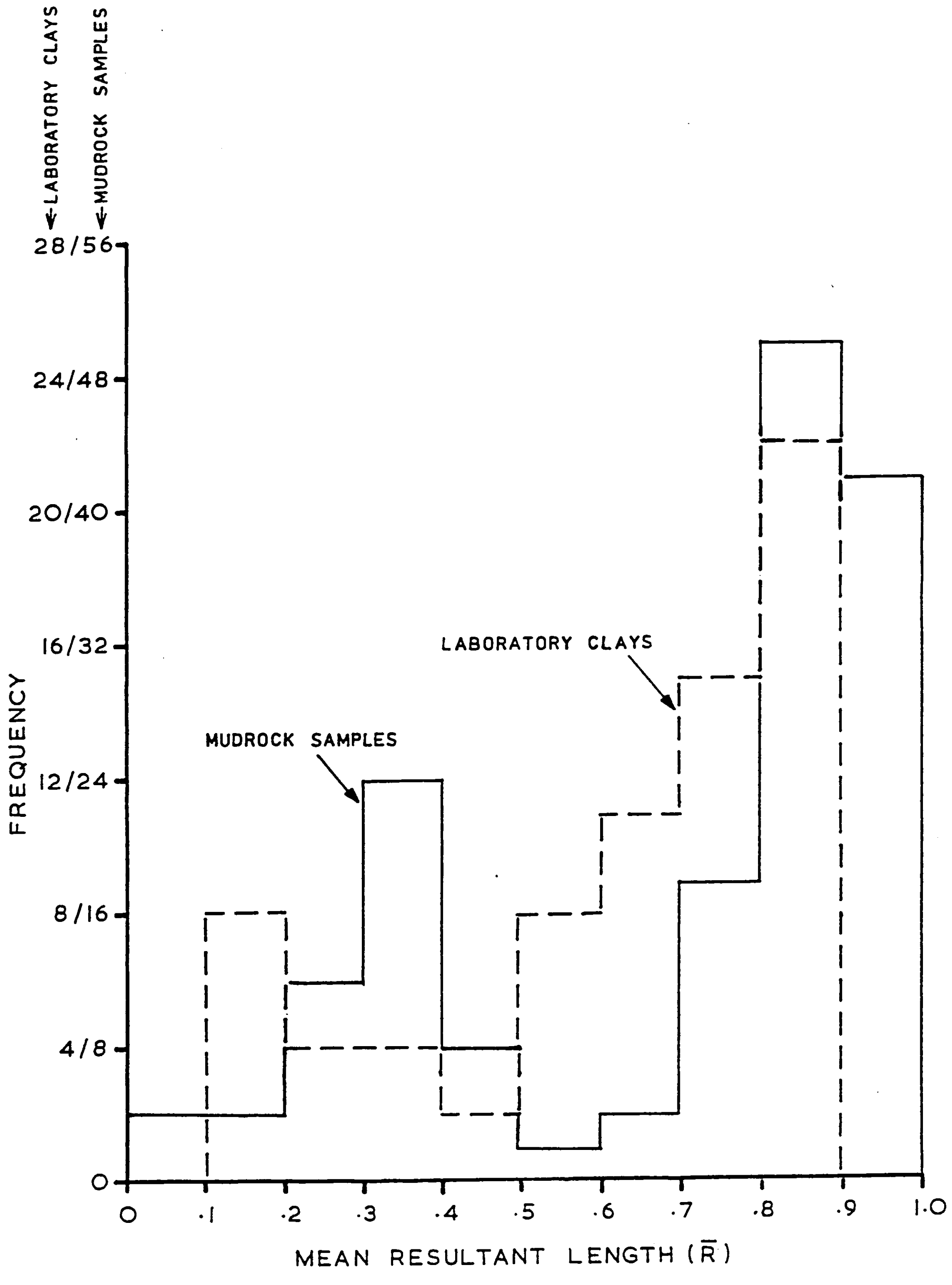


Figure 66 A COMPARISON OF PARTICLE ORIENTATION IN NATURAL AND LABORATORY SEDIMENTS

7.6.1 Fresh clay sediments

Freshly deposited sediments at the mudline are characterised by a high water content (200-450%) corresponding to void ratios in the order of e = 6 to 12. The shear strengths of such sediments are expectedly low, around 1-6 kN/m^2, depending on the sediment density. There is normally a rapid reduction in void ratio within a few centimetres of the mudline, its value falling to about e = 2 to 3. Thereafter the void ratio reduces gradually in response to overburden pressure. A typical void ratio profile for a deltaic sediment is given in Fig. 67. With a reduction in void ratio there is a natural increase in the shear strength associated with the increased density. It should be stressed that the void ratio and shear strength profile are only typical of a normally consolidated sediment. A deviation from these profiles can be expected depending on the rate of deposition and the mineral content and grain size distribution of the sediment.

7.6.2 Consolidated clay sediments

It is not easy to define the changeover from a fresh sediment to a consolidated sediment since the process of consolidation is a gradual one. The sediments referred to in this section are considered to have undergone some diagenetic alteration leading to an interparticle bonding that gives the sediment a true cohesive strength. In general terms such sediments are rocks by definition, but the writer is hesitant to apply such an ambiguous word.

Any attempt to assign typical values to the geotechnical properties of consolidated clay sediments is immediately confronted by the

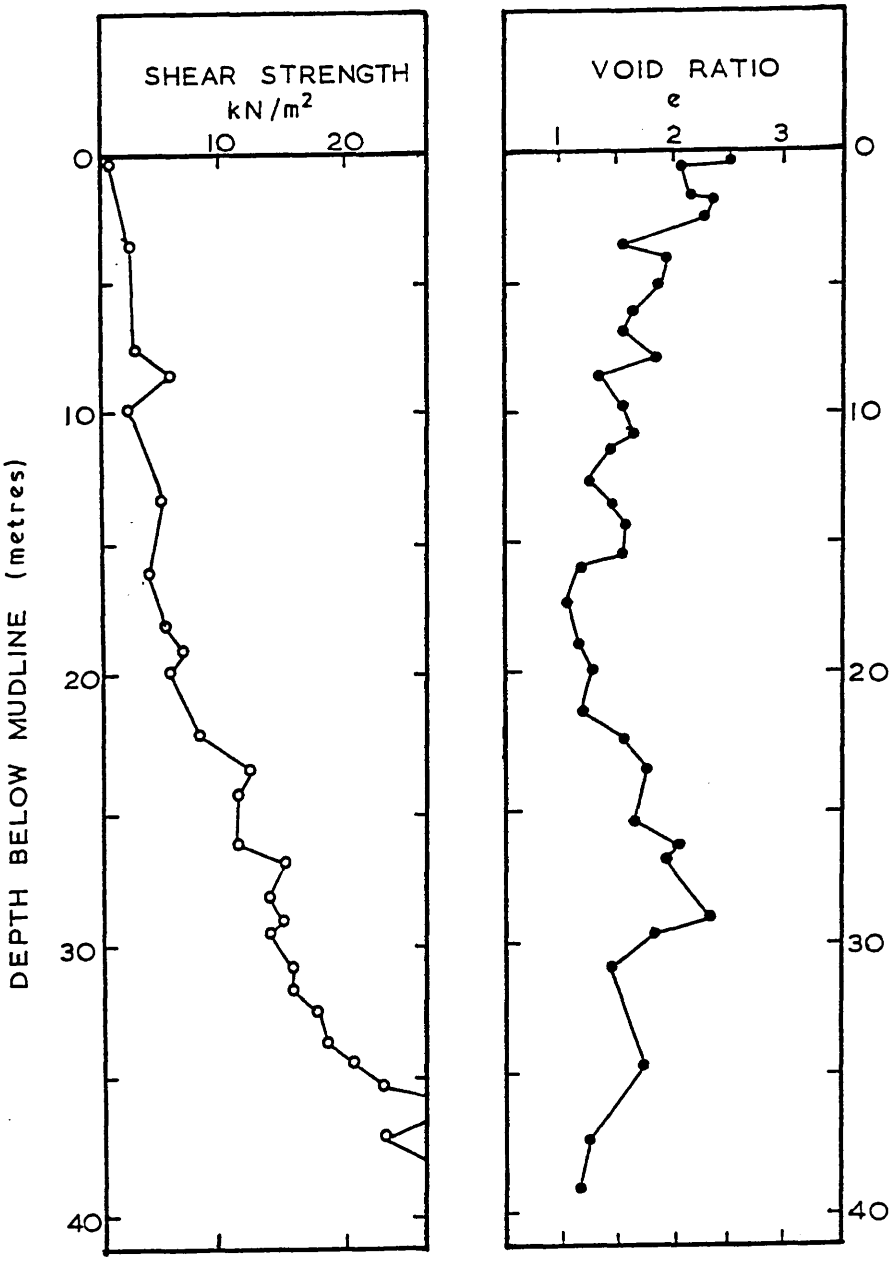


Figure 67 TYPICAL SHEAR STRENGTH AND VOID RATIO VARIATIONS WITH DEPTH IN A DELTAIC SEDIMENT (Bohlke and Bennett 1980)

tremendous variation in measured values. Cripps and Taylor (1981) have conducted a comprehensive study of mudrock properties and concluded that the magnitude of any particular parameter was the product of a number of genetic factors, including composition, degree of overconsolidation and diagenetic changes. The type and method of testing used to determine the particular parameter also contributed to the variation in its value.

From the data given by Cripps and Taylor (op cit) the variation of void ratio (obtained from the porosity data) and the undrained shear strength of a number of UK mudrocks have been plotted with respect to geological time (Fig. 68). The expected reduction in void ratio commensurate with age and depth of overburden is confirmed. A reverse trend is just discernable for the shear strength values, but is somewhat masked by other variables related to geological history, lithology and amount of weathering.

Consolidation test results have been reported for a number of post-Carboniferous sediments mainly with respect to the assessment of foundation settlement. Average values for the coefficient of consolidation (c_v) lie within the range 0.04 to 5.4 mm^2/min. For Oxford Clay, Jackson and Fookes (1974) quote c_v = 1.0 to 1.3 mm^2/min.

7.6.3 Laboratory sediments

One of the aims of this study has been to consider the variation in certain geotechnical properties with respect to sediment microstructure. The experimental data obtained from consolidation and shear strength tests performed on the various laboratory sediments

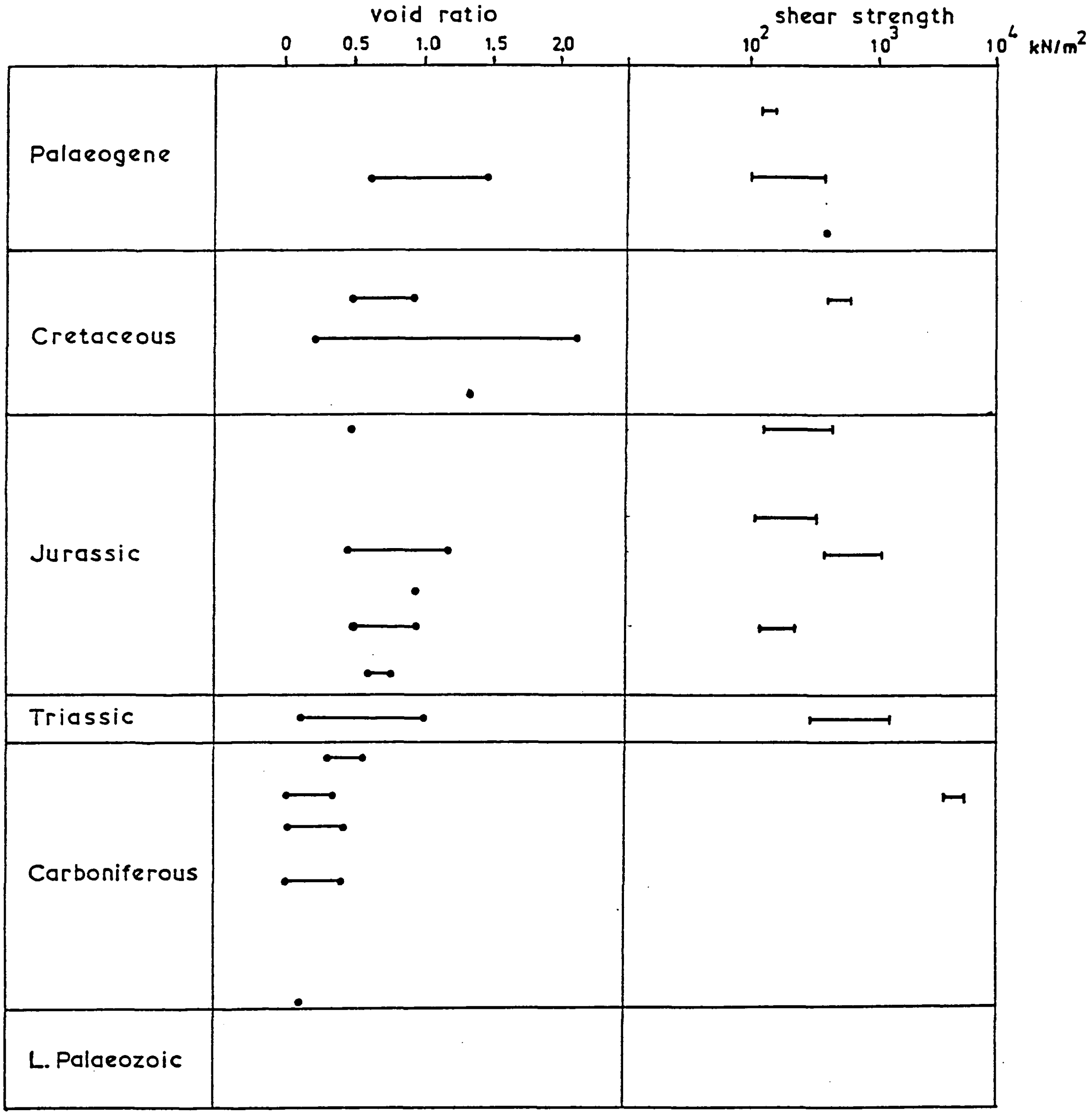


Figure 68 SHEAR STRENGTH AND VOID RATIO VARIATIONS IN UK MUDROCKS

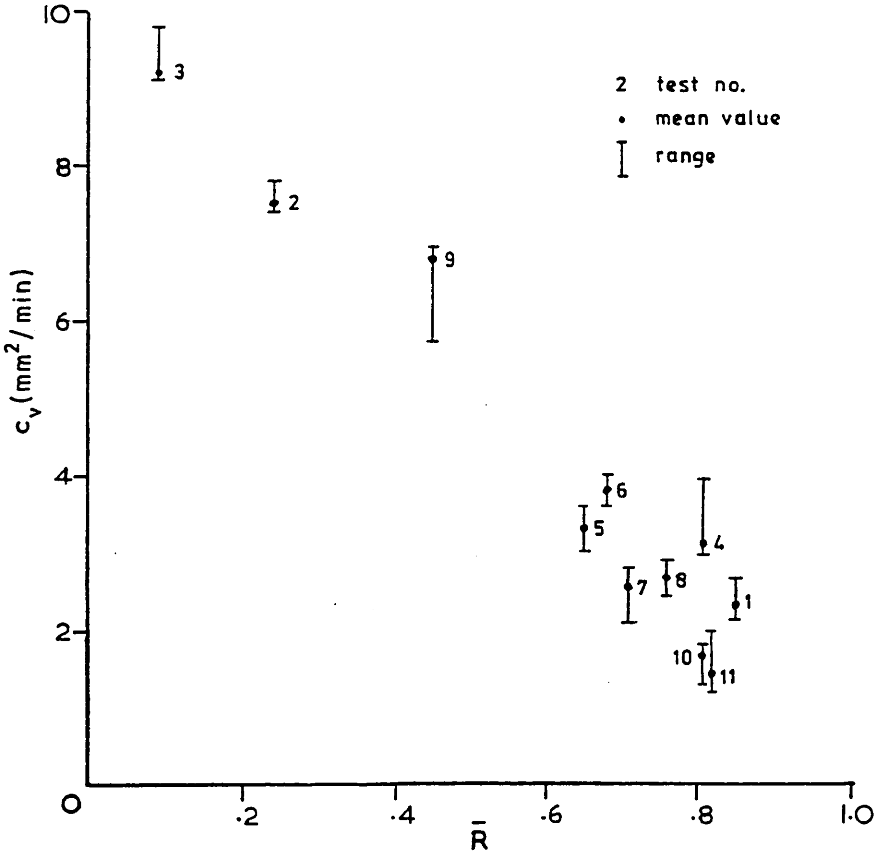


Figure 69 COEFFICIENT OF CONSOLIDATION (c_v) VERSUS MEAN RESULTANT LENGTH

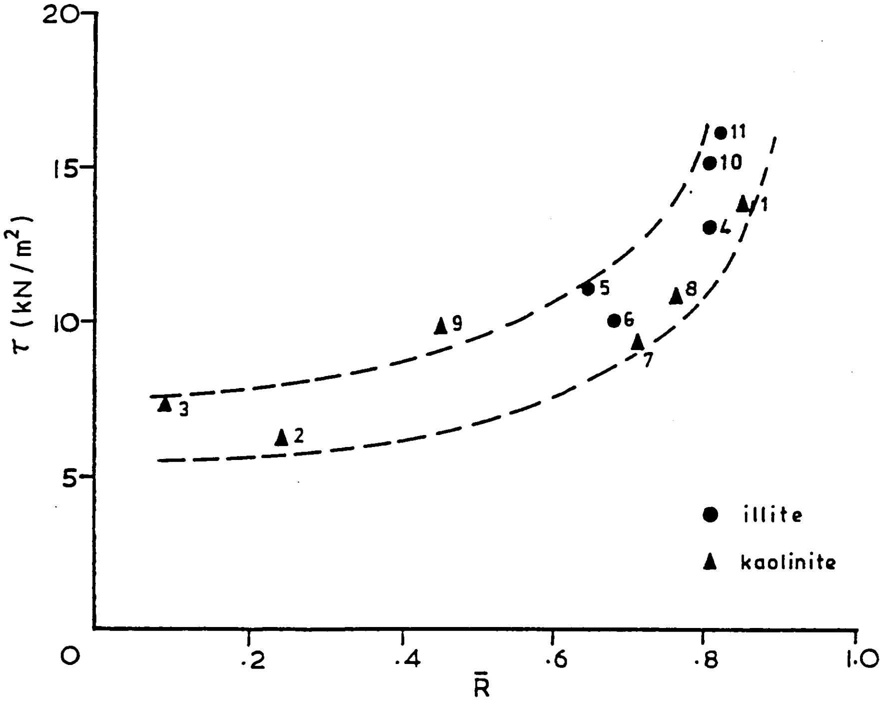


Figure 70 SHEAR STRENGTH (τ) VERSUS MEAN RESULTANT LENGTH

have been presented in Chapter 6 (Table 11). The statistical parameter chosen to describe the microstructure is the mean resultant length ($\bar{R}$) of the measured particle orientations. Values of $\bar{R}$ for each laboratory sediment have been plotted against the corresponding value of coefficient of consolidation and shear strength. These graphs are given in Figs. 69 and 70 respectively.

