

# Clay mineralogy and shale instability: an alternative conceptual analysis

M. J. WILSON<sup>1,\*</sup> AND L. WILSON<sup>2</sup>

<sup>1</sup> James Hutton Institute, Craigiebuckler, Aberdeen, and <sup>2</sup> Corex(UK) Ltd, Howe Moss Drive, Aberdeen, UK

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**ABSTRACT:** The instability of shales in drilled formations leads to serious operational problems with major economic consequences for petroleum exploration and production. It is generally agreed that the nature of the clay minerals in shale formations is a primary causative factor leading to their instability, although the exact mechanism involved is more debateable. Currently, the principal cause of shale instability is considered to be volume expansion following the osmotic swelling of Na-smectite. However, illitic and kaolinitic shales may also be unstable, so that interlayer expansion cannot therefore be considered as a universal causative mechanism of shale instability. This review considers alternative scenarios of shale instability where the major clay minerals are smectite, illite, mixed-layer illite-smectite (I/S) and kaolinite respectively. The influence of interacting factors that relate to shale clay mineralogy such as texture, structure and fabric are discussed, as are the pore size distribution and the nature of water in clays and shales and how these change with increasing depth of burial. It is found from the literature that the thickness of the diffuse double layer (DDL) of the aqueous solutions associated with the charged external surfaces of clay minerals is probably of the same order or even thicker than the sizes of a significant proportion of the pores found in shales. In these circumstances, overlap of the DDLs associated with exposed outer surfaces of clay minerals on opposing sides of micropores (<2 nm in diameter) and mesopores (2–50 nm in diameter) in a lithostatically compressed shale would bring about electrostatic repulsion and lead to increased pore/hydration pressure in smectitic, illitic and even kaolinitic shales. This pressure would be inhibited by the use of more concentrated K-based fluids which effectively shrink the thickness of the DDL towards the clay mineral surfaces in the pore walls. The use of soluble polymers would also encapsulate these clay mineral surfaces and so inhibit their hydration. In this scenario, the locus of action with respect to shale instability and its inhibition is moved from the interlamellar space of the smectitic clays to the charged external surfaces of the various clay minerals bounding the walls of the shale pores.

**KEYWORDS:** shale instability, smectitic shale, illitic shale, kaolinitic shale, shale water, diffuse double layer, shale pores.

The problem of wellbore instability in shales<sup>1</sup> is well known to drilling engineers as these rocks make up over 75% of drilled formations. It is one of the most significant technical problems in petroleum exploration and a major source of lost time and revenue.

More than 90% of wellbore instability problems are caused by problematic shales (van Oort *et al.*, 1996). Nearly twenty years ago it was estimated that these problems cost the industry at least half a billion

\* E-mail: jeff.wilson@hutton.ac.uk  
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<sup>1</sup> The term “shale” is used in this paper in a broad sense as denoting a fine-grained sedimentary rock with a relatively high clay content, as currently understood in the oil industry.

dollars per annum (Anderson *et al.*, 2010), and the probable current cost approaches or may even exceed a billion dollars per annum.

Shale instability manifests itself in a number of different ways, leading in turn to a variety of different problems. Thus, the wellbore may collapse through caving, sloughing or heaving, inevitably leading to enlarged holes. Cuttings from the drilled shale may disintegrate and disperse through the drilling fluid, or the shale may agglomerate around the drill bit (bit-balling) and drill pipe, accreting onto the walls of the wellbore and significantly reducing its diameter. These problems result in tight holes and stuck drill pipes that may even lead to hole abandonment. Indirect problems include clogging of surface flow lines and shakers following shale dispersion and disintegration, lost circulation, as well as difficulties in logging and in running casing.

Clay mineral composition is often thought to be a major causal factor in shale instability, although there are usually a variety of interacting mechanical factors which may exacerbate the situation. These may be associated with the drilling operation itself including, for example, contact between the drill string and the shale formation, fluid erosive action and pressure surges. Also the distribution of the overall *in situ* vertical and horizontal stresses, and particularly the existence of overpressure, may be major or contributory factors in causing such instability. Nevertheless, the nature of the clay minerals that make up shales, together with the overall shale texture, structure and fabric, remain as the most often cited primary causes of wellbore instability. The fundamental cause of this instability is considered to be the hydrophilic and charged nature of clay minerals which enables them to swell and participate in cation exchange reactions. For expandable clays, such as smectites, this results in

an increase in swelling pressure, either directly through hydration of the clay structure or indirectly where cation exchange increases osmotic pressure and where the shale itself acts as a semi-permeable membrane. A recent review of clay swelling (Anderson *et al.*, 2010) maintained that osmotic swelling of Na-smectites at the interlamellar level was the principal cause of shale instability during drilling operations. Although this mechanism is certainly important, it cannot be a universal cause of shale instability, as many problem shales are non-smectitic. The main objectives of this review are, therefore, to critically examine the present widely accepted mechanism for shale instability based on the swelling of smectitic clays and to put forward an alternative conceptual model relating clay mineralogy to shale instability that does not involve such interlamellar expansion.

## CLAY MINERALOGY AND SHALE INSTABILITY

### *Historical*

O'Brien & Chenevert (1973) were among the first to try to directly relate the instability of shales to their clay mineral composition. They classified problem shales into five main categories, where clay mineralogy was related to their relative hardness and their tendency to hydrate and disperse, thus leading to problems such as sloughing, caving, dispersion and bit-balling (Table 1). It may be noted from this classification that the clay minerals considered to be most active in causing shale instability are smectite, illite and mixed-layer clays (primarily mixed-layer illite/smectite, I/S). Chloritic clay minerals are considered to be of secondary importance and kaolinite is not mentioned, thereby implying that it is relatively inactive.

TABLE 1. Problem shale classification according their characteristics and clay mineralogy (after O'Brien & Chenevert, 1973).

Class	Characteristics	Clay minerals
1	Soft, highly dispersive (Gumbo). Mud making.	High smectite, some illite
2	Soft, fairly dispersive. Mud making.	High illite, fairly high smectite
3	Medium hard, moderately dispersive, sloughing.	High in mixed-layer, illite, chlorite
4	Hard, little dispersion, sloughing	Moderate illite, moderate chlorite
5	Very hard, brittle, no dispersion, caving	High illite, moderate chlorite

TABLE 2. Quantitative clay mineral composition (%) and H<sub>2</sub>O content of specific North American problem shales (after O'Brien & Chenevert, 1973)

Shale	Class	H <sub>2</sub> O (%) RH 50%	Smectite	Illite	Mixed-layer I/S	Chlorite
Anahuac	1	>4.0	40.4	5.5	—	—
Vermilion	2		25.4	42.0	—	6.7
Atoka	3		—	38.8	18.2	13.0
Midway	3	2.4–2.8	—	35.0	15.0	15.0
Wolfcamp	4	1.5–2.0	—	14.8	—	3.2
Canadian Hard	5	0.4–1.0	—	48.3	—	8.3

O'Brien & Chenevert (1973) illustrated the utility of their classification by quantitatively analysing the clay mineral composition of specific North American shales which had recognizable problem characteristics (Table 2). Thus, the clay mineralogy of the Anhuac Shale (Class 1) is dominated by smectite (40%), forms a so-called "gumbo" clay when drilled and gives rise to a wide variety of problems including bit-balling and dispersion. The Vermilion Shale (Class 2), which like the Anhuac Shale is also highly dispersive although without forming a gumbo clay, is dominated by illitic clay (42%) with significant amounts of smectite (25%). The Class 3 Atoka and Midway Shales contain no smectite but are dominated by illite and mixed-layer I/S clays and are prone to moderate dispersion and sloughing. The Class 4 Wolfcamp Shale is hard, shows little tendency to disperse while being prone to sloughing. It contains no smectite or mixed-layer clay and only moderate amounts of illite (15%). In contrast, the Class 5 Canadian Hard Shale shows no dispersion but is prone to caving, yet is dominated by illite (48%) with significant amounts of chlorite (8%). Table 2 also shows that the water content of the shales at 50% relative humidity decreases steadily from Class 1 to Class 5.