The laboratory results show that the sediments with randomly orientated particles have marginally higher void ratios, higher values of c_v and lower shear strengths than those with preferred orientation. The void ratio relationship is as expected since a random structure is naturally more open. Correspondingly, the increased water content of the sediment gives a reduction in the shear strength. The value of the coefficient of consolidation is related to the permeability of the sediment. In the preferentially orientated sediment the diffuse double layers on the clay particles are fully developed. These expanded layers contain a more viscous water which constricts the pathways and reduces the flow velocity. In addition the parallel arrangement of particles, at right angles to the flow, creates a longer flow path and hence a reduction in the hydraulic gradient causing fluid movement. These effects combine to give a reduced permeability which leads to a lower c_v in the sediments with preferred orientation.

The microstructure is also shown to be altered by the addition of the particular organic compounds chosen in tests 7 to 11, and consequently the geotechnical properties are modified. For example, amino acids are thought to increase the dielectric constant of the water and this causes an expansion of the diffuse double layer resulting in particle repulsion and a reduction in permeability. The lower value of c_v measured in test 7 (compared with test 3) substantiates this.

The compression indices for the sediments have been determined from their compression curves (e against log p). Example curves for kaolinite and illite are plotted in Fig. 71. It is noted that the sediment compression curves are in general agreement with other published data plotted on the same figure. The value of C_c is slightly reduced for the organic tests in comparison to their equivalent saltwater-only test and this is attributed to the alteration of the electrolyte concentration and the dielectric constant of the water.

An insert to Fig. 71 shows the full compression curve for kaolinite (test no. 3) and reveals that most of the reduction in void ratio occurs at a pressure below 50 kPa. This was true for all the tests regardless of the clay mineral type or depositional conditions. This would suggest that the initial structures formed within the sediment undergo similar alteration during the early stages of consolidation. McConnachie (1971) found a distinct change occurs in the consolidation process at about 15 kPa, which may account for the sudden deviation in the full compression curve referred to above. Furthermore, Bowles et al (1969) noted a very distinct difference of microstructure in their marine sediment when consolidated to 50 kPa and 400 kPa. It is thus concluded that most particle rearrangement occurs at low pressure (< 50 kPa).

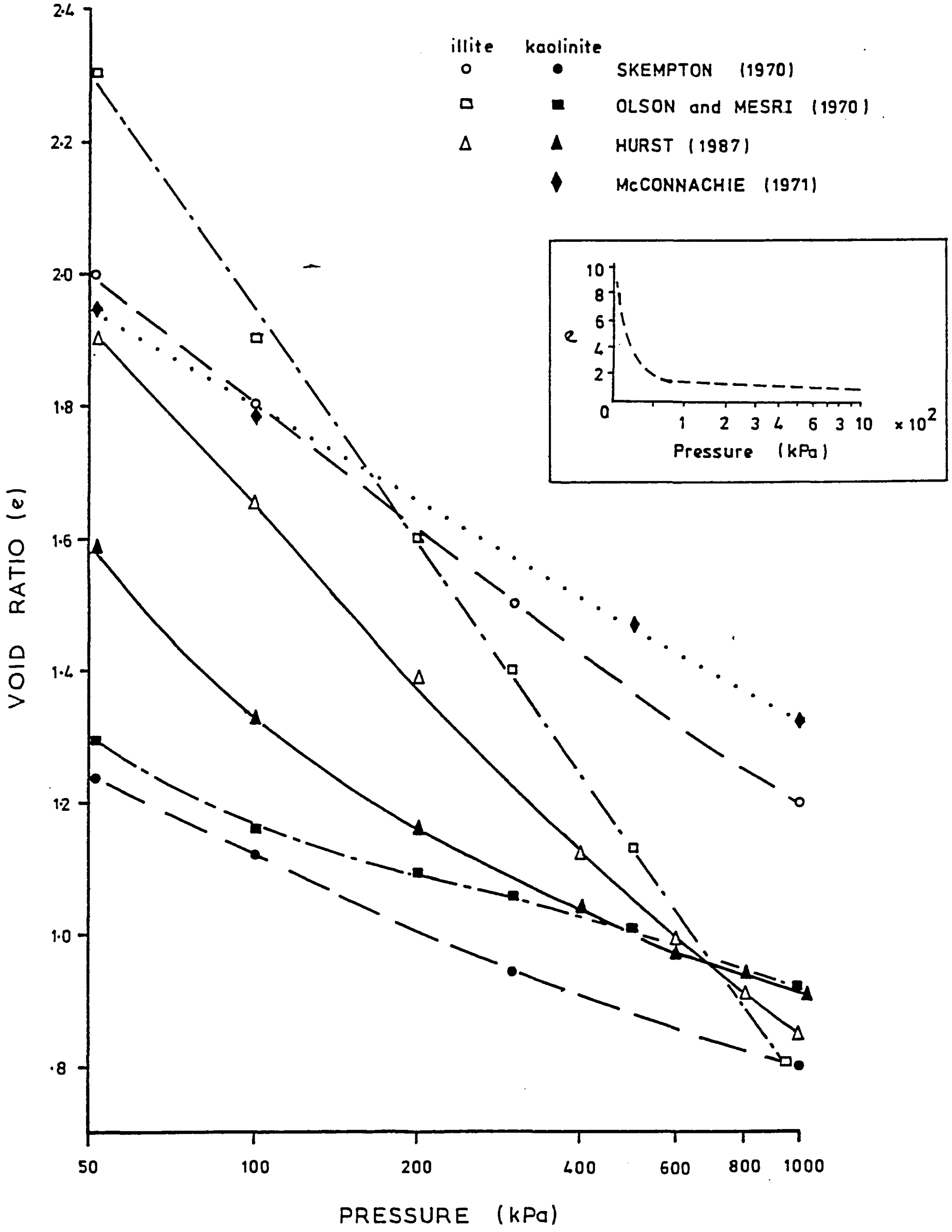


Figure 71 COMPRESSION CURVES OF ILLITE AND KAOLINITE SEDIMENT

7.7 Authenticity of the laboratory simulation

The validity of the various comparisons between natural and laboratory sediments will depend upon how near the laboratory model is to the true geological situation. The model was chosen to represent a body of water in which a specific volume of clay particles is being sedimented under quiescent conditions and thereafter consolidated by gravitational compaction. The basic arrangement of the model to simulate those physical aspects consists of a settling column with a consolidation cell attached to its base. The model is fully described in Appendix I.

Clay particles sedimented into the column remain in suspension until the gravitational force is significant to cause downward motion. The concentration of particles is small enough to allow free settling through the water column. When sufficient particles have arrived at the column base a bed begins to form which defines the sediment-water interface. The growth of this bed is determined by the balance between new particles arriving and the consolidation of the existing sediment. The model is thus a reasonable attempt at simulating the natural deposition of clay particles in a low energy environment.

The clay sediment formed in the consolidation cell is characterised by a high moisture content corresponding to its high void ratio. The model allows for the expulsion of this water by providing a drainage outlet at the top of the cell. Vertical drainage is likely in the natural situation. The process of sediment consolidation is modelled by the standard laboratory procedure. The model differs from true geological consolidation in several respects. The pressure on the sample (simulating the overburden) is increased in steps, each value being held constant until the associated increase in pore pressure has dissipated. This is contrary to the field situation since in most cases the slow rate of sediment accumulation would not cause pore pressures to increase. The stepped pressure increments in the model produce a 'shock' load rarely experienced by natural sediments. More importantly all time-dependent effects, such as inter-particle bonding, cannot be simulated in the laboratory.

The chemical and mineralogical changes experienced by clay minerals during diagenesis are quite varied. Many of the changes take place at pressures and temperatures beyond the capability of the chosen model. However since the depositional environment rather than diagenetic effects are being modelled these shortcomings are considered acceptable. The main requirement of the model is to provide a means of allowing the particles in suspension to be acted upon by the environment in which they settle, since this is considered fundamentally important to the microstructure of the resulting sediment.

7.8 Clay diagenesis, organic matter and oil generation in argillaceous rocks

It was suggested in Chapter 1 that the model proposed by the writer is relevant to the formation of petroleum. It is almost universally agreed that petroleum is generated within organic-rich sediments comprising organic matter derived from biological materials. In section 4.3.3 the writer has highlighted the most probable environments in which organic-rich sediment accumulates. Fundamental to the formation of petroleum are diagenetic changes in the sediment and the interaction between clay minerals and organic matter. The following sections outline these processes and describe the generation of oil in the context of the writer's model.

7.8.1 Clay sediment diagenesis

After deposition argillaceous sediments undergo important physical and mineralogical changes with respect to time. One consequence of these changes is that the clay mineralogy of a sediment bears a close relationship to its geological age. It is observed that the younger argillaceous sediments are enriched in kaolinite, and the expandable mixed-layer clays, whereas the older sediments are almost exclusively rich in illite and chlorite (Weaver 1967, Shaw 1981). The variation of clay mineralogy with age is primarily a reflection of the effects of burial diagenesis. The changes associated with diagenesis have been reviewed by Dunoyer de Segonzac (1970) and are essentially dependent on pore-water chemistry and burial temperature, although the length of time the sediment is buried may also be important.

The first stage of burial diagenesis involves the removal of pore-water from the sediment. This is achieved ostensibly by gravitational compaction due to the increasing overburden as the sediment thickens. It is usually limited to the first few hundred metres of burial. If the expulsion of the pore-water is hindered by poor drainage a state of compacted disequilibrium occurs wherby the pore pressures are abnormally high and the resulting sediment is under-compacted. The experimental data given in Table 11 suggests that one effect of adding organic compounds to the clay-water system is to cause a reduction in the coefficient of consolidation and compression index values. It is therefore probable that organic-rich clay sediments are slower to consolidate than other clay sediments. This being the case then a rapid rate of deposition, for example in a deltaic environment, is likely to lead to an under-compacted mudrock. According to Magara (1975) in such a situation high subsurface temperatures are experienced which means that the temperature dependent clay mineral reactions may take place at shallow depths. This may be significant since the essential mechanism for petroleum formation is that organic compounds in the sediment undergo thermal cracking assisted by the catalytic effect of the clay mineral fraction.

One aspect of further burial diagenesis is the conversion of smectites to illites and this is accompanied by the release of interlayer water. Powers (1967) suggests that the water released from the interlayers may flush out the already formed hydrocarbons from the sediment, thus providing the mechanism by which the hydrocarbons are transferred to give a reservoir rock. Of course if the proportion of smectite in the original sediment is small then the hydrocarbons would remain the the source rock unless another migratory mechanism becomes operative. One such mechanism described by Perry and Hower (1972) is linked to ordered illite/smectite interlayering at greater depths.

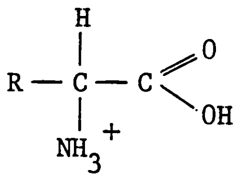
7.8.2. Clay-organic interaction

In order that organic matter is preferentially preserved there must either be an absence of the destructive processes causing its degradation or a means by which the organic matter is screened from attack. It has already been mentioned in Chapter 4 that in the first case reducing conditions usually suffice and in the second case preservation is often achieved by rapid burial. However the writer believes that an important role is played by the reaction between the organic matter and clay minerals.

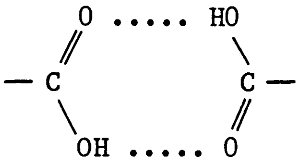
It has been established in section 4.4.4 that clay minerals are able to adsorb organic molecules onto their crystal structure. When this occurs the organic compounds are essentially protected against microbial degradation and therefore are preserved. The writer suggests that this mechanism also has implications for the control of preferred orientation in respect of the proposed model. This may be explained by considering the interaction between amino acids and clay minerals.

The principal source of amino acids found in sediments are plankton proteins which have been hydrolyzed during and after settlement to the sediment surface. The amino acids are present in both the overlying waters and the upper sediment layers. When the two zones are compared it is found that the distribution of amino acids is uneven (Degens 1965). For example, in the overlying water aspartic acid is quite dominant but within the sediment its concentration falls rapidly within the first half metre, perhaps as a result of adsorption onto the clay mineral surfaces.

The capability of amino acids to be adsorbed arises from their structure. By virtue of having both carboxyl (acid) and amino (basic) functional groups in the same structure amino acids are able to exist in one of three forms as determined by the prevailing pH conditions. In acid conditions the preferential form is cationic and may be represented by the following formula:



The amino acid cation (NH_3^+) is thus able to be adsorbed onto the negative clay-solution interface or in the interlayer space depending on proton transfer and cation exchange. When the cation attaches itself to the clay surface one of two effects may happen. The simplest effect is that the surface charge is neutralised by the surrounding swarm of amino acids. In this way flocculation is prevented. A second effect could be that the carboxyl groups of amino acids adsorbed on adjacent particles form a double hydrogen bond as shown here:



The outcome would be that the clay particles are flocculated in a strong face to face mode and such an arrangement could make the development of preferred orientation that much easier. The writer admits that there is no striking visual evidence for this in the micrographs of the amino acid sedimented clays.

The adsorption of fatty acids by clays occurs principally as a result of interaction with broken bond surfaces, in which case it is pH dependent (section 4.4.2). In saline waters, where the diffuse double layer is depressed, the possibility of association is enhanced.

The test results for glucose indicate that its adsorption is weak since there is a marginal effect on the orientation parameter. According to Theng (1974) this is to be expected since the glucose molecule becomes strongly hydrated in aqueous solutions and this weakens its ability to displace water molecules from the clay surface.

7.8.3 Oil generation

The generally accepted source of petroleum precursors is the group of organisms called phytoplankton. When they die these algae drop to the sea floor and are vigorously attacked under aerobic and anaerobic conditions by bacteria. The degradation products are essentially amino acids, sugars and lipids (fatty acids). These organic compounds are adsorbed by clay minerals, falling through the water column and because they are 'removed' from further degradation they become enriched in the sediment. After burial the organic matter undergoes thermal alteration and polymerises into a substance called kerogen. As the burial period and temperature increase the kerogen is progressively transformed resulting in the formation of bitumen and the release of fatty acids. The liberated acids experience decarboxylation, in which clay minerals act as catalysts, to form hydrocarbons and herald the final stage in petroluem formation:- the creation of a source rock.

The type and composition of hydrocarbon produced from the source rock is dependent on the expected form of kerogen. Demaison et al (1983) have related the type of kerogen to early depositional factors. They identified three main environments of deposition of organic matter based upon the degree of oxygenation. In the oxic (aerobic) environment mainly vitrinite (gas-prone) and inertinite (non-source) kerogens are expected. The sub-oxic (dysaerobic) environment produces vitrinite and exinite (moderately oil-prone). In both cases the potential for either oil or gas is reduced by low rates of sedimentation. The anoxic environment is strongly oil-prone with liptinite and exinite being formed. These three environments are analogous to the model in Fig. 14.

The reference to sedimentation rate is pertinent to the observation made in section 7.7.1 concerning the creation of undercompacted mudrocks. Since source rocks for oil must be capable of expelling the hydrocarbons formed, the availability of large volumes of water is necessary to cause primary migration. Apart from the interlayer water source alluded to in section 7.7.1 the other source is apparent in undercompacted sediments. One cause of undercompaction is rapid deposition and this is particularly effective when the flow of water is restricted as is the case for organic-rich sediments.

7.9 Validity of the model

According to the model proposed by the writer oil source rocks should present a stronger preferred orientation than gas source rocks.

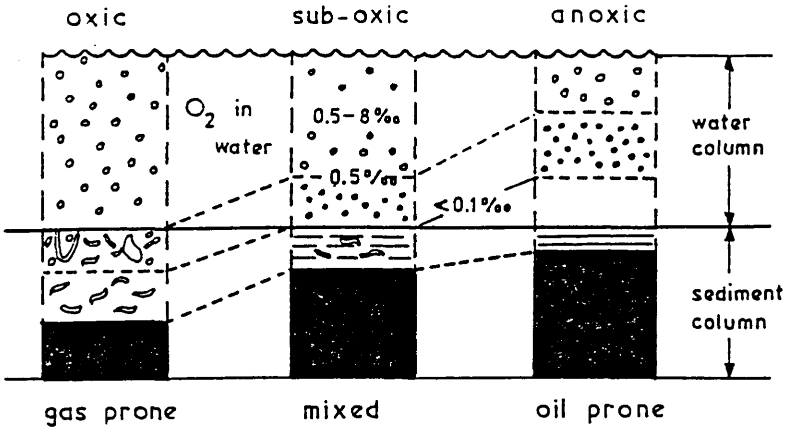
Demaison and Moore (1980) have provided a means by which the model can be tested. They matched the three kerogen type zones to the facies interpretation of the lower Toarcian of Yorkshire by Morris (1979). His division comprises a normal shale, a restricted shale and a bituminous shale. The relationship is shown in Fig. 72.

The normal shale is described as a homogeneous, bioturbated sediment with abundant benthic body-fossils and common sideritic nodules. This facies is equivalent to the gas-prone oxic environment. It is illustrated by the micrograph of Fig 73(a) which on analysis gave an orientation value of $\bar{R}$ = 0.27.