Where smectite is a major or significant clay component of a shale, as in Classes 1 and 2, then the shale instability can be readily rationalized in terms of the well known swelling behaviour of the smectite minerals in water and when saturated with different cations (Mooney *et al.*, 1952; Norrish, 1954). Thus, Wyoming montmorillonite saturated with strongly hydrated divalent cations like Ca<sup>2+</sup> and Mg<sup>2+</sup> yields interlayer basal spacings of ~15 Å over a wide range of relative humidities (32–79%) associated with a double layer of water, whereas saturation with the less strongly hydrated monovalent K<sup>+</sup> and NH<sub>4</sub><sup>+</sup> ions yields smaller interlayer spacings of ~12.0–12.4 Å related to a single water layer under the same conditions (MacEwan & Wilson, 1980). Such observations are entirely consistent with the inhibiting effects of K-based drilling fluids on shale instability. Many unstable shales have a clay mineralogical composition approaching that of Class 2 and for such shales O'Brien & Chenevert (1973) clearly demonstrated that K-based fluids dramatically affected their swelling and dispersion behaviour, particularly when the fluid included a polymer (Table 3). Comparison of the inhibiting effects of various chloride solutions on the linear swelling of Class 3

TABLE 3. Swelling and dispersion behaviour of Class 2 shale in various fluids (after O'Brien &amp; Chenevert, 1973).

Solution	% Linear swelling	Appearance	% Shale recovery
Water	—	Total disintegration	1.3
10% CaCl <sub>2</sub>	2.18	Partial disintegration	5.0
10% NaCl	2.00	Intact, easily crumbled	8.8
10% KCl	1.49	Intact, firm	46.0
10% KCl +Polymer	0.0	Intact, firm	91.6

shales again demonstrated the effectiveness of the  $K^+$  cation compared with  $Ca^{2+}$ ,  $Mg^{2+}$  and  $Na^+$  (Table 4). It should be noted, however, that such shales contained no discrete smectite, although smectitic layers do occur in the mixed-layer clays which, with illite, dominate the composition of the clay fraction.

### Mechanisms

In attempting to elucidate the role of clay mineralogy in relation to the instability of shales, emphasis is almost always placed on the expandability of smectite, particularly when saturated with  $Na^+$  (Anderson *et al.*, 2010 and references therein). There is currently a wealth of evidence derived from both experimental and modelling techniques that has characterized the nature of swelling in smectite at increasing levels of detail. In analysing the relationship between smectite swelling and shale instability, Van Oort (2003) described what is considered to be a typical situation where the swelling pressure following the interlamellar expansion and hydration of smectitic clay minerals, when combined with pore pressure, overcomes *in situ* vertical and horizontal stresses, and any cementation bonds holding the mineral particles together. Essentially the same mechanism was thought to be involved where shale instability is manifested by the dispersion of cuttings. Thus, cuttings travelling up the annulus are relieved of hydrostatic pressure so that dispersion will occur if swelling and pore pressures exceed mud pressure and the binding force of cementation between the shale mineral constituents.

This interpretation attributes a vital role to the physico-chemical activity within the interlamellar

space of the expandable clay minerals derived primarily from the hydration/solvation of the clay surfaces and exchangeable cations in the interlamellar space, thus creating a “hydration stress/pressure” or “swelling stress/pressure”. There are two types of swelling that can occur in smectitic minerals (Norrish, 1954). The first is “crystalline” swelling where expansion of the interlayer spacing takes place in more or less discrete steps which are related to the number of water layers in the interlamellar space. A one-water-layer structure yields a basal spacing of  $\sim 12.5 \text{ \AA}$ , a two-water-layer structure a spacing of  $\sim 15 \text{ \AA}$ , and a three-water-layer structure a basal spacing of  $\sim 20 \text{ \AA}$ . The second type is “osmotic” swelling and in the natural environment is thought to occur only when the smectitic clay is  $Na^+$ -saturated and where the external fluid is at very low ionic concentrations. In osmotic swelling, the basal spacing of the swollen smectite usually exceeds  $40 \text{ \AA}$  and may reach much higher values. As indicated above, osmotic swelling of  $Na^+$  smectite is considered to be the main cause of shale instability.

It is evident, therefore, that swelling pressure should be highly clay specific and that the effectiveness of inhibitors in reducing swelling pressures should be different for different clays, with the strongest effects being confined to smectites and mixed-layer clays with expandable interlayers. There should be little effect on illite and other non-swelling clays. The swelling indices of four pure clay minerals which typically occur in shales were estimated by Bol *et al.* (1994) through a determination of the relationship between stress and void ratio (water content) by means of an odometer test. The swelling index measured in this test characterises the hydration stress of a particular clay mineral/fluid combination and the results indicate that inhibition essentially affects hydration of smectitic clays but not that of other clay minerals tested (Table 5).