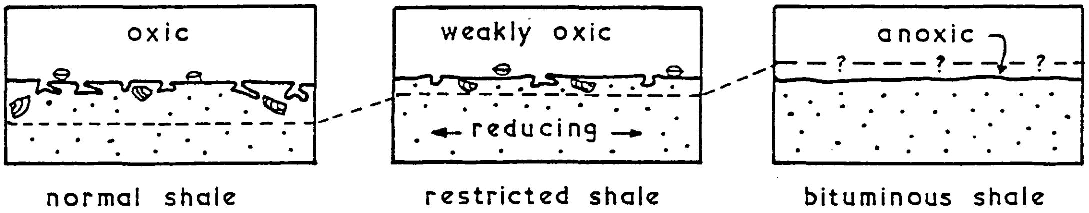
The restricted shale is poorly laminated with scattered calcareous concretions and sparse benthic fauna. It is classed as a mixed organic facies appropriate to the sub-oxic environment. A micrograph of the shale is given in Fig 73(b) and this yielded an orientation value of 0.52.

Pyritic concretions, little bioturbation and a sparse benthic fauna provide evidence that the finely laminated bituminous shale represents the anoxic facies. An orientation ratio of 0.79 was recorded from its micrograph shown by Fig. 73(c).

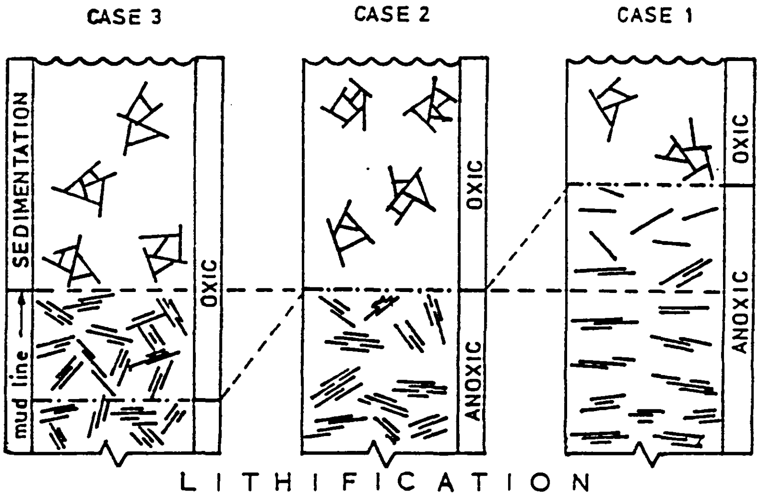
The values for particle orientation strengthen the belief that preferred orientation is related to the conditions in the depositional environment and add weight to the validity of the model.



DEMAISON ET AL (1983)

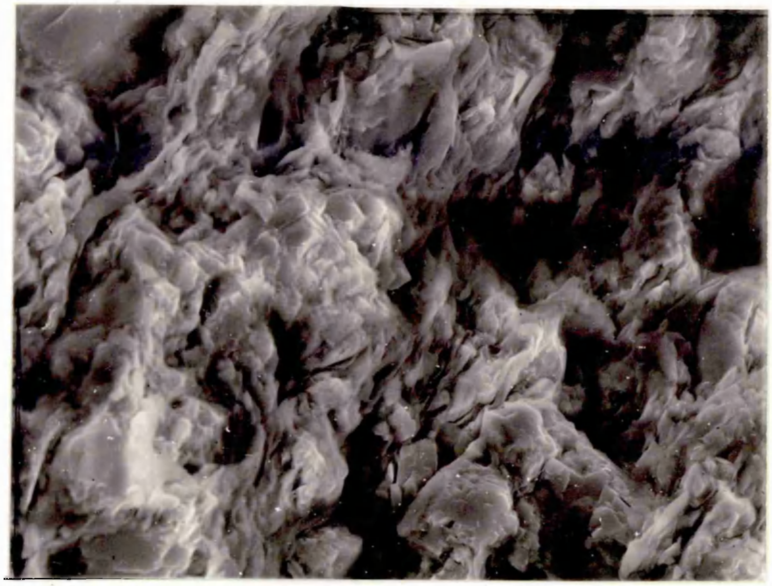


MORRIS (1979)



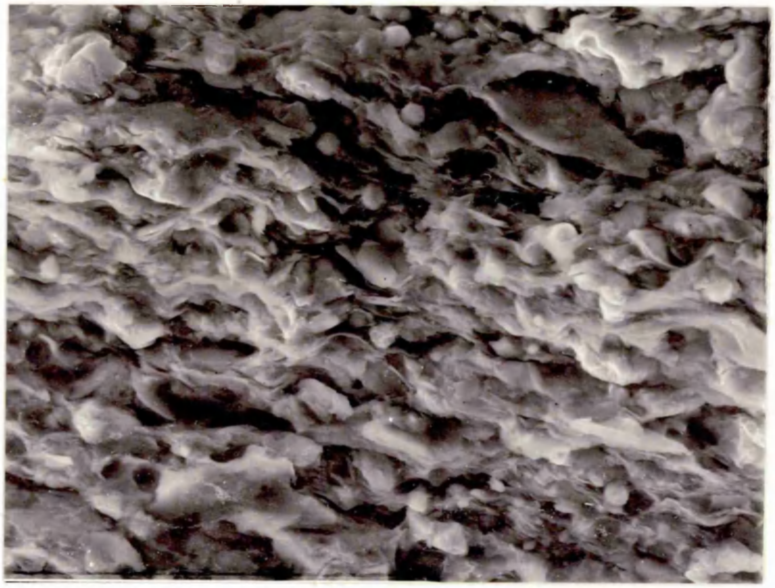
HURST (1987)

Figure 72 COMPARISON OF SEDIMENTARY MODELS



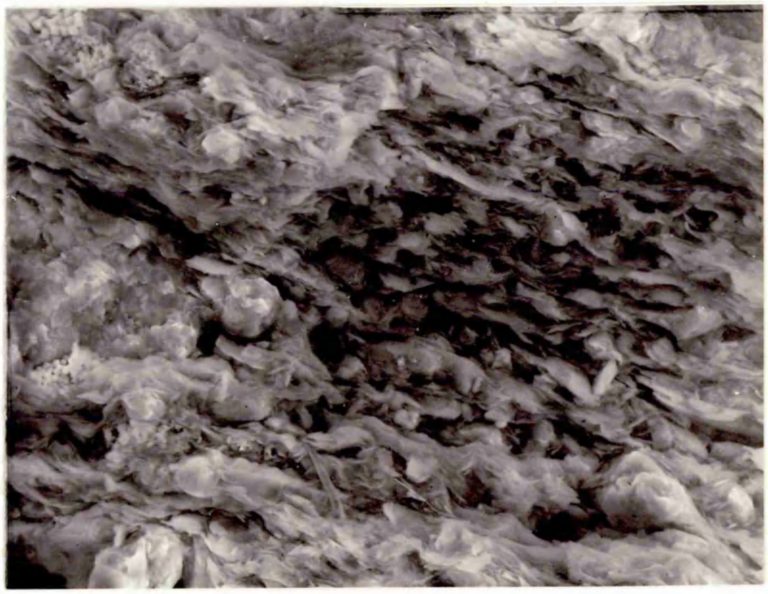
(a)

normal shale



(b)

restricted shale



(c)

bituminous shale

20 µm

Figure 73 SEM microgaphs of the Liassic (lower Toarcian) of Yorkshire

CHAPTER 8

CONCLUSIONS AND SUGGESTIONS FOR FUTURE RESEARCH

8.1. General

This chapter summarises the findings of this investigation into changes in the microstructure of clay sediments and confirms the importance of organic matter in controlling the development of fissility in shales. The last section of the chapter is devoted to suggestions for further research in this field.

8.2. Concluding summary

A simple model of the depositional environment and subsequent consolidation of clay sediments has been used in an investigation into the fabric changes associated with post-depositional effects. Much work has been required in the attempt to simulate the physical and chemical aspects of the model. New pieces of equipment have been built to assist in this investigation. A novel method of creating the correct saline conditions and the use of natural organic deflocculants lend authenticity to the chemical aspects of the model.

Analysis of particle orientations has required the application of directional data statistical methods to evaluate the degree of particle anisotropy in natural and laboratory samples. The writer suggests using the mean resultant length ($\bar{R}$) of the data, when plotted on a unit circle, to provide a measure of the anisotropy index. The value of $\bar{R}$ can be mathematically defined as:

$$\bar{R} = (\bar{C}^2 + \bar{S}^2)^{\frac{1}{2}}$$

where $\bar{C}$ = sum of cosines of angles divided by no of angles and $\bar{S}$ = sum of sines of angles divided by no of angles.

Statistical evidence for preferred orientation is given by comparing the value of $\bar{R}$ with its critical value at a given level of significance. The results from this study indicate that the critical values for proving random orientation could possibly be relaxed slightly. The writer suggests that at the 0.1% level of significance the critical value of $\bar{R}$ appropriate to defining random orientation is 0.45. Preferred orientation would be defined by a value of $\bar{R}$ greater than 0.7. Values in between would indicate a weak preferred orientation. The writer stresses that this suggestion is only appropriate to fabric studies of the type presented here.

8.2.1 Microstructural changes

With respect to the development of preferred particle orientation in mudrocks three important phases of sedimentation are recognised:

(i) colloidal phase
(ii) interface phase
(iii) sediment phase

The colloidal phase exists when the particle concentration in the water medium is very low. During this phase the particles form the structures defined by Val Olphen (1963) as shown in Fig. 4. Both single-plate cardhouse type structures and small domain-like packets are present. The stability of these structures will depend on the pH, Eh and salinity conditions of the water medium.

The sediment-water interface is a distinct zone where the biological activity and chemical reactions of an organic and inorganic nature help to create special conditions. This zone is important because clay particles normally will have a longer residence time here than the period spent settling through the water column. Reactions in which time spent in contact with the overlying water is important are more likely to occur in this zone. The important physical characteristic of the interface phase is that there is a sudden increase in particle concentration. This results in the formation of more domain-type structures at the expense of a diminishing number of single particles. The microfabric tends to be of an open flocculated structure, particularly in the marine environment.

The sediment phase marks the change from the 'soupground' (Ekdale 1985) conditions of the interface zone, where the moisture content is around the liquid limit, to the more plastic conditions associated with a rigid structure. Particles may undergo rearrangement during this phase in response to overburden pressure.

With respect to microstructural changes, the writer suggests that during the colloidal and interface phases the overwhelming influence on preferred particle orientation is the chemistry of the environment. Once the sediment phase is entered this influence is negligible and compaction provides the means of furthering the degree of preferred orientation. The evidence from this study, discussed in section 7.5, indicates that most reorientation must occur at low pressure (<1000 kPa). It is concluded that primary fabric, where the particle orientation is parallel with the bedding, occurs very early in the sediment's history. The hypothesis is supported by this study and the experimental work of Krizek et al (1975).

8.2.2 Geotechnical properties

Test results have shown that the compression-pressure relationship, as defined by the compression index, does not alter significantly with variations in the chemistry of the depositional environment. This would suggest that initial soil structure has negligible effect on the compressibility of clay sediments. However, the coefficient of consolidation is shown to vary with changes in the chemistry and hence soil structure is significant to this parameter. These findings are in agreement with the experimental investigation of Woo et al (1977).

The writer considers it unwise to relate the absolute values of the geotechnical properties of his laboratory sediments to those of natural sediments, since the model neglects the influence of non-clay particles and time-dependent effects. It is thought acceptable, however, that the trends indicated by this work are a reflection of complex natural clay-water-electrolyte mixtures.

8.2.3 Particle orientation

The analysis of mudrock microstructure overwhelmingly points to a relationship between particle orientation and the environment of deposition. It would appear that strong preferred orientation is associated with anoxic environments.

In the laboratory investigations, organic matter has been shown to increase the parallelism of clay particles. The preferred orientation in both cases is in accord with the direction of bedding. The writer concludes that fissility along the bedding planes is related to the environment of deposition and that it is primarily organic matter in this environment which is the cause of fissility under anoxic conditions. Since bioturbational effects were not modelled during this study it is not possible to form a conclusion on this aspect from the experimental evidence. The writer does accept that in the aerobic environment bioturbation will cause disturbance to the primary fabric.

8.3 Suggestions for further work

The writer has attempted to simulate some important factors of the environment of deposition in order to understand the processes of sedimentation and compaction. It has been expressed that the simulation is not perfect. However, the simple approach adopted has provided valid information.

It would be gainful to consider other aspects for the simulation which were beyond the scope of this study. A more realistic approach would be to use natural sediments instead of the monomineralic clays. SEM evidence shows that clay microstructure is affected by silt and sand sized particles and the inclusion of these into the simulation would prove beneficial.

It has been noted that bioturbation destroys primary fabric. This aspect of sedimentation could be modelled and would shed further light on the role played by benthic activity in determining fabric types.

There are many organic compounds present in the environment of deposition. The writer has shown that in his tests at least one of those compounds (glucose) was not very active in promoting preferred orientation. Other potential peptizing compounds, such as glutamine, behenic acid, stearic acid and arginine, could be added individually or as mixtures to the depositional model, to gain further knowledge of clay-organic interaction.

The writer has suggested that the critical period for realignment of particles by external forces occurs at low pressures. Presumably because of the difficulty in preparing undisturbed specimens at a high void ratio, there is less information available on sediments at this early stage of compaction. Methods need to be developed whereby sediment can be compacted within the SEM so that a direct observation of changes to microstructure is attainable.

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EXPERIMENTAL WORK

1.0 Introduction

The experimental phase of the research involved modelling the natural processes of sedimentation and consolidation in a laboratory. The method chosen was to mix dry clay powder in a water medium to form a suspension. The clay was allowed to settle naturally out of suspension into a consolidation cell. Pressure was applied to the cell to simulate the sediment increasing in thickness. The consolidation aspect was monitored for pressure and settlement to determine certain geotechnical parameters. Details of the equipment used and the method of obtaining the sediment are described in the following sections.

1.1 Sedimentation equipment

The model proposed in Section 4.5 suggests that the environment of deposition contributes to the formation of different mudrock fabrics. To test the model a simulation of the process of sedimentation was carried out in the laboratory. Because no existing facilities were available most of the equipment had to be designed and built to suit. The main factor influencing design was the physical process of sediment accumulation. The following elements of the model were fabricated:

1. preparation tank and delivery system
2. sedimentation column

3. consolidation cell.

Preparation tank and delivery system

It was decided to mix together the clay powder and the water prior to delivery into the sedimentation column. A recirculatory system was adopted to achieve a thorough mixing. The system is illustrated in Fig. A1. Adjustment of valves provided switching from mixing mode to delivery mode. An industrial water pump delivered the clay mixture along a plastic tube down to the base of the sedimentation column. Weep holes at the top of the column indicated when to switch off the pump. The tube was then withdrawn.

Sedimentation column

The design of the column was influenced by the need to allow time for settling particles to be acted upon by the 'chemistry' of the waters. For this reason the maximum height of the laboratory was used and this gave a column length of 5 m. The column was constructed from proprietary plastic pipe and pipe fittings. Tapping points were provided at intervals down the column so that samples could be drawn off for turbidity measurement and to allow the column to be drained. Figure A2 shows detail at the base of the column.

Consolidation cell

The consolidation cell was similar to the design of the Rowe cell (Rowe and Barden, 1965) and had internal dimensions of 152 mm in diameter and 120 mm in height. The height was dictated by the need to have a reasonably thick sample at the end of consolidation. The