Van Oort (2003) described the physicochemical nature of shale-water interactions by considering the transport phenomena involved between the wellbore and the shale formation. In considering separately the direct Darcy flow of water which is driven by hydraulic gradients, and the diffusion of solutes which is driven by the chemical potential gradient between the drilling fluid and the shale, it was proposed that the shale-fluid system could act as a “leaky osmotic membrane” that could sustain chemical osmosis. Thus, by using high-salinity

TABLE 4. Swelling behaviour of Class 3 shale with polymer/salt solutions (after O'Brien & Chenevert, 1973).

Solution	% Linear swelling (after 24 hr)	% Reduction (compared with $H_2O$ )
Saturated KCl	0.56	79.0
3% KCl	0.94	64.0
3% NaCl	1.68	36.0
3% $MgCl_2$	1.83	30.0
3% $CaCl_2$	1.57	40.0

TABLE 5. Swelling indices of clay minerals (after Bol *et al.*, 1994).

Fluid	Montmorillonite $C_s \times 100$	Illite $C_s \times 100$	Chlorite $C_s \times 100$	Kaolinite $C_s \times 100$
H <sub>2</sub> O	>70	4.4	2.2	9.9
1 M NaCl	13.1	4.0	2.2	10.4
1 M KCl	10.9	3.5	2.0	11.2

fluids (such as KCl, NaCl, CaCl<sub>2</sub>, KCOOH, etc.) it is possible to stimulate osmotic backflow of shale pore water towards the wellbore, effectively offsetting the hydraulic inflow of mud filtrate (van Oort, 1997). These studies are consistent with the accepted wisdom in the oil industry that if the salinity of water in the drilling mud is greater than that in the pores of the shale then osmosis acts to dehydrate and stabilize the shale. Conversely, if the salinity of the water in the pores of the shale is greater than that in the drilling mud then the shale may become more hydrated and destabilized.

Further refinements and additional details to the general concept of shale acting as a semi-permeable membrane, resulting in the shale becoming unstable through the development of osmotic pressure, have been described by others. Thus, Schlemmer *et al.* (2003) found that the interactions of shales with various drilling fluids developed pore pressures generally compatible with osmotic theory, but depending on their clay mineralogy, porosity, pore water salinity and especially the composition of the drilling fluid. Water-based muds yielded the lowest osmotic effect whereas silicate-based muds, which formed impermeable deposits or precipitates, as well as invert emulsions, produced the greatest effect.

Many other studies could be cited which appear to be consistent with the concept that the primary relationship between the clay mineralogy of shales and their instability involves the occurrence of expandable smectitic clays or mixed-layer I/S and that the mechanism responsible involves increased swelling/pore pressures consequent on the hydration and expansion of the interlayer spaces in these clays. Other studies also appear to be consistent with the concept that shale in the wellbore may act, albeit imperfectly, as a semi-permeable membrane, leading to a situation where the shale can be stabilized by the use of drilling fluids of appropriate chemical composition through a process of osmosis

and cation exchange, effectively contracting the interlamellar space of the smectitic clays and thereby reducing the swelling/pore pressure.

There are, however, some studies which are at odds with these concepts and in any case the above explanations cannot account for the instability of illitic shales which contain no expandable clays (Classes 4 and 5 in Table 1) or perhaps even shales with no discrete smectite and where the mixed-layer illite/smectite mineral may be predominantly illitic (Class 3 in Table 1).

#### *Studies critical of conventional concepts of shale instability*

The fact that illitic shales can be unstable (Table 1), where there is no possibility that destabilization reactions take place through reactions involving the interlamellar space, indicates that there must be other mechanisms causing instability, and these might also apply to smectitic shales in addition to those dominated by illite. It should also be noted that the physical evidence for interlamellar swelling, particularly of the osmotic kind, of smectitic minerals has been obtained primarily when the clays are in the dispersed state or in thin films. Whether such swelling is able to occur just as readily in relatively impermeable, compacted shales under basin conditions is a moot point. It is, therefore, a legitimate question to ask whether shales do actually swell (or shrink) downhole *in situ* when interacting with drilling fluids? This question was posed by Santarelli & Carminati (1995) and they concluded that the evidence that shale swelling does occur downhole was open to other interpretations. In particular, they considered that laboratory tests on shale cores and plugs that appeared to show swelling were influenced by capillary effects as a consequence of air being trapped after drying. It may also be noted that the commonly used linear swelling tests

TABLE 6. Mineralogy of drilled shale formations where only Shale "A" is unstable (after Santarelli &amp; Carminati, 1995).