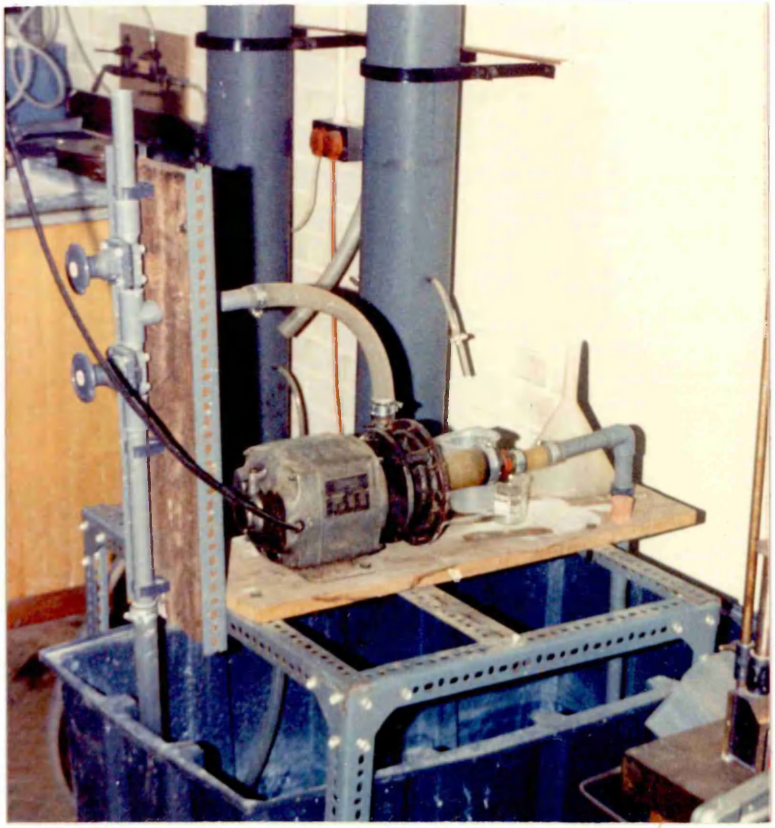


Figure A1 PREPARATION TANK AND DELIVERY SYSTEM

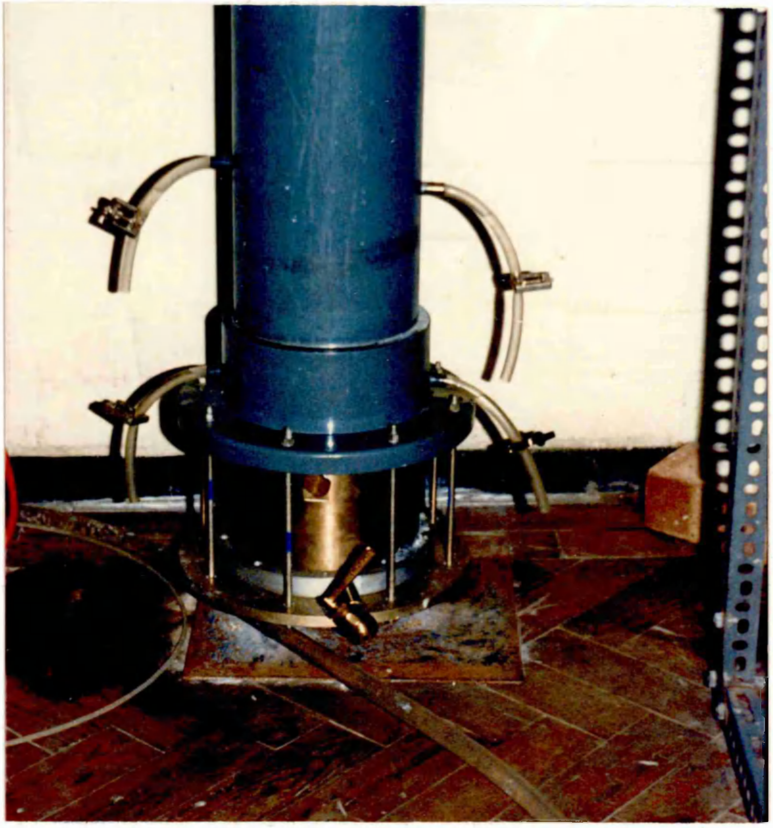


Figure A2 COLUMN BASE DETAIL

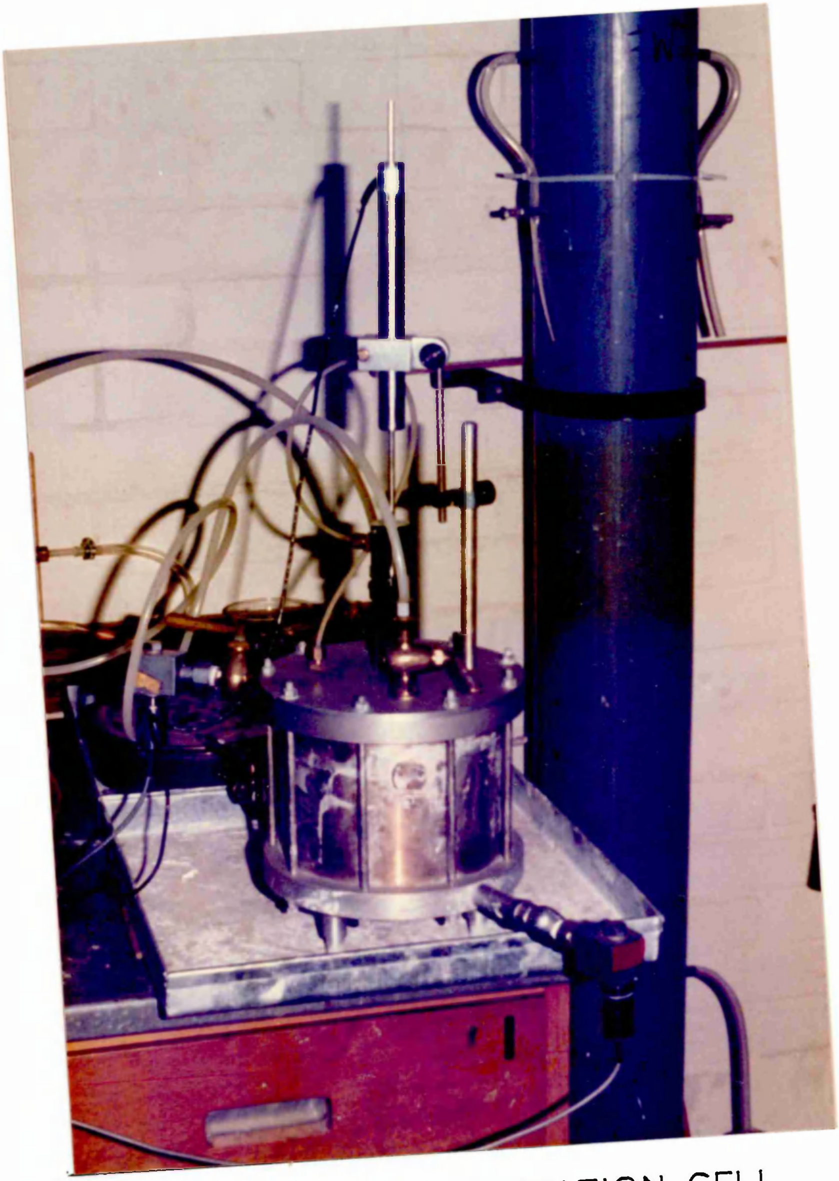


Figure A3 CONSOLIDATION CELL

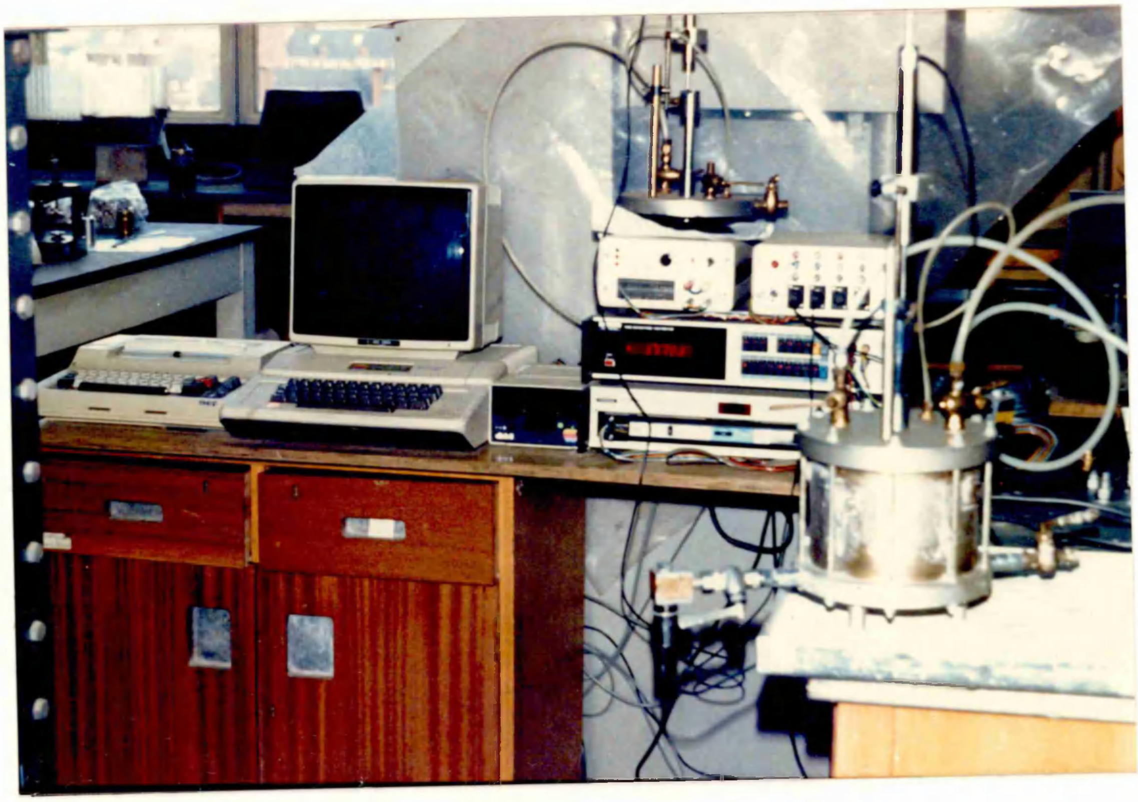


Figure A4 MONITORING EQUIPMENT

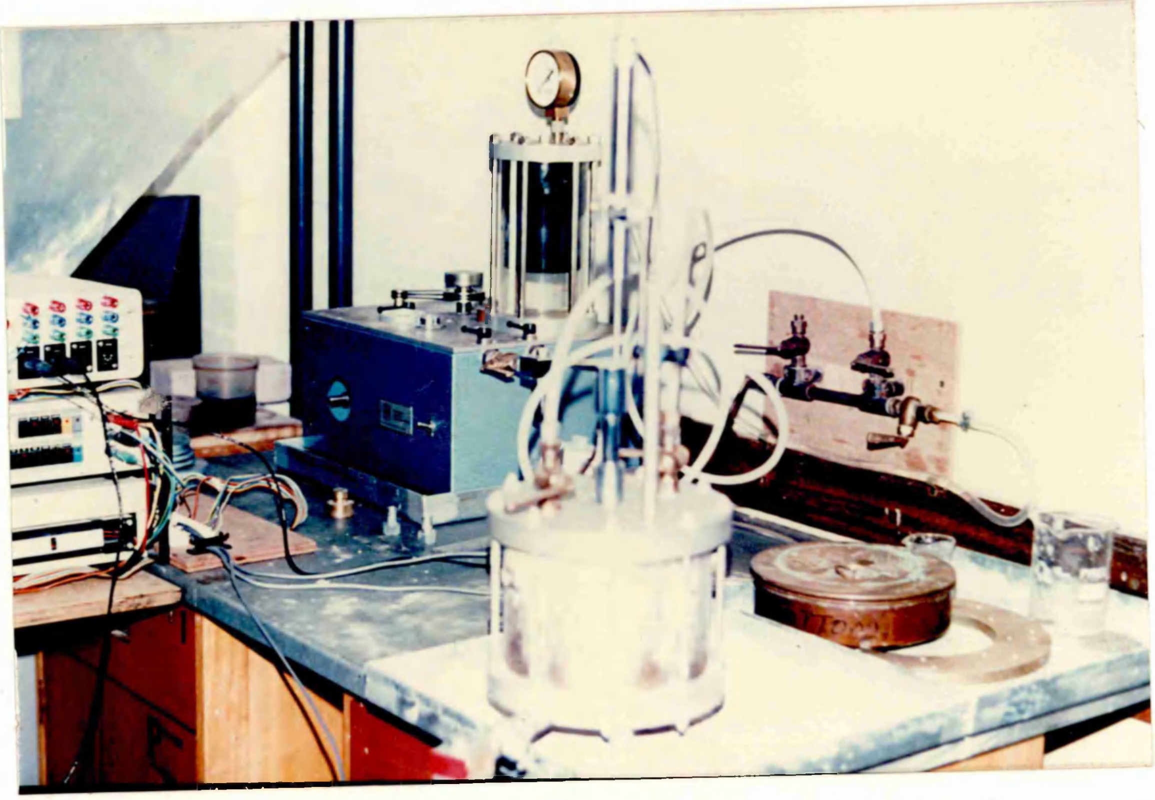


Figure A5 CONSTANT PRESSURE APPARATUS

cell is illustrated in Fig. A3. During sedimentation the cell was clamped to the base of the column as shown in Fig. A2. It could be removed when the column was drained.

1.2 Monitoring and pressure equipment

The consolidation phase of the experiment was monitored for both settlement and pressure. Displacement and pressure transducer readings were recorded automatically under instruction from a computer program written by the author. The program is presented in Appendix VI.

Consolidation pressure was maintained by an oil and water constant pressure apparatus. The monitoring and pressure equipment are shown in Figs. A4 and A5.

2.0 Experimental procedure

The procedure for obtaining consolidated samples of laboratory sedimented clay was as follows:

1. prepare water medium
2. add clay powder and mix for 15 mins
3. leave mixture for 48 hrs

4a. remix for 15 mins

4b. add organic compounds (optional)

5. fill sedimentation column
6. draw off samples
7. drain column
8. remove consolidation cell

9. allow to stand for 48 hrs
10. remove surface water and pre-consolidate (cell open)
11. consolidate sample (cell closed)
10. release pressure
11. determine moisture content and shear strength
12. cut sediment samples for SEM analysis.

The preparation tank was filled with fresh water and if saline conditions were required an amount of 'instant ocean' crystals were added to give the desired salinity. Clay minerals in powdered form were then added slowly to a concentration of 8 g/1, appropriate for natural settling. The clay suspension was thoroughly mixed by recirculation and then left to settle. This allowed time for the clay powder to absorb water and achieve natural equilibrium.

Before delivery to the sedimentation column the suspension was remixed. At this stage the chosen organic compounds could be added if required. The column was filled and the clay particles allowed to settle into the consolidation cell at the base. Periodic checks were made of the stage of settlement by drawing off samples from the drain tubes and checking the turbidity value of the fluid. When it was confirmed that most of the clay had settled the column was drained. The drainage points were spaced so that any turbulence created was minimised.

The consolidation cell was unclamped from the column base and transferred to the workbench. The clay sediment at this stage was too 'thin' to place under pressure from the constant pressure equipment. Pre-consolidation was necessary and this was achieved by placing a perspex disc on the sediment surface with drainage allowed through 'weep holes' in the disc. The pressure was raised from 0.1 kPa to 10 kPa by the addition of more discs. At this pressure the clay sediment had achieved sufficient stiffness for consolidation to be carried out by the pressure equipment itself.

With some modification the lowest attainable pressure provided by the constant pressure apparatus was approximately 31 kPa. By adding further weights this was slowly raised to the nominal minimum pressure of 100 kPa. Thereafter the sediment was consolidated in stages to the maximum of 1000 kPa, each increment being held for 24 hours. Full monitoring of displacement, supply pressure and pore-water pressure (pwp) was carried out with data being recorded by the computer at pre-set time intervals.

On completion of the final increment the pressure was released and the sediment allowed to swell. Moisture content determination of the sediment enabled the void ratio to be calculated for the various steps of the consolidation process.

Before the sediment was removed from the cell the shear strength was determined using the falling cone method of Hansbo (1957).

Samples were obtained from undisturbed parts of the sediment and prepared for electron microscopy work as outlined in Appendix IV.

3.0 Additional experimental work

Part of this study was conducted away from the sponsoring establishment and this required the fabrication of new equipment. The new set-up was based around the standard 75 mm diameter oedometer cell of the Casagrande type. Although not as elaborate the system consisted of the same basic units of mixing tank, sedimentation column and consolidation cell.

There was no facility with the Casagrande oedometer to measure pore water pressure which meant that only settlement could be monitored. This was not such a drawback since pwp measurement in the Rowe Cell had proved difficult.

The experimental procedure was similar to that explained in Section 2 above, except that pressure was provided by the normal loading arrangement of the Casagrande-type oedometer.

4.0 Experimental calculations

The consolidation and strength properties of the sedimented clays have been presented in Chapter 6. The following example illustrates the calculation procedure used to determine those properties.

4.1 Settlement-time record

Test No 3 : Kaolinite sedimented in salt water

Consolidation pressure = 100 kPa

Initial sample thickness = 31.19 mm

Final sample thickness = 28.15 mm

$\sqrt{\text{time}}$ (mins)	0	2	4	7	11	15	19	24	29	38
settlement (mm)	0	.58	1.34	2.28	2.74	2.84	2.86	2.88	2.91	3.04

The coefficient of consolidation (c_v) is given by the equation

$$c_v = \frac{T_v d^2}{t_{90}}$$

where time factor T_v = 0.848 at 90% consolidation

d is the mean drainage path length

t_{90} is the time taken for 90% consolidation.

The value of t_{90} is obtained from the graph of settlement against time (Fig. A6) using Taylor and Merchant's method. This is shown to be 84.6 mins.