Shale	Quartz	Total clay	% by weight of the total clay			
			Illite/smectite	Kaolinite	Illite	Chlorite
A	10	90	80	15	5	—
B	23	75	85	—	5	10

on rocks (such as those shown in Tables 3 and 4) involve crushed and powdered material where the original fabric of the rock is completely destroyed. Santarelli & Carminati (1995) also conducted simulation tests on small scale boreholes and showed that there was no difference between the response to inhibitive fluids such as KCl and non-inhibitive fluids such as lignosulfonate. They also stated that in their experience, field observations of shale behaviour were inconsistent with the swelling hypothesis. For example, in the field case study of two shales, identified only as A and B, only Shale A was unstable despite having a similar clay mineralogy in terms of I/S content (Table 6) to that of Shale B, which was stable. On the basis of such evidence as well as doubts concerning the reality of osmotic flow in shales, Santarelli & Carminati (1995) concluded that shales do not swell downhole *in situ*.

Similar doubts were expressed by Santos & Fontoura (1997) who also maintained that experiments showing swelling on unpreserved shale cores or reconstituted shale core material were invalid. In their experience, preserved shale samples did not usually react when immersed in water. It may be noted that Méring & Oberlin (1971) early demonstrated observable differences in the swelling of bentonitic samples when placed in water. Thus, pieces of Wyoming bentonite showed easily observable swelling, whereas bentonite from Camp Berteaux showed no swelling at all. Both samples consisted predominantly of montmorillonite. It was suggested that the higher porosity of the Camp Berteaux sample enabled swelling of the smectitic clay to take place without an overall volume increase of the sample. Santos & Fontoura (1997) found that where swelling of shale samples in the laboratory is observed it is related to the amount and distribution of water within the shale and is due to surface hydration, which affects all clay minerals, rather than to osmotic hydration, which

only affects swelling clays. They further pointed out that surface hydration exerts a high swelling pressure over a small volume compared with osmotic swelling of Na montmorillonite which exerts a low swelling pressure over a large volume.

Ballard *et al.* (1994) carried out a carefully designed experiment using radioactive tracers to measure the rate of water and ion transport through cores of a variety of British shales, in which water composition and applied pressure drop could be varied at will. Fluids were circulated past both ends of the cores in separate systems, radioactive tracers being added to the fluid at the front end of core, whereas the fluid in the back end of the core contained synthetic pore water (PW) with no tracers. Diffusion rates of ions from pore water and diluted pore water are shown in Table 7 and it is clear that there is little difference between them. If osmosis had occurred, the diffusion rate from 10% PW should be faster than that of PW because

TABLE 7. Diffusion Rates ( $1 \times 10^{-10} \text{ m}^2 \cdot \text{s}^{-1}$ ) from pore water and diluted pore water (after Ballard *et al.*, 1994).

Shale	Outlet fluid	Diffusion rate
London Clay	PW	1.90
	10%PW	1.80
Oxford Clay	PW	1.9
	10%PW	2.00
Tertiary A	PW	1.25
	1%PW	1.24
Tertiary B	PW	3.05
	1%PW	2.88
Kimmeridge	PW	0.17
	10%PW	0.17

of water mass transfer. Even Kimmeridge shale which has an exceptionally low porosity and therefore offers the best possibility of forming a semi-permeable membrane, showed no sign of osmosis. The results therefore indicate that the shales have not acted as semi-permeable membranes and that osmosis has not occurred. Under zero pressure, ion transport was found to be controlled by diffusion, rates depending on concentration gradients and porosity. Transport rates of water depended on shale type and at the drilled surface advection is the dominant process with rates depending on overbalance pressure and shale permeability.

Bostrom *et al.* (1998) and Horsrud *et al.* (1998a) experimented on the effect of KCl on preserved smectitic North Sea shales and also concluded that ion transport took place by diffusion and not osmosis. The mechanism of stabilization was considered to be by cation exchange and shale shrinkage. However, with increasing KCl concentrations, stability problems also increased so that it seems that an optimum KCl concentration exists for the stabilization of these shales.