The mean drainage path length is given by

$$d = \tfrac{1}{2}(31.19 + 28.15) = 29.67 \text{ mm}.$$

Substituting values into the equation gives

$$c_v = \frac{0.848 \times 29.67^2}{84.6} = 8.82 \text{ mm}^2/\text{min}$$
$$= 4.64 \text{ m}^2/\text{year}.$$

4.2 Void ratio - effective stress record

Test No 3: kaolinite sedimented in salt water

pressure range 0 - 1000 kPa

initial sample thickness = 116.0 mm

final sample thickness (H_o) = 24.25 mm

final moisture content (w_o) = 38.1%

specific gravity of kaolinite (G_s) = 2.65

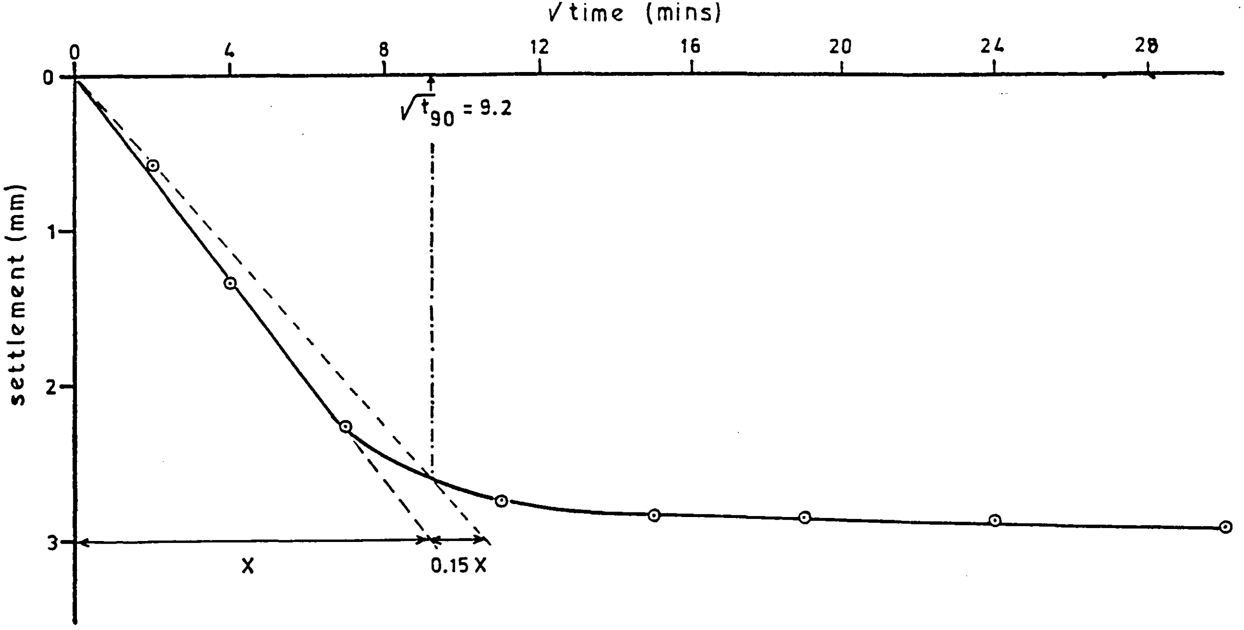


Figure A6 SETTLEMENT-TIME RELATIONSHIP (TEST 3)

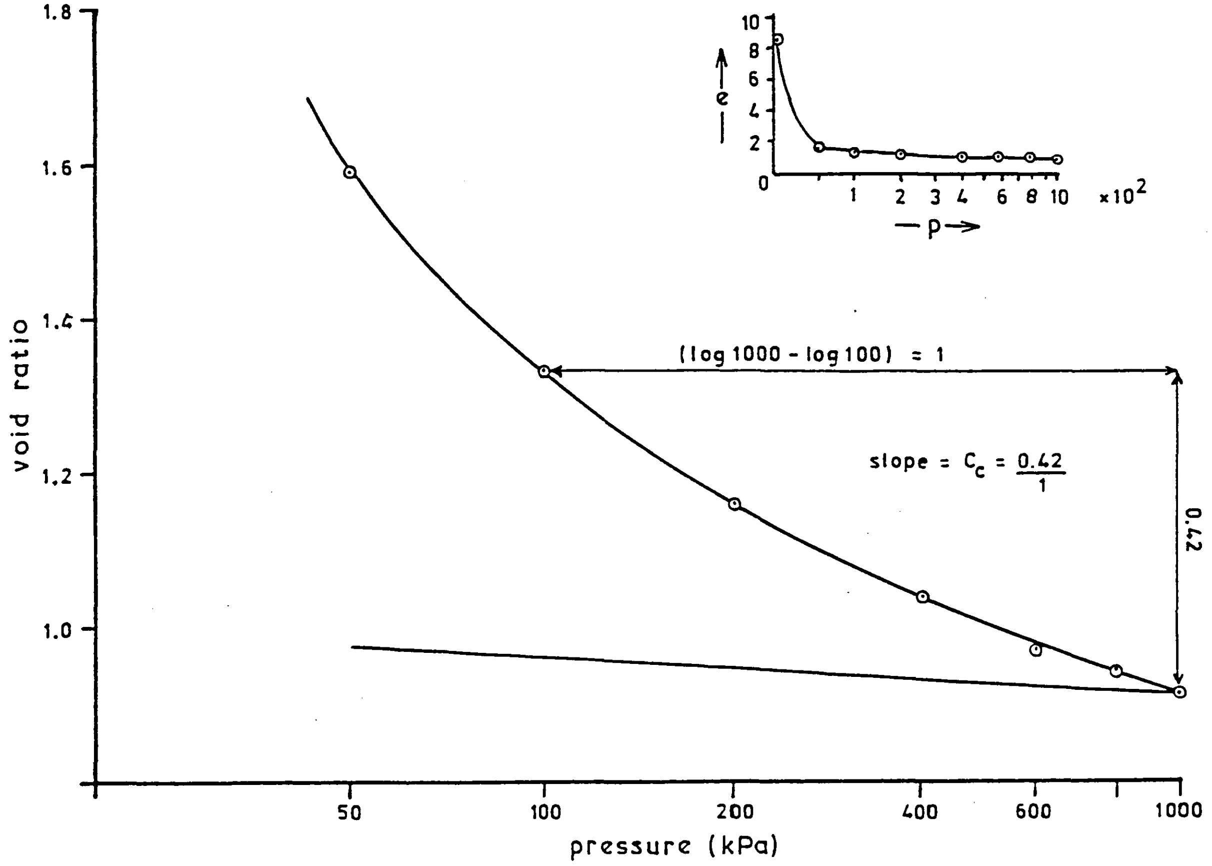


Figure A7 VOID RATIO-PRESSURE RELATIONSHIP (TEST 3)

Under one-dimensional consolidation conditions the change in void ratio (Δe) is related to the change in sample thickness (ΔH) by the equation

$$\frac{\Delta H}{H_o} = \frac{\Delta e}{1+e_o}$$

hence $$\Delta e = \left(\frac{1+e_o}{H_o}\right) \Delta H$$

At the end of the test the clay is saturated and the final void ratio is obtained from $e_o = w_o G_s$ (where G_s is the specific gravity of the clay particles.)

The calculations are tabulated as follows:

pressure (kN/m^2)	sample thickness (mm)	ΔH (mm)	Δe	e
0	116.00	91.75	7.600	8.61
50	31.19	6.94	0.575	1.59
100	28.15	3.90	0.323	1.33
200	26.08	1.83	0.152	1.16
400	24.62	0.37	0.031	1.04
600	23.76	-0.49	-0.041	0.97
800	23.35	-0.90	-0.075	0.94
1000	23.03	-1.22	-0.101	0.91
0	24.25	0	0	1.01

The graph of void ratio against pressure (i.e. effective stress) is shown in Fig A7. The slope of the line is called the compression index C_c and is shown to have a value of 0.42 for the pressure range 100-1000 kPa.

4.3 Shear strength determination

Hansbo (1957) outlined a method of obtaining the undrained shear strength from the penetration depth measured in the standard fall-cone test. The relationship between cone penetration (h) and undrained shear strength (c_u) is given by

$$c_u = \frac{KQ}{h^2}$$

where Q is the weight of the cone

K is a constant whose magnitude depends upon the cone angle.

Hansbo provides tables of h and c_u for different combinations of weight and cone angle.

A number of tests were carried out on each sample and the mean penetration in each case determined. For test 6 of illite sedimented in saltwater the following readings were taken:

penetration (mm): 10.2, 9.4, 9.0, 9.7, 11.3, 9.5

average penetration = 9.9 mm

for a 100 g - 30° cone $c_u = 1.03\ t/m^2$

$= 10.1\ kN/m^2$

5.0 Experimental record: note on pore-water pressure measurements

The measurement of pore-water pressures for the clays consolidated in the Rowe Cells was not successful and therefore pwp values are not given. Difficulty was experienced in flushing the pressure transducers and it is thought that either clay deposits blocked the porous caps in the pressure line to the transducers, or became lodged in the transducer housing preventing the pressure membrane from operating. The problem was not overcome in the time available and the measurement of pore-water pressure was therefore abandoned.

STATISTICAL THEORY

1.0 Introduction to directional data

Directional data is the name given to data which arises when the observations are measured in angles. Whereas linear data can range from $-\infty$ to ∞, directional data can only range from 0° to 360°, around the circle.

The periodic nature of directional data requires that analysis is carried out using special statistical techniques. Linear statistics cannot be used for theoretical reasons as well as for empirically obvious reasons stemming from the arbitrariness of the zero point on the circular scale. For example, consider the arithmetic mean of 359° and 1°. Linear statistics provides the arithmetic mean calculation of $(359^{\circ} + 1^{\circ})/2 = 180^{\circ}$ which is clearly absurd since it is obvious that the true mean is 0° (or 360°).

The statistical methods for describing and analysing directional data are relatively new but comprehensive accounts can be found in Batschelet (1965) and Mardia (1972). Its application specific to clay particle orientation has been reported by Gadson and Kanji (1983).

Angular observations may be represented on a circle of unit radius (Fig A8). A single observation θ° is then a unit vector with the angle θ° measured anti-clockwise to the positive x direction. The cartesian coordinates of the vector are $(\cos\theta, \sin\theta)$ and the polar coordinates are $(1, \theta^{o})$. If the vector is non-directed, ie angle θ^{o} and $(\theta+\pi)^{o}$ are not distinguished, the data is referred to as axial data.

The most common form of axial data arises where it is not possible to differentiate between whether an observation is facing forwards or backwards. This is the case for particles in mudrocks when only a check on preferred orientation is required. In this situation the angles θ^{o} and $(\theta+\pi)^{o}$ are indistinguishable and hence the data can only be measured within the range $(0, \pi)$.

In order to use the appropriate techniques the data must first be transformed. If θ^* is defined as a continuous random variable within the range $(0, 2\pi/L)$ the transformed data for a circular distribution becomes

$$\theta' = L\theta^* \qquad 0 < \theta' \leqslant 2\pi$$

θ' is interpreted as a circular random variable whose probability density function (pdf) is given by

$$f(\theta') = g(\theta'/L)/L$$

where g is the pdf of θ^* on the line.

For axial data $L = 2$,

hence $$\theta' = 2\theta^* \qquad 0 < \theta^* \leqslant \pi$$

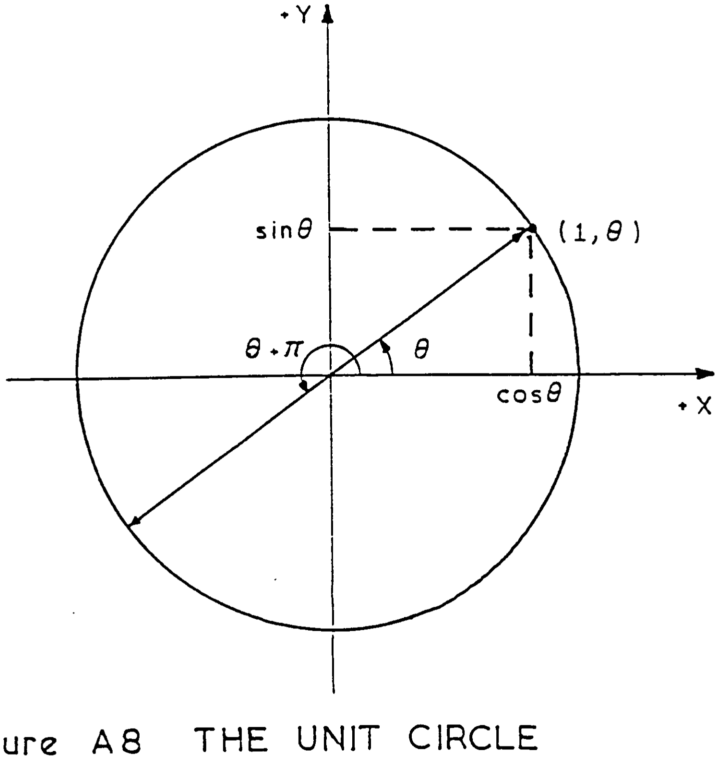


Figure A8 THE UNIT CIRCLE

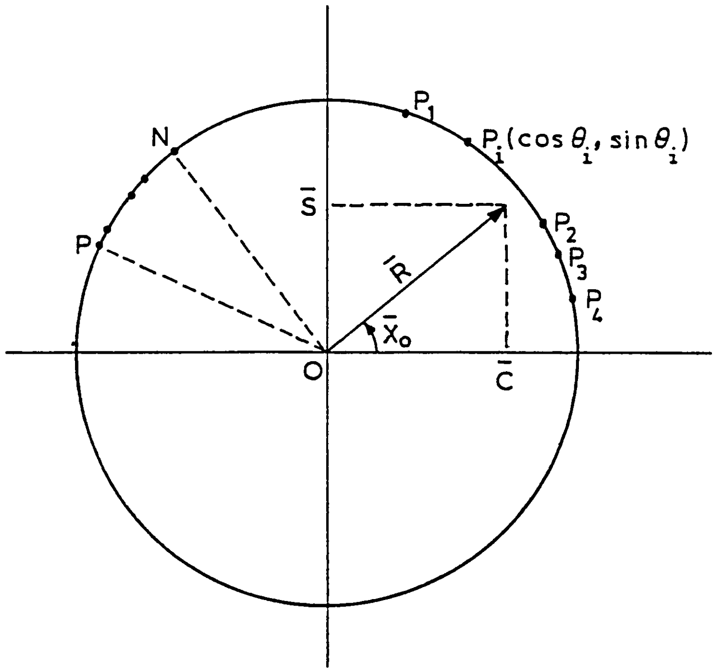


Figure A9 MEAN DIRECTION, MEAN RESULTANT LENGTH AND CIRCULAR VARIANCE

Thus the angles are doubled for axial data before the analysis is carried out.

Data may be presented in a diagrammatic form using either circular histograms, linear histograms or rose diagrams. With linear histograms the point of cut used in unrolling the data should be chosen carefully or an erroneous impression of the data will result. It is usual to chose a cut which gives the maximum concentration of data around the centre of the linear histogram.

The distribution of data about the circle is evident from observing the linear histogram. The most common frequency distributions for directional data are uniform, unimodal and bimodal. As with the linear case it is often unimodal distributions which predominate.

2.0 Statistics of directional data

In order to evaluate unimodal circular distributions it is necessary to introduce some descriptive measures. These are treated in full by Mardia (1972) and the following are employed in this analysis.

2.1 The mean direction ($\bar{X}_o$)

The mean direction of a series of angles, θ_1, θ_2 ... θ_n is defined as the direction of the resultant of the unit vectors OP_1, OP_2 ... OP_n. If P_i is the point on the circumference of the unit circle corresponding to the angle θ_i then the cartesian coordinates of P_i are ($\cos \theta_i$, $\sin \theta_i$), where i = 1, 2, ... n. (Fig A9).

The centre of gravity of these points ($\bar{C}$, $\bar{S}$) is given by

$$\bar{C} = \frac{1}{n}\sum_{i=1}^{n} \cos\theta_i \qquad \bar{S} = \frac{1}{n}\sum_{i=1}^{n} \sin\theta_i \tag{2.1.1}$$

and the mean resultant length is defined as

$$\bar{R} = (\bar{C}^2 + \bar{S}^2)^{\frac{1}{2}} \tag{2.1.2}$$

whereby the length of the resultant, $R = n\bar{R}$.

The mean direction is found by solving the equations

$$\bar{C} = \bar{R}\cos\bar{X}'_o \quad \text{and} \quad \bar{S} = \bar{R}\sin\bar{X}'_o \tag{2.1.3}$$

The solution provides $\bar{X}'_o = \arctan(\bar{S}/\bar{C}), -\frac{\pi}{2} \leqslant \bar{X}'_o \leqslant \frac{\pi}{2}$

so that in order to obtain the mean direction, the following conditions are applied

$$\bar{X}_o = \begin{cases} \bar{X}'_o & \text{if} \quad \bar{S} \geqslant 0,\ \bar{C} \geqslant 0 \\ \bar{X}'_o + \pi & \text{if} \quad \bar{C} \leqslant 0 \\ \bar{X}'_o + 2\pi & \text{if} \quad \bar{S} < 0 \quad \bar{C} \geqslant 0 . \end{cases}$$

It may be shown that $\bar{X}_o$ has two important properties:

(1) $\bar{X}_o$ is invariant to a change in zero direction;

(2) the sum of the deviations about $\bar{X}_o$ is zero.

2.2 Circular variance (S_o)

In linear distributions a measure of the concentration of data is given by the standard deviation. With circular distributions it is more appropriate to measure the dispersion of data which is referred to as the circular variance (Mardia 1972).

For any set of directional data the circular dispersion is measured by the smallest angle which accommodates all the data. Thus in Fig A9 the circular dispersion of data lying between OP and ON on the unit circle is given by the angle NOP.

If α is the zero direction then a measure of the dispersion of the points P_i (for i = 1 to n) is given by

$$D = \frac{1}{n} \sum_{i=1}^{n} \left[1 - \cos(\theta_i - \alpha) \right] \qquad (2.2.1)$$

and if D is minimised with respect to then

$$\frac{dD}{d\alpha} = \sum_{i=1}^{n} \sin(\theta_i - \alpha) = 0 \qquad (2.2.2)$$

But from (2.1.2) and (2.1.3)

$$\sum_{i=1}^{n} \sin(\theta_i - \bar{X}_o) = 0$$

hence $\alpha = \bar{X}_o$.

It may be concluded that the dispersion is smallest about the mean direction.

If the value of D about $\bar{X}_o$ is defined as S_o, such that

$$S_o = 1 - \frac{1}{n} \sum_{i=1}^{n} \cos(\theta - X_o) \qquad (2.2.3)$$

then using (2.1.3) $\quad S_o = 1 - \bar{R}$

S_o is called the circular variance and $\bar{R}$ is the mean resultant length.

It can be shown that S_o is invariant under a change of zero direction.

The range of S_o is $0 \leqslant S_o \leqslant 1$.

If the data is clustered about the mean then $\bar{R}$ is close to unity and S_o is very small. For data distributed evenly about the circle then $\bar{R}$ becomes small and S_o is close to unity.

3.0 Circular distribution models

There is no single distribution on the circle which has all the desirable properties that the normal distribution possesses on the line. There are many circular models, most of which have been derived from transformations of the standard univariate/bivariate models (e.g. uniform) or as circular analogues of important univariate characterisations (e.g. Von Mises).

Of the many circular distributions, three are found to cover most statistical applications. These are:

(1) the uniform distribution

(2) the wrapped normal distribution

(3) the Von Mises distribution.

The latter was recommended as appropriate for the statistical analysis of data in this study.

3.1 The Von Mises distribution

Introduced by Von Mises (1918) for his study of the derivations of measured atomic weights, the Von Mises distribution is as important in circular statistics as the normal distribution is in linear statistics.

The Von Mises distribution has two properties which are common to the linear normal distribution.

(1) the maximum likelihood characteristic

$\bar{X}_o$ is the maximum likelihood estimate of μ_o only if θ is Von Mises;

(2) the maximum entropy characteristion

the Von Mises distribution has the maximum entropy under the condition that the mean direction and circular variance are fixed.

A circular random variable θ is said to have a Von Mises distribution if its probability density function is given by

$$g(\theta;\, \mu_o,\, K) = \frac{1}{2\pi I_o(K)}\, e^{K\cos(\theta - \mu_o)} \qquad \begin{array}{c} 0 < \theta \leqslant 2\pi \\ K > 0 \\ 0 \leqslant \mu_o < 2\pi \end{array} \qquad (3.1.1)$$

where $I_o(K)$ is the modified Bessel function of the first kind, and of order zero.

$$\text{i.e.} \qquad I_o(K) = \sum_{r=0}^{\infty} \frac{1}{r!^2} (\tfrac{1}{2}K)^{2r}$$

μ_o is the mean direction

K is a concentration parameter.