It can be concluded from this brief review that the instability of shales during drilling operations cannot necessarily be accounted for by the volume expansion affecting the interlayer spaces of smectitic clay minerals and that shales may not act as semi-permeable membranes enabling osmotic hydration to occur. It is clear too that shale instability where the dominant clay mineral is illite cannot be accounted for by a mechanism related to smectitic swelling. Recent evidence has also emerged indicating that kaolinite-dominant shales can be highly unstable when drilled, and can give rise to bit-balling problems, so that here too a mechanism other than that of smectite interlayer expansion must be implicated (Carpacho *et al.*, 2004).

#### *Textural and structural factors*

In discussing the impact of clay mineralogy on shale instability there are a number of interacting factors to take into account, other than the nature of the clay minerals themselves. Most importantly these include the overall texture, structure and fabric of the shales, as well as the size distribution and connectivity of the shale pores, because it is these factors which determine the extent to which the drilling fluid can interact with the shale clay

minerals. In this context, shale “texture” may be taken to refer to the relative proportions of clay- (<2  $\mu\text{m}$ ), silt- (2–62  $\mu\text{m}$ ) and sand-size (>62  $\mu\text{m}$ ) particles that make up the shale. “Structure” refers to the arrangement of features and constituents of the rock which can be easily observed with the naked eye or with an optical microscope. Such features could include bedding planes, fissility, cleavage, sedimentary laminations, grading by particle size, microfractures, burrows, faults etc. Shale “fabric” refers to the spatial relationship between the clay mineral particles themselves as well as with non-clay mineral particles and the extent to which this creates voids or pores within the volume of the shale. These relationships can be most easily observed at the SEM scale in fine-grained rocks such as shales as illustrated in Fig. 1.

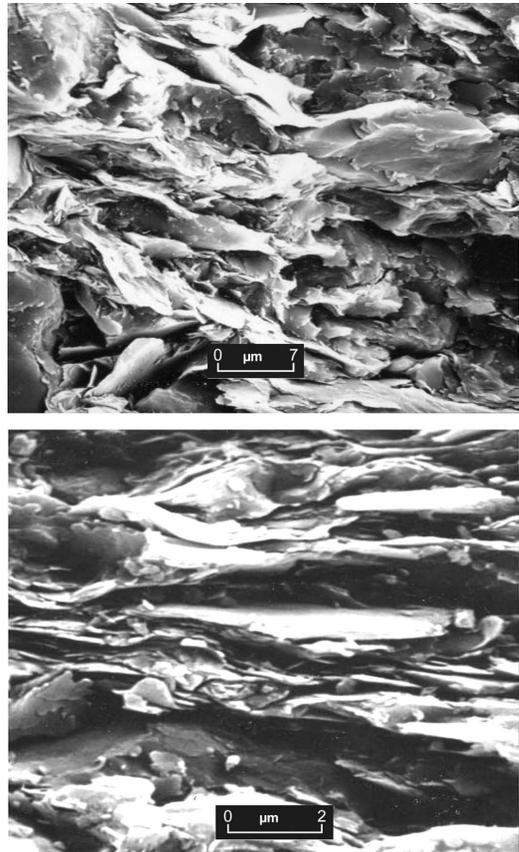


FIG. 1. Photomicrographs showing (upper) weakly oriented shale with open pore structure (inter-aggregate pores) and (lower) well oriented shale with slit and wedge-shaped pores between the platy clay aggregates.

It should be emphasized here that quantitative determination of the overall clay mineralogy of a shale, no matter how accurate, cannot on its own predict the susceptibility of the shale to instability problems.

*Shale texture.* The proportion of clay-size material in rocks designated as shales varies widely. For example, the clay contents of the North American shales studied by O'Brien & Chenevert (1973) ranged from ~17 to 70%. For North Sea shales Horsrud *et al.* (1998) recorded clay contents ranging from 32 to 82%, whilst for shales from Mexico and Brazil, Diaz-Perez *et al.* (2007) found clay contents ranging from ~17 to 66%. Clearly such variations in texture could have a significant impact on shale reactivity whatever the clay mineral composition of the rock.