The density is often abbreviated as $M(\mu_o, K)$.

The larger the value of K, the greater the clustering of the distribution about μ_o.

The Von Mises distribution is unimodal, with the mode at $\theta = \mu_o$ and the antimode at $\theta = \mu_o + \pi$. It is symmetrical about $\theta = \mu_o$ and has two points of inflexion. For $\mu_o = 0$ the points of inflexion are given by

$$\pm \arccos \left[-\frac{1}{2K} + \left(1 + \frac{1}{4K^2}\right)^{\frac{1}{2}} \right]$$

The density of the Von Mises distribution for values of K and μ_o is illustrated in Fig A10. Note that the Von Mises distribution is uniform when K = 0.

3.2 The maximum likelihood estimates of μ_o and K for the Von Mises distribution

In order to predict that a set of data fits a Von Mises distribution it is necessary to evaluate the maximum likelihood estimates for μ_o and K.

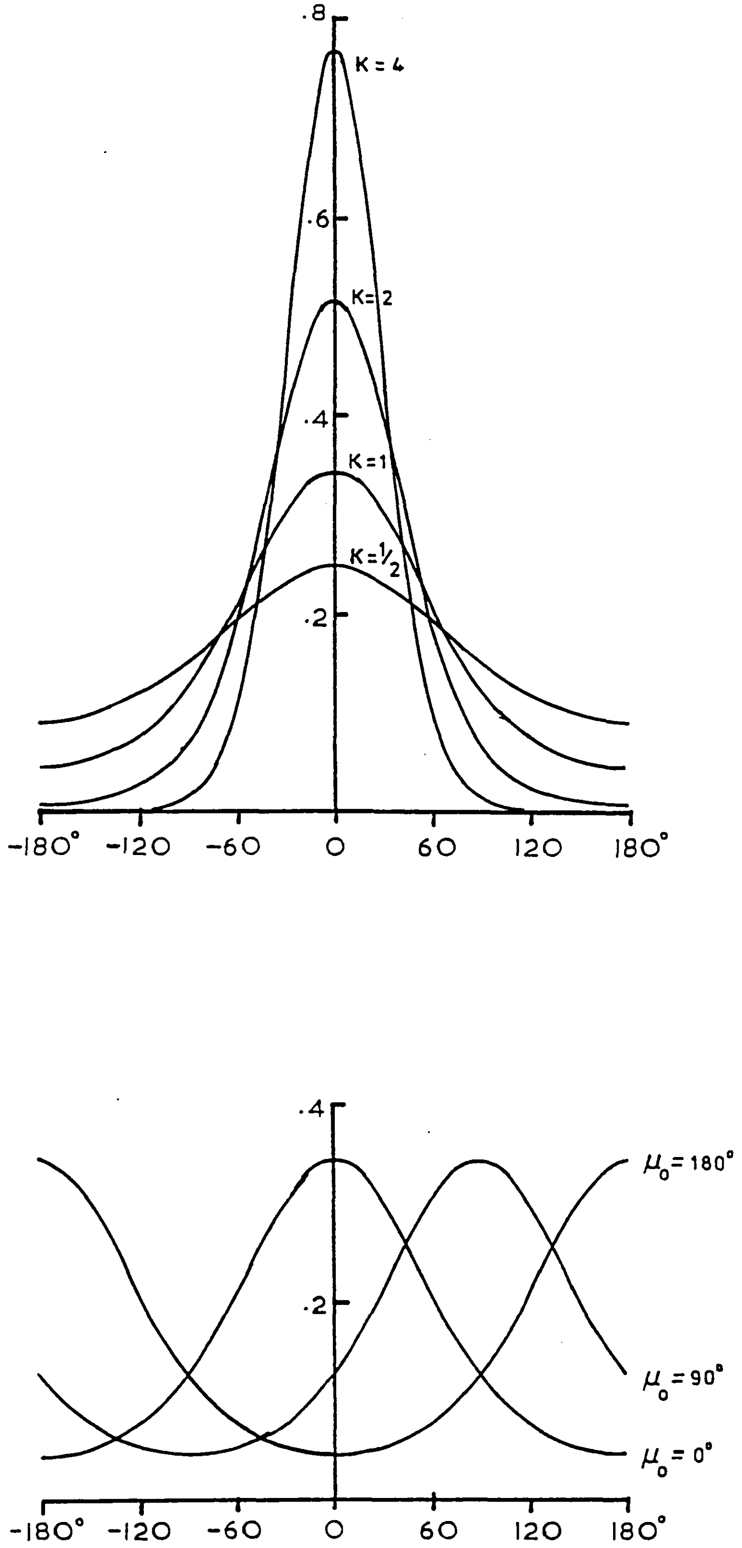


Figure A10 DENSITY OF THE VON MISES DISTRIBUTION (Mardia 1972)

If $\theta_1 \ldots\ldots \theta_n$ are a random sample from a Von Mises distribution of density $M(\mu_o, K)$ then by taking the loglikelihood of the pdf and differentiating with respect to the parameter being estimated, this gives

$$\mu_o = \overline{X}_o$$

The m.l.e. $(\hat{K})$ of K is a numerical solution of

$$\hat{K} = A^{-1}\ (\bar{R})$$

where A = ratio of modified Bessel function I_1/I_o.
Values of $\hat{K}$ corresponding to values of $\bar{R}$ are given by Batschelet (1965) and appear in Table A1.

Approximations for the values of K may be derived from the following formulae

For very small $\bar{R}$ $\quad \hat{K} = 2\bar{R}$

for small $\bar{R}$ (<0.45) $\quad \hat{K} = \bar{R}(12 + 6\bar{R}^2 + 5\bar{R}^4)$

for large $\bar{R}(>0.80)$ $\quad \hat{K} = [2(1 - \bar{R}) - (1 - \bar{R})^2 - (1 - \bar{R})^3]^{-1}$

for $\bar{R}$ nearly 1 $\quad \hat{K} = \dfrac{1}{2(1 - \bar{R})}$

3.3 Goodness of fit for a Von Mises distribution

A test for the goodness of fit of a sample of data to a Von Mises distribution is carried out by finding the values of μ_o and K and then using these estimates to determine the expected values of the distribution by integrating the p.d.f.

$\bar{R}$	k	$\bar{R}$	k	$\bar{R}$	k
0.00	0.00000	0.35	0.74783	0.70	2.01363
.01	.02000	.36	.77241	.71	2.07685
.02	.04001	.37	.79730	.72	2.14359
.03	.06003	.38	.82253	.73	2.21425
.04	.08006	.39	.84812	.74	2.28930
.05	.10013	.40	.87408	.75	2.36930
.06	.12022	.41	.90043	.76	2.45490
.07	.14034	.42	.92720	.77	2.54686
.08	.16051	.43	.95440	.78	2.64613
.09	.18073	.44	.98207	.79	2.75382
.10	.20101	.45	1.01022	.80	2.87129
.11	.22134	.46	1.03889	.81	3.00020
.12	.24175	.47	1.06810	.82	3.14262
.13	.26223	.48	1.09788	.83	3.30114
.14	.28279	.49	1.12828	.84	3.47901
.15	.30344	.50	1.15932	.85	3.68041
.16	.32419	.51	1.19105	.86	3.91072
.17	.34503	.52	1.22350	.87	4.17703
.18	.36599	.53	1.25672	.88	4.48876
.19	.38707	.54	1.29077	.89	4.85871
.20	.40828	.55	1.32570	.90	5.3047
.21	.42962	.56	1.36156	.91	5.8522
.22	.45110	.57	1.39842	.92	6.5394
.23	.47273	.58	1.43635	.93	7.4257
.24	.49453	.59	1.47543	.94	8.6104
.25	.51649	.60	1.51574	.95	10.2716
.26	.53863	.61	1.55738	.96	12.7661
.27	.56097	.62	1.60044	.97	16.9266
.28	.58350	.63	1.64506	.98	25.2522
.29	.60625	.64	1.69134	.99	50.2421
.30	.62922	.65	1.73945	1.00	∞
.31	.65242	.66	1.78953		
.32	.67587	.67	1.84177		
.33	.69958	.68	1.89637		
.34	.72356	.69	1.95357		

Table A1

The maximum likelihood estimate $\hat{k}$ for given $\bar{R}$ in the von Mises case.
For the solution $k = A^{-1}(p)$, replace $\hat{k}$ by k, $\bar{R}$ by p
(after Batschelet (1965))

$$\int_{L}^{U} \left[2\pi I_o(K)\right]^{-1} \exp\left[K \cos(\theta - \mu_o)\right] d\theta \qquad \begin{array}{c} K > 0 \\ 0 < \theta \leq 2\pi \end{array}$$

where U and L are the upper and lower bounds of the requuired interval.

Once the expected values for the Von Mises distribution are found, a chi-squared goodness of fit test is carried out on the observed and expected frequencies according to

$$\chi^2_{\nu,\alpha} = \sum \frac{(O - E)^2}{E}$$

where O = observed frequency

E = expected frequency

ν = degrees of freedom

α = required probability level.

If the expected value of frequency in a cell is less than 5, then cells must be combined until a frequency of 5 or greater exists for every cell or combination of cells.

4.0. Tests for samples from a Von Mises population

Assuming that the data conforms to a Von Mises distribution, $M(\mu_o, K)$, then the following parametric tests (Watson and Williams 1956) may be carried out.

4.1 Rayleigh test of uniformity

This test is designed to check whether a set of data has a preferred orientation on the circle. The null hypothesis (H_o) is that the data comes from a uniform distribution. The alternative hypothesis (H_A) is that the data conforms to a Von Mises population with a preferred direction (μ_o) and a concentration parameter K.

Thus if $\theta_1 \ldots\ldots \theta_n$ are a random sample from a population with p.d.f. $f(\theta)$ then

$$H_o : f(\theta) = 1/2\pi \qquad 0 \leqslant \theta \leqslant 2\pi$$

$$H_A : f(\theta) = g(\theta) = \left[2\pi I_o(K)\right]^{-1} \exp\left[K \cos(\theta - \mu_o)\right] \qquad 0 \leqslant \theta \leqslant 2\pi$$
$$K > 0$$

The critical region for the test is that

$$\bar{R} > K$$

where $\bar{R}^2 = \bar{C}^2 + \bar{S}^2$

and K is found from tabulated critical values.

Stephens (1969) tabulated the critical values of $\bar{R}$ for given sizes (n) of sample data, and these are shown in table A2.If $\bar{R} > K$ then the null hypothesis of uniformity is rejected.

4.2 Confidence interval for μ_o

For a population mean direction of μ_o a check can be made to see if the sample mean direction $\bar{X}_o$ differs significantly from it.

n \ α	0.10	0.05	0.025	0.01	0.001
5	0.677	0.754	0.816	0.879	0.991
10	.478	.540	.594	.655	.775
15	.391	.443	.489	.542	.649
20	.339	.385	.425	.472	.569
25	.303	.344	.381	.423	.512
30	.277	.315	.348	.387	.470
35	.256	.292	.323	.359	.436
40	.240	.273	.302	.336	.409
45	.226	.257	.285	.318	.386
50	.214	.244	.270	.301	.367
100	.150	.170	.190	.210	.260

Table A2. CRITICAL VALUES OF THE RAYLEIGH TEST OF UNIFORMITY WITH THE TEST STATISTICS $\bar{R}$. $Pr(\bar{R} \geq \bar{R}_o) = \alpha$

(after Batschelet 1971)

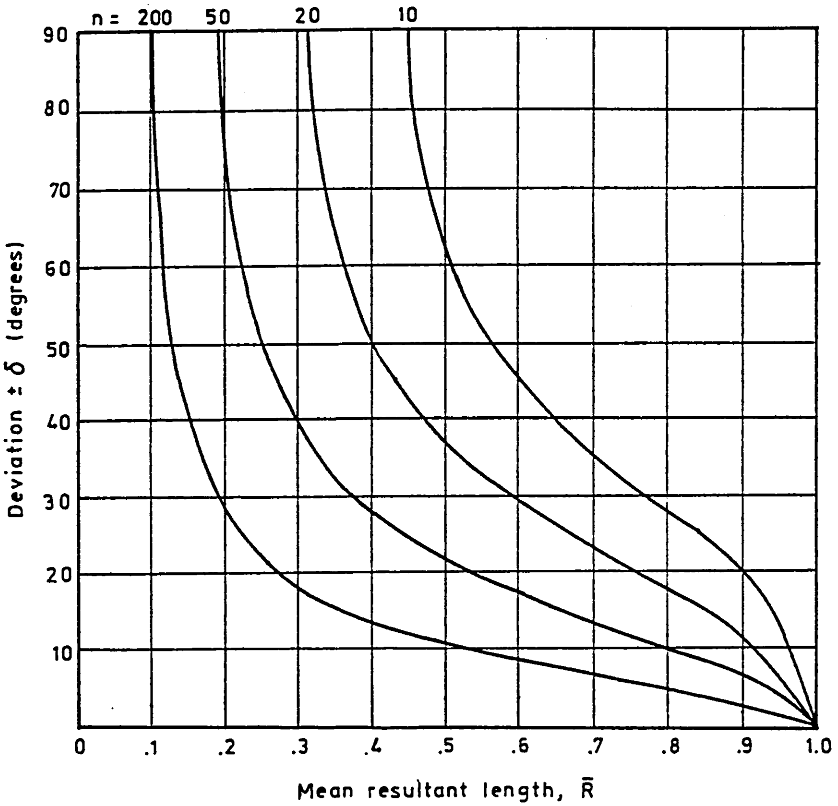


Figure A11. CONFIDENCE INTERVALS FOR μ_o
BATSCHELET CHART FOR OBTAINING A 95% CONFIDENCE INTERVAL FOR THE MEAN DIRECTION μ_o. $Pr(-\delta < \mu_o < \delta) = 0.95$

(after Batschelet 1971)

The $(1 - \alpha)$ confidence interval for μ_o is obtained by applying the values of δ from Fig. A11 to the arc containing $\bar{X}$, ie $\bar{X}_o \pm \delta$.

The condition $(\bar{X} - \delta) < \mu_o < (\bar{X} + \delta)$ satisfies the null hypothesis that there is no significant difference between the population and sample mean directions.

4.3 Two sample testing : Tests for mean directions

Suppose that $\theta_{1,1}$ ..... $\theta_{1,n}$ and $\theta_{2,1}$ ..... $\theta_{2,n}$ are two independent random samples of size n from a Von Mises population $M(\mu_{o,i}, K_i)$; $i = 1, 2$.

The null hypothesis is $H_o : \mu_{o,1} = \mu_{o,2} = \mu_{o,i}$.

For large values of K, Watson and Williams (1956) give the following approximation

$$2K(R_1 + R_2 - R) \backsim \chi_1^2$$

where R_1 and R_2 are the resultant lengths for samples 1 and 2 respectively and R is the resultant length for the combined sample. If the value of $2K(R_1 + R_2 - R)$ is less than the corresponding chi-squared value then the null hypothesis cannot be rejected.

4.4 Multi-sample test : One-way classification

Suppose θ_{i1}, ... θ_{in_i} , $i = 1, \ldots, q$, are q independent random samples of size n_i from a Von Mises population $M(\mu_{o,i}, K_i)$, $i = 1, \ldots, q$.

If R_i and R are the lengths of the resultant for the ith sample and combined sample respectively then

$$R_i^2 = C_i^2 + S_i^2 \quad \text{and} \quad R^2 = C^2 + S^2$$

where C_i, S_i are the sums of the cosines and sines for the ith sample and C, S are the corresponding sums for the combined sample.

For a one way classification the null hypothesis $H_o : \mu_{o,1} = \mu_{o,2} = \ldots\ldots = \mu_{o,q}$ is tested against the alternative hypothesis that at least one of the equalities does not hold. It is assumed that $K_1 = K_2 = \ldots\ldots = K$ where K is unknown.

Because the p.d.f. of $R_1 \ldots\ldots R_q$ (given R) is independent of K the critical region is taken as

$$R_1 + \ldots\ldots + R_q > K$$

for a given resultant length R. The critical points for K are unknown but this difficulty is overcome by using the following method.

For values of $K \geqslant 1$ and $q > 2$ it may be proven under H_o that

$$F_{q-1,\ n-q} = 1 + \frac{3}{8\hat{K}} \left[(n-1)(\Sigma R_i - R)/(n - \Sigma R_i)(q-1)\right] \qquad (4.4.1)$$

where $\hat{K}$ is the m.l.e. of K based upon $\bar{R}$

$(\Sigma R_i - R)/q-1)$ is the variance of the sample mean about the population

$(n-\Sigma R_i)/(n-q)$ is the variance of the sample about the sample mean.

This may be arranged in the form of an analysis of variance (ANOVA) table in which case equation (4.4.1) may be rewritten as

$$\frac{F_{q-1,\ n-q}}{1+\frac{3}{8\hat{K}}} = (n-q)(\Sigma R_i - R)/(n-\Sigma R_i)(q-1)$$

which yields the following ANOVA table

source	degrees of freedom	sum of squares	mean square	$F/(1+\frac{3}{8\hat{K}}$
between samples	$q-1$	$\Sigma R_i - R$	$(\Sigma R_i - R)/(q-1) = I$	I/II
within samples	$n-q$	$n-\Sigma R_i$	$(n-\Sigma R_i)/(n-q) = II$	
total	$n-1$	$n-R$		

If $0 < K < 1$ and $q > 2$ (ie $\bar{R} < 0.45$) the likelihood criterion is

$$-2\log_e \lambda \backsim \frac{2}{n}(\Sigma R_i^{\ 2} - R^2) = U$$

For large n, U is distributed as a chi-squared variable with $(q-1)$ degrees of freedom when the null hypothesis is true.

5.0 Non-parametric tests

When a set of data does not conform to a Von Mises distribution it is necessary to apply appropriate analogues of non-parametric tests on the line for continuous populations. Because there is no unique way of ordering a sample on the circle, these circular analogues are invariant under rotations. Tests for goodness of fit on the circle are often applied to test the hypothesis of uniformity.

5.1 Watson's U^2 test of uniformity

This is a test of uniformity where the hypothesis $H_o : F = F_o$ is tested against $H_A : F \neq F_o$.

For a given F_o the above problem can be reduced to testing uniformity on applying the probability integral transformation

$$U = F(\theta) \tag{5.1.1}$$

which implies that U is distributed uniformly on (0, 2π) provided that F is continuous.

Watson (1961) proposed the statistic

$$U^2 = n \int_0^{2\pi} \left[S_n(\theta) - F(\theta) - \mu \right]^2 dF(\theta) \tag{5.1.2}$$

where

$$\mu = \tfrac{1}{2} - \frac{1}{n} \sum_{i=1}^{n} F\theta_i \tag{5.1.3}$$

U^2 may be interpreted as a type of variance of the Cramer - Von Mises statistic, W^2, which is measured about the origin. This statistic may be expressed by

$$W^2 = \sum_{i=1}^{n} \left[U_i - \left\{ (2i - 1)/2n \right\} \right]^2 + 1/12n$$

where U_i are the uniform order statistics.

If $\bar{U} = (U_1 + \ldots\ldots + U_n)/n$ then from (5.1.2) and (5.1.3)

$$U^2 = \sum_{i=1}^{n} \left[U_i - \bar{U} - \left\{(2i-1)/2n\right\} + \tfrac{1}{2}\right]^2 + 1/12n$$

In this case $U_i = \frac{\theta_i}{360}$

For $n \geqslant 8$, Stephens (1970) has shown that a perfectly adequate modification of U^2 is given by

$$U^{*2} = (U^2 - 0.1/n + 0.1/n^2)(1 + 0.8/n)$$

where the percentage points for this statistic are listed below.

α	0.1	0.05	0.025	0.01
U^{*2}	0.152	0.187	0.221	0.267

Table A3

5.2 Two-sample testing : Watson's two-sample U^2 test

Watson (1962) produced a two-sample analogue of the one-sample U^2 test. The null hypothesis is that the two samples come from the same population or from two populations with the same directions.

Let the ith observation of sample 1 be a_{1i} and the jth observation of sample 2 be a_{2j}. The relative cumulative frequencies of the observations in sample 1 are i/n_1 and in sample 2 are j/n_2.

The test statistic is

$$U^2 = \frac{n_1 n_2}{N^2} \left[\sum_{k=1}^{n} d_k^2 - \frac{\left(\sum_{k=1}^{n} d_k\right)^2}{N}\right]$$

where $N = n_1 + n_2$

$$d_k = \frac{i}{n_1} - \frac{j}{n_2}$$

and k runs from 1 to N.

Critical values of $U^2_{\alpha, n1, n2}$ are given by Batschelet (1981).

If the calculated U^2 value is less than the critical then H_o is not rejected.

5.3 Multi-sample testing : Uniform Scores Test

This tests the equality of q-populations for a set of q independent random samples.

Let θ_{ij}, $j = 1, \ldots, n$ and $i = 1, \ldots, q$ be q independent random samples of size $n_1, \ldots, n_q$ and let r_{ij}, $j = 1, \ldots, n_i$ be the ranks of the angles for the ith sample.

The test statistic is:

$$W = 2 \sum_{i=1}^{q} (C_i^2 + S_i^2)/n_i \backsimeq \chi^2_{2(q-1)}$$

where

$$C_i = \sum_{j=1}^{n_i} \cos\ (2\pi r_{ij}/n) \qquad S_i = \sum_{j=1}^{n_i} \sin\ (2\pi r_{ij}/n)$$

and $n = \sum n_i$.

The null hypothesis is rejected for values of W greater than the chi-squared value.

Because the ranks of the angles are being used rather than the angles themselves then a transformation ($\theta' = 2\theta$) is not necessary. A random sample of ten angles from each population is considered adequate for the test.

If two populations are being compared then $q = 2$ and $W = \frac{2nR_1^2}{n_1 n_2}$

STATISTICAL PROCEDURE

1.0 Introduction

The usual approach to the interpretation of rock and soil microstructure from SEM micrographs is one of qualitative assessment (e.g. Barden 1972b). For there to be a meaningful comparison between the microstructure of various samples then quantitative measurement is essential (e.g. Tovey 1971, 1973). Once data has been captured then appropriate statistical analyses may be carried out (e.g. Gipson 1965, Smart 1973).

In this study the SEM micrographs were to be assessed for preferred particle orientation. Angular data in this form is amenable to directional data analysis as described by Batschelet (1965) and Mardia (1972). The statistical theory has been presented in Appendix II. The objective of the analysis was to determine if a preferred orientation is present and if this orientation has significance to any macro-feature (i.e. fissility). It was also possible to check for consistency of orientation between similar specimens and assess the effects of a change in magnification. The procedure and applicable statistical tests are outlined in Table A4.

2.0 Collection of data

In order to carry out the study, micrographs of mudrock and laboratory clay specimens were taken using an SEM. The specimens had been fractured and mounted to give a view parallel to the natural 'grain', in the case of rock, or at right angles to the applied pressure for laboratory clays. To ensure a randomly chosen view was obtained the positioning dials of the SEM stage were set randomly before recording the image. Several micrographs were taken for each specimen at different places across its surface. In some cases the place was held fixed and micrographs were taken at varying magnifications.

The angular data from micrographs was obtained by using the computer aided design system 'DOGS' supplied by PAFEC Ltd of Nottingham. Micrographs were fixed to a tablet and the alignment of particles was recorded with a 'mouse'. The values of angles were automatically given relative to the base of the micrograph. Correct orientation of the micrograph was essential.

To measure the orientation of every particle on the micrographs was not feasible so a sample of 50 particles from each micrograph was taken. To reduce sampling bias a grid system was used and random numbers were then chosen for the x and y coordinates. Random numbers which gave coordinate positions of 'directionless' particles and featureless areas were rejected.

3.0 Analysis of data

The data for each micrograph was read by a statistical program compiled by the author (see STAT1 in Appendix IV). Because the data is axial the program first sorts the values into angular intervals on the circle. It then proceeds to calculate the standard parameters of mean direction and circular variance in the range 1 to

180°. The value of circular variance is an indication of the tightness with which the observations cluster about the mean direction. A low value of circular variance suggests a preferred orientation (see Appendix II Section 2.2). The significance of uniformity is not tested at this stage.

To undertake further tests on the data a chi-squared goodness of fit is carried out to confirm a Von Mises distribution. This is completed by the second part of program STAT1. The value of chi-square obtained is checked against values from standard tables (see Batschelet 1981).

3.1 Test of uniformity

This test is carried out to check if the clay particles in a micrograph are aligned in a preferred direction. For Von Mises distributed data the Raleigh test is used (Appendix II, Section 4.1). For data not from a Von Mises population Watson's U^2 test is applied using Stephens' (1970) modified statistic U^{*2} (Appendix II, Section 5.1).

3.2 Confidence inverval for μ_o

For a Von Mises population this check allows a comparison between the preferred direction of particles (mean direction $\bar{x}_o$) and the direction of the fissile plane (true mean direction μ_o). It is only applied where the angle of the fissile plane is obvious in a low magnification micrograph. There is no equivalent check for a non-Von Mises population.

3.3 Two-sample testing

This test is carried out on two micrographs of the same stub specimen to compare their microstructure. Von Mises populations use Watson and Williams two sample test and the equivalent non parametric test is Watson's U^2 test (Appendix II, Sections 4.3 and 5.2 respectively).

3.4 Multi-sample testing

Where more than two micrographs are available then a comparison of microstructure is achieved by multi-sample testing. In this way a measure of the change in any preferred direction due to differences in micrograph magnification is made. The two tests for Von Mises and non-Von Mises populations are the One-way Classification (Appendix II, Section 4.4) and the Uniform Scores Test (Appendix II, Section 5.3) respectively. The latter test may also be applied to two-sample testing.

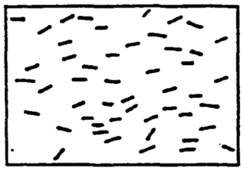
	Requirement	Statistical test	
1.	Derive parameters	$\bar{C}$, $\bar{S}$, $\bar{R}$, X_o, S_o	
2.	Goodness of fit	χ^2 test for Von Mises distribution	
		pass	fail
3.	Test of uniformity	Rayleigh	Watson's U^2
4.	Comparison to fissile plate	Confidence interval for μ_o	none
5.	Two-sample comparison	Watson and Williams	Watson's U^2
6.	Multi-sample comparison	One-way classification	Uniform Scores test

TABLE A4 STATISTICAL TESTS FOR DIRECTIONAL DATA

4.0 Example analyses

By way of illustrating the statistical procedure followed in the study some example calculations are presented here. The particle observations from three increasing magnification shots of a specimen of Stockdale Shale are used as data. The recorded particles from the micrographs are also shown.

OBSERVED PARTICLES DATA ANGLES



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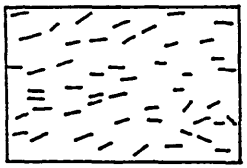
7	31	21	1	18	179	21	179	15	23
172	38	30	2	3	4	20	179	13	6
2	177	9	10	2	30	20	48	176	18
22	49	32	179	26	178	21	168	10	174
14	2	18	7	167	9	171	17	18	13



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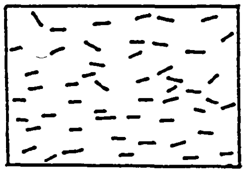
11	22	2	16	180	7	3	22	11	33
38	4	15	31	3	14	26	29	9	10
9	12	26	177	12	4	16	33	1	19
32	27	2	25	179	176	165	40	22	18
8	8	170	176	28	19	172	140	151	176



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21	179	6	21	19	176	4	22	120	179
30	3	13	11	14	32	14	152	8	176
151	10	5	2	10	3	8	7	18	25
10	14	176	174	27	158	41	10	9	22
178	1	177	166	170	17	179	40	1	38

4.1 Determination of standard parameters

The output from program STAT1 for the data associated with each micrograph gave the following standard parameters.

	$\bar{C}$	$\bar{S}$	S_θ	$\bar{R}$	$\bar{X}_\theta$
Pic 187	0.774	0.376	0.037	0.860	25.9
Pic 188	0.790	0.336	0.038	0.858	23.0
Pic 189	0.803	0.228	0.044	0.835	15.8

The results show that the data angles are tightly clustered about mean particle directions of 26°, 23° and 16° in each case.

4.2 Goodness of fit test for Von Mises distribution

The observed and expected frequencies for the class intervals were as follows for Pic 187.

CLASS INTERVAL	OBSERVED (O)	EXPECTED (E)	$(O-E)^2/E$	CELL
1 - 30	16	17.4	0.113	1
31 - 60	15	15.0	0.000	2
61 - 90	4			3
91 - 120	3			
121 - 150	0			
151 - 180	0			
181 - 210	0 (7)	6.31	0.075	
211 - 240	0			4
241 - 270	0			
271 - 300	0			
301 - 330	0			
331 - 360	12 (12)	9.84	0.476	

Chi-squared value = 0.664

At the 5% level $\chi^2_{n-3} = \chi^2_1$ = 3.84 (from standard tables).

Thus, since the calculated value (0.664) is less than 3.84, we conclude that there is no evidence at the 5% level that the Von Mises distribution is a bad fit.

Similarly for Pic 188 $\chi^2_1 = 0.388$

and Pic 189 $\chi^2_1 = 1.113$

Thus for all three micrographs the angular data fit a Von Mises distribution.

4.3 Rayleigh test of uniformity

From Table A2 the critical value of $\bar{R}$ at the 5% level for $n = 50$ is 0.244. The values of $\bar{R}$ for each micrograph are given in 4.1, previously. Hence for all three micrographs $\bar{R} > R_{crit}$ and the null hypothesis, H_o, is rejected and we conclude that each has a strong preferred particle orientation.

4.4 Confidence interval for μ_o (note - doubled angles)

The angle of the bedding plane for the specimen was measured as 20°. From Table A3 at the 5% level $\delta = 8^o$. Thus the confidence interval for μ_o in each micrograph is

Pic 187	25.9 ± 8^o
Pic 188	23.0 ± 8^o
Pic 189	15.8 ± 8^o

Since the bedding plane angle falls within the confidence interval for all three micrographs we can accept the null hypothesis at the 5% level of significance. Thus the preferred particle orientation of the samples is not significantly different from the angle of the bedding (fissile) plane of the specimen.

4.5 One-way classification

This test yields an analysis of variance table which is formed in the following manner.

number of micrographs, $q = 3$

number of particles, $n = 150$

For the combined sample

$\Sigma Ri = (0.860 + 0.858 + 0.835) \times 50 = 127.65$

$\Sigma \bar{R}^2 = [(0.774 + 0.790 + 0.803) \times 50]^2 + [(0.376 + 0.336 + 0.228) \times 50]^2$

$\Sigma R = 127.34$

Source	D.F.	S.S	MS	RATIO
between micrographs	2	0.31	0.155	1.02
within micrographs	147	22.35	0.152	
total	149	22.66		

overall mean resultant $\bar{R} = 0.849$ and from table A1, $\hat{K} = 3.68$.

The calculated ratio is multiplied by $(1 + \frac{3}{8K})$
where $K = \hat{K} = 3.68$.

The ratio converted to an F-ratio becomes $F_{2,147} = 1.12$. At the 5% level from standard tables $F_{2,147}$ is 3.055 and thus we conclude that there is no significant difference between the micrographs. This confirms that the change in magnification in this case does not alter the assessment of preferred particle orientation.

4.6 Conclusions

These tests have shown that the specimen of Stockdale Shale has a preferred particle orientation which is not significantly different from the orientation of the fissile plane. Furthermore, the orientation is not significantly altered by viewing at different magnifications.

THE PREPARATION AND OBSERVATION OF SAMPLES

1.0 Introduction

The specimen chamber in an electron microscope is held under conditions of high vacuum whilst in operation. Because of this any moisture within a specimen will evaporate and the escaping gas and evaporation may cause unwanted electrical discharge from the high voltage terminal. A loss of resolution due to scattering of the electrons by vapour molecules can also be expected. Thus the specimens either have to be satisfactorily dried or be specially prepared.

Specimen preparation techniques have been reported by a number of authors and excellent reviews are given by Tovey and Wong (1973) and by Lohnes and Demirel (1978).

2.0 Specimen preparation

2.1 Preparation of laboratory clays

The need for the removal of water from specimens has been explained. In the consolidated clays obtained from the experimental part of the study the moisture content is a significant proportion of the total volume. Because the natural moisture content lies within the plastic zone for the clay the removal of water causes shrinkage of the specimen. The stresses arising during shrinkage will most likely disturb the microstructure. The effects of sample disturbance have been discussed by Tovey (1970) and by Barden and

Sides (1971). To overcome this problem a freeze drying method was used similar to that reported by Tovey and Wong (1973). The specimen was frozen rapidly and then transferred to a high vacuum chamber to allow the ice to sublimate at the low temperature. Water removal was usually completed in 24 hours.

A fresh surface was exposed by cutting a groove in the specimen to facilitate fracturing. After mounting on stubs using silver 'dag' as a conductive adhesive the specimen was dusted with compressed air and its fracture surface peeled with adhesive tape to remove loose material. In order to prevent charging in the microscope the specimen was coated with a very thin (200-300 Å) layer of gold in a vacuum chamber. All specimens were kept in dessication jars.

Specimens were obtained from the mid-region of the consolidated sample so as to minimise edge effects. Knill et al (1976) found that at the drained end of the sample preferred orientation was strongest. It was considered essential to the study that this kind of additional influence should be avoided.

2.2 Preparation of mudrocks

The moisture content of all mudrock samples was minimal and hence specimens cut for observation in the SEM could be oven-dried without fear of damage caused by shrinkage.

The major problem with the mudrock specimens was that to fracture the stronger ones a reasonable sized piece of rock had to be cut. This meant that further trimming to size contaminated the fracture surface. This problem was overcome by following the procedure illustrated in Fig. A12.

With the softer or more fissile samples a surgical blade was used to cut and trim the specimen. Tweezers were used to hold the loose layers together.

Once the specimen was trimmed to size it was mounted, cleaned and coated in the same manner as for the laboratory clays.

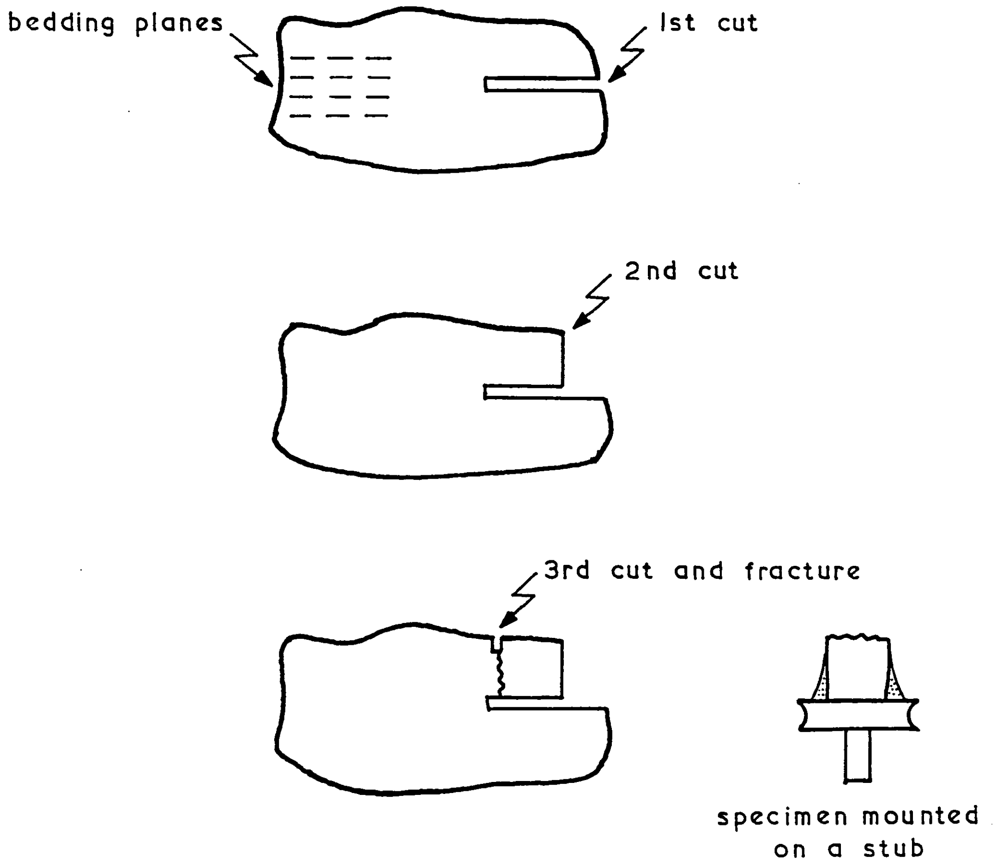

Figure A12. METHOD OF OBTAINING STUB SPECIMEN

3.0 Scanning Electron Microscopy

Because of the small size of particles within a specimen electron microscopy has to be used to observe the microstructure. The two instruments used in this study were a Philips PSEM 500 and a Jeol JSM 840A scanning electron microscope.