*Shale structure.* It is becoming more widely appreciated that the structure of shales may vary considerably both between and within individual formations. They are, therefore, best regarded as heterogeneous materials, despite first impressions of a monotonous appearance. For example, an assumption that shales are relatively impermeable materials may not always be correct, bearing in mind that internal microfractures may act as efficient fluid conduits. Thus, Capuano (1993) found for the geopressured Oligocene shales of the Frio Formation in Texas, that microfractures supported a fluid flow comparable to that of the Frio Sandstone reservoir. Again, Gale *et al.* (2007) investigated the natural fractures in the Barnett Shale in the Fort Worth Formation, Texas, and concluded that these features were common at a range of scales. Some sets were relatively narrow, sealed with calcite and contributed little to enhance permeability, although they acted as planes of weakness and were able to be reactivated after hydraulic treatment. Other fractures were larger and occurred in widely spaced clusters so enhancing permeability at a local scale. Dewhurst *et al.* (1999) concluded that the permeability of mudstones may vary by ten orders of magnitude and even by three orders of magnitude at a single porosity. It may be anticipated therefore that variation in the nature, intensity and orientation of shale structures will ensure that for many shales the transmission of drilling fluids from the wellbore into the shale will occur preferentially at selected points, rather than in the radial and uniform way envisaged by van Oort (2003).

*Shale fabric.* The microfabric of shales is also relevant to their stability. Bennett *et al.* (1991)

reviewed the formation of shale microfabrics and showed how there is a continuum between physicochemical, bio-organic and compaction/diagenetic processes. In the context of shale instability, however, the most important point would be in whether the clay minerals were strongly oriented or arranged in a random fashion, forming anisotropic and isotropic fabrics respectively (Fig. 1). An oriented fabric would be typical of many shales and would be readily revealed by their laminated and fissile nature, in the main formed by a combination of sedimentation and burial compaction processes. Such shales may be susceptible to delamination and caving when in contact with water-based muds. Many shales, however, are of a massive nature with little or no sign of sedimentary laminations and when examined under the SEM show a randomly oriented clay fabric with many edge-to-face contacts leading to the creation of numerous voids. Such shales are often bioturbated or may even be classed as palaeosols as indicated by traces of rootlets and ped-like features. The ball clays described later exemplify such features to a high degree. In general, shales with randomly oriented fabric have a higher porosity than those with well oriented fabrics and consequently are more prone to disintegration and dispersion when hydrated.

#### *Pore size distribution and microstructure in clays and shales*

The pore size distribution of individual clay minerals after drying and experimental compaction shows significant differences dependent on their aggregate structure and particle size (Diamond, 1970; Vasseur *et al.*, 1995). This was well illustrated by a study of three clay materials of different mineralogy (Djéran-Maigre *et al.*, 1998). One clay contained major amounts of illite (77%), a second was smectitic (52%) and a third contained equal amounts (28%) of mixed-layer I/S, illite and kaolinite.

The illitic clay consisted of tiny individual crystals, ~0.15  $\mu\text{m}$  in diameter and 20 nm in thickness, which were randomly oriented and became somewhat better oriented with increasing stress, but not to the same extent as the clay with mixed mineralogy. The void ratio decreased from 1.40 to 0.5 as applied stress increased and TEM observations showed that the pores became very much smaller and that consequently hydraulic

conductivity was low both in the natural and compacted states. The smectitic clay had the most open microstructure of all the clays examined, although at 10 MPa applied stress all the pores observed in the initial state could no longer be observed under the TEM. Both illitic and smectitic clays in their initial state had a relatively more open structure than that of the clay with mixed mineralogy. This clay consisted largely of randomly oriented aggregates in which individual particles were disposed in a face-to-face manner, and showed inter-aggregate pores of up to 0.5  $\mu\text{m}$  in diameter. With compression up to 20 MPa the particle aggregate structure and the inter-aggregate pores had largely disappeared. Similar results for individual clay minerals were obtained by Iñigo *et al.* (2000) who also showed that particle orientation on compaction to 100 MPa strongly depended on the size of the clay mineral basal surface, so that aggregates of large (>10,000 Å), well crystallized kaolinite yielded a well oriented structure, whereas aggregates consisting of small particles (<1000 Å) of illite and smectite produced a poorly oriented more open structure.

Shales would naturally be expected to show a broader distribution of pore sizes than pure clay minerals because of their larger content of non-clay minerals. Thus, for North Sea shales ranging in age from Jurassic to Tertiary and of mixed clay mineralogy, Horsrud *et al.* (1998b) showed that 20% of the pore volume was found in the 6 to 12 nm size range. It would be anticipated, however, that with deeper burial and increasing compaction the finer pore sizes would become more predominant. This has been shown by Connell-Madore & Katsube (2006) in their study of Cretaceous and Palaeocene shales from the Beaufort and Mackenzie Basin in the Northwest Territories