The scanning type of electron microscope forms its image from the electrons which are reflected back and emitted from the specimen due to surface bombardment by an electron beam. The beam is produced by an electron gun and is focussed by condenser lenses to a fine high energy beam. It is made to scan the specimen by deflector coils. The electrons issuing from the specimen are collected by a detector and then transmitted to two cathode ray tubes. One is used for viewing the image and the other is a high resolution screen for photographic recording.

Movement of the specimen is controlled by dials which are connected via mechanical linkage to the specimen stage. Two dimensionl 'horizontal' movement (x, y) is provided this way. Vertical movement is effected by manipulation of the microscope geometry. Rotation of the specimen is also possible with some instruments and this allows 3D imagery to be achieved.

The addition of energy-dispersive and wavelength spectrometers enables quantitative data to be extracted on the mineral composition of specimens. Developments in this field and in back scattered electron detectors ensures a bright future for research in the fine-grained sediments.

MUDROCK SAMPLE LOCATIONS

During the course of this study samples of the following mudrocks were observed to assess the particle orientation within the sample. A number of SEM micrographs have been presented in Chapter 5.

Sample name	Geographical location	O.S. Grid reference
Bituminous Shales	Saltwick Bay	NZ 917114
Cautley Mudstone	Ingleton	SD 695747
Cement Shales	Saltwick Bay	NZ 917114
Cleveland Ironstone	Port Mulgrave	NZ 798175
Dolwyddelan Shale	Dolwyddelan	SH 721522
Freshwater Shale 1	Bradfield Moors	SK 243900
Freshwater Shale 2	Ughill	SK 260902
Hard Shales	Saltwick Bay	NZ 917114
Jet Rock	Saltwick Bay	NZ 917114
Kimmeridge Clay	Kimmeridge Bay	SY 907792
Littleham Mudstone	Littleham Cove	WY 038803
Ludlovian Shale	Malham	SD 912656
MCM Mudstone	Rotherham	SK 445947
MCM Shale	Rotherham	SK 445947
Onny Shale	Craven Arms	SO 426 853
Oxford Clay	Calvert	SP 690235
Pot Clay	Bradfield Moors	SK 243900
Red Shale	Caerfai Bay	SM 762244
Siderite Mudstone	Saltwick Bay	NZ 917114
Speeton Clay	Speeton Cliffs	TA 151759

Stockdale Shale	Kentmere	NY 446043
Tertiary Mudstone	Zakynthos, Ionian Islands, Greece	
Yoredale Shale	Fountains Fell	SD 858722

Stratigraphical and depositional details of mudrocks presented in Chapter 5.

Tertiary	Greek Mudstone	Shallow marine basins - oxic
Cretaceous	Speeton Clay	Shallow sea - anoxic
Jurassic	Kimmeridge Clay	Shallow sea - fluctuating anaerobic
	Oxford Clay	Shallow sea - oxic
	Bituminous Shale	Shallow sea - fluctuating anoxic
Permo-Triassic	Littleham Mudstone	Overbank muds - oxic
Carboniferous	Coal Measure Shales	Paludal - highly organic, anaerobic
	Yoredale Shales	Prodeltaic - oxic
Devonian	no sample	
Silurian	Stockdale Shale	Deep troughs - anoxic
Ordovician	Cautley Mudstone	Calcareous shelf sea - oxic
	Dolwyddelan Shale	Shallow troughs - anaerobic
Cambrian	Red Shale	Shelf sea - oxic

PROGRAM STAT1

```
      IMPLICIT DOUBLE PRECISION (A-H,O-Z)
      INTEGER A
      BYTE DATFIL(10)
      EXTERNAL FCT
      COMMON T,DIRMN,Z
      DIMENSION IFR(12),A(50),MP(12),INTS(12),INTF(12)
      DIMENSION ACF(12),ASF(12),AUX(12)
      DIMENSION NOBS(12),E(12),XL(12),XU(12),P(12),NXL(12),NXU(12)
      DATA IFR/12*0/
      TYPE *,' ENTER DATA FILENAME'
      READ(5,10) DATFIL
   10 FORMAT(10A1)
      OPEN (UNIT=1,FILE=DATFIL,TYPE='OLD')
      READ (1,*) A
      CLOSE (UNIT=1)
      DO 15 I=1,50
         IF(A(I).GE.1.AND.A(I).LE.15) IFR(1)=IFR(1)+1
         IF(A(I).GE.16.AND.A(I).LE.30) IFR(2)=IFR(2)+1
         IF(A(I).GE.31.AND.A(I).LE.45) IFR(3)=IFR(3)+1
         IF(A(I).GE.46.AND.A(I).LE.60) IFR(4)=IFR(4)+1
         IF(A(I).GE.61.AND.A(I).LE.75) IFR(5)=IFR(5)+1
         IF(A(I).GE.76.AND.A(I).LE.90) IFR(6)=IFR(6)+1
         IF(A(I).GE.91.AND.A(I).LE.105) IFR(7)=IFR(7)+1
         IF(A(I).GE.106.AND.A(I).LE.120) IFR(8)=IFR(8)+1
         IF(A(I).GE.121.AND.A(I).LE.135) IFR(9)=IFR(9)+1
         IF(A(I).GE.136.AND.A(I).LE.150) IFR(10)=IFR(10)+1
         IF(A(I).GE.151.AND.A(I).LE.165) IFR(11)=IFR(11)+1
         IF(A(I).GE.166.AND.A(I).LE.180) IFR(12)=IFR(12)+1
   15 CONTINUE
   16 CLOSE (UNIT=1)
      MP(1) = 15
      INTS(1) = 1
      INTF(1) = 30
      DO 20 I=2,12
         MP(I) = MP(I-1) + 30
         INTS(I) = INTS(I-1) + 30
         INTF(I) = INTF(I-1) + 30
   20 CONTINUE
      TYPE 25
   25 FORMAT('1',13X,'INTERVAL',3X,'MID-POINT',3X,'FREQUENCY')
      DO 35 I=1,12
         TYPE 30, INTS(I),INTF(I),MP(I),IFR(I)
   30    FORMAT(' ',13X,I3,'-',I3,7X,I3,9X,I2)
   35 CONTINUE
      CFSUM = 0.0
      SFSUM = 0.0
      FSUM  = 50.0
      DO 45 I=1,50
         RA = (A(I)*2.0)/57.29577951
         ACF(I)=COS(RA)
         ASF(I)=SIN(RA)
         CFSUM =CFSUM + ACF(I)
         SFSUM =SFSUM + ASF(I)
   45 CONTINUE
      CBAR=CFSUM/ FSUM
      SBAR=SFSUM/ FSUM
      RBARD=(CBAR*CBAR+SBAR*SBAR)**0.5
```

```
      XBARD=ATAN(SBAR/CBAR)
      XBARD=XBARD*57.29577951
      IF (SBAR.GT.0.0.AND.CBAR.GT.0.0)XBARD=XBARD+0.0
      IF (CBAR.LT.0.0)XBARD=XBARD+180.0
      IF (SBAR.LT.0.0.AND.CBAR.GT.0.0)XBARD=XBARD+360.0
      XBAR=XBARD/2
      SZERO=1.0-RBAR
      TYPE 50
      TYPE 60,CBAR,SBAR,SZERO,RBARD,XBARD
   50 FORMAT('0',5X,'CBAR',5X,'SBAR',6X,'SZERO',5X,'RBARD',5X,'XBARD')
   60 FORMAT(1X,5F10.4)
      CHISQ = 0.0
      TYPE 65
   65 FORMAT('$HOW MANY CELLS ?')
      ACCEPT *,NG
      TYPE 68
   68 FORMAT('$UPPER VALUE OF K ?')
      ACCEPT *,AKU
      TYPE 70
   70 FORMAT('$LOWER VALUE OF K ?')
      ACCEPT *,AKL
      CRBARD = RBARD*100
      ICRBARD = CRBARD
      DRBARD = CRBARD-ICRBARD
      T = (AKU-AKL)*DRBARD + AKL
      TYPE *,T
      DIRMN = XBARD
      IFAIL = 1
      RI0 = S18AEF(T,IFAIL)
      IF (IFAIL.NE.0) THEN
         TYPE *, ' ERROR IN BESSEL FUNCTION I0'
         STOP
      END IF
      DIRMN = DIRMN/57.29577951
      Z = RI0*2*4*ATAN(1.0)
      TYPE 72
   72 FORMAT(' INPUT INTERVAL LIMITS & OBS.FREQUENCY')
      DO 80 I=1,NG
         ACCEPT *, NXL(I),NXU(I),NOBS(I)
         XL(I)=NXL(I)/57.29577951
         XU(I)=NXU(I)/57.29577951
         EPSR=0.0001
         IFAIL = 1
         NLIMIT = 0
         Y = D01AHF(XL(I),XU(I),EPSR,NPTS,RELERR,FCT,NLIMIT,IFAIL)
         IF (IFAIL.NE.0) THEN
            TYPE *, ' ERROR IN VON MISES'
            STOP
         END IF
         E(I)=Y*50
         P(I)=(NOBS(I)-E(I))**2/E(I)
         CHISQ = CHISQ + P(I)
   80 CONTINUE
   85 TYPE 90
   90 FORMAT('0',10X,'INTERVAL',2X,'OBS',4X,'EXP',3X,'(O-E)**2/E')
      DO 95 I=1,NG
         TYPE 100, NXL(I),NXU(I),NOBS(I),E(I),P(I)
  100    FORMAT(' ',10X,I3,'-',I3,3X,I2,3X,F5.2,3X,F6.4)
```

```
 95 CONTINUE
    TYPE 110, CHISQ
110 FORMAT('0',5X,'CHI-SQUARE = ',F7.4)
    STOP
    END
    DOUBLE PRECISION FUNCTION FCT(X)
    IMPLICIT DOUBLE PRECISION (A-H,O-Z)
    COMMON T,DIRMN,Z
    FCT=EXP(T*COS(X-DIRMN))/Z
    RETURN
    END
```

```
10 REM A TIME CONTROLLED PROGRAM FOR MONITORING CONSOLIDATION TESTS
20 REM FOR SLUMBERGER 7066 AND 7010 SCANNERS
25 REM MAX SCANS = 100 ,MAX CHANNELS = 10
30 REM FOR APPLE IIe MICROCOMPUTER
40 REM S1 = NO OF SCANS  :  C1 = NO OF CHANNELS
50 PRINT"CONSOLIDATION TEST PROGRAM : NOTES FOR USE":PRINT
55 PRINT"1  INITIAL SCAN RECORDED AUTOMATICALLY":PRINT
60 PRINT"2  START CONSOLIDATION WHEN INITIAL SCAN COMPLETED":PRINT
65 PRINT"3  REMOVE PROGRAM DISK ONLY AFTER TIME INTERVALS READ"
70 PRINT:PRINT:INPUT"PRESS RETURN TO CONTINUE";Z$
80 DIM A(10,100),C1$(10),IBS(100),T$(100)
85 REM FOLLOWING DATA FOR 4 CHANNELS READ IN FIXED ORDER
90 C1 = 4
100 C1$(1) = "C10"
110 C1$(2) = "C11"
120 C1$(3) = "C12"
130 C1$(4) = "C13"
140 REM READ TIME INTERVALS AS SET BY 'STINT' PROGRAM
150 HOME:VTAB(5):PRINT"READING TIME INTERVALS - PLEASE WAIT"
160 D$ = CHR$(4)
170 PRINT D$;"OPEN INTERVAL"
175 PRINT D$;"READ INTERVAL"
180 FOR I = 1 TO 40
190 INPUT IBS(I)
200 NEXT I
210 INPUT S1
220 PRINT D$;"CLOSE INTERVAL":HOME
230 VTAB(8):PRINT"INITIALISING DATASTORE - PLEASE WAIT"
240 N$ = CHR$(14)
250 PRINT D$;"PR#2"
260 PRINT CHR$(24)
270 PRINT D$;"PR#0"
280 FOR W = 1 TO 3000: NEXT W : REM TIME DELAY
290 PRINT D$;"PR#2"
300 PRINT "K1" + N$
310 PRINT CHR$(25)
320 PRINT "MOROTOF101H1N1"
325 PRINT D$;"PR#0"
330 VTAB(12):PRINT "DATASTORE NOW READY"
340 INPUT "PRESS RETURN TO START PROGRAM - START TEST AFTER SCAN 0";Z$
345 PRINT:PRINT:PRINT"REMOVE PROGRAM DISK"
350 GOSUB 1000
360 ST = SEC : REM START TIME
370 FOR W = 0 TO S1
380 ET = IBS(W) * 60
390 RT = ST + ET ;REM READING TIME
400 IF RT = ST THEN 440
410 GOSUB 1000
420 CT = SEC : REM CURRENT TIME
430 IF CT < RT THEN 410
440 T$(W) = T$
450 PRINT "SCAN NUMBER";W
460 FOR R = 1 TO C1
470 PRINT D$;"PR#2"
480 PRINT C1$(R) + "G;"
490 PRINT D$;"PR#0"
500 PRINT D$;"IN#2"
510 INPUT A(R,W)
520 PRINT D$;"IN#0"
```

```
 530 NEXT R
 540 IF W = S1 THEN 570
 550 NEXT W
 560 HOME
 570 INPUT "ALL SCANS COMPLETED - PRESS RETURN TO CONTINUE";Z$ : HOME
 580 INPUT "INSERT DATA STORAGE DISK - PRESS RETURN WHEN READY";Z$
 590 PRINT "WRITING DATA TO DISK - PLEASE WAIT"
 600 GOSUB 2000
 610 HOME : PRINT "DUMPING COMPLETED - REMOVE DATA DISK"
 620 PRINT "TEST COMPLETED"
 630 END
 990 REM ***********************************************
1000 REM *** SUBR - GET TIME ***
1010 D$ = CHR$(4) : SLOT = 4
1020 REM NOT LEAP YEAR LY = 0 , LEAP YEAR LY = 1
1030 LY = PEEK (49280 + 16 * SLOT)
1040 LY = INT(LY/64)
1050 IF LY > 1 THEN LY = LY - 2
1060 SLOT = 4 : PRINT D$;"NOMON I,O,C"
1070 PRINT D$;"IN#";SLOT
1080 PRINT D$;"PR#";SLOT
1090 INPUT " ";T$
1100 PRINT D$;"IN#0"
1110 PRINT D$;"PR#0"
1120 HOME : PRINT "REAL TIME = ";T$
1130 REM  CONVERT TO SECONDS
1140 MT = VAL (MID$(T$,1,2))
1150 D  = VAL (MID$(T$,4,2))
1160 H  = VAL (MID$(T$,7,2))
1170 M  = VAL (MID$(T$,10,2))
1180 S  = VAL (MID$(T$,13,6))
1190 REM CALCULATE DAYS TO DATE
1200 RESTORE
1210 DTD = 0
1220 FOR L = 1 TO MT
1230 READ J
1240 DTD = DTD + J
1250 NEXT L
1260 DATA 0,31,28,31,30,31,30,31,31,30,31,30,31
1270 REM ADD IN DAYS AND LEAP YEAR DAY
1280 DTD = DTD + D
1290 IF MT > 2 AND LY = 1 THEN DTD = DTD = 1
1300 REM FIND SECONDS TO DATE
1310 SEC = DTD*36400 + H*3600 + M*60 + S
1320 RETURN
1330 REM ***********************************************
2000 REM *** SUBR DUMP TO DISK ***
2010 REM FILE BASE NAME : DATA-CONSOL-
2020 REM CONVERSION CONSTANTS FOR TRANSDUCERS
2030 P1 = 10296
2040 D1 = 10.67
2050 P2 = 10252
2060 P3 = 10463
2070 FOR I = 0 TO S1
2080 A(1,I) = A(1,I) * P1
2090 A(2,I) = A(2,I) * D1
2100 A(3,I) = A(3,I) * P2
2110 A(4,I) = A(4,I) * P3
2120 NEXT I
```

```
2130 REM DUMPING
2140 HOME : F$ = "DATA-CONSOL-"
2150 INPUT "FILE NAME = DATA-CONSOL-";E$
2160 PRINT D$;"OPEN ";F$;E$
2170 PRINT D$;"DELETE ";F$;E$
2180 PRINT D$;"OPEN ";F$;E$
2190 PRINT D$;"WRITE ";F$;E$
2200 PRINT F$ + E$
2210 PRINT C1 : PRINT S1
2220 FOR L = 1 TO C1
2230 PRINT C1$(L)
2240 NEXT L
2250 FOR Y = 0 TO S1
2260 PRINT T$(Y)
2270 PRINT IBS(Y)
2280 FOR Z = 1 TO C1
2290 PRINT A(Z,Y)
2300 NEXT Z
2300 NEXT Y
2320 PRINT D$;"CLOSE ";F$;E$
2340 RETURN
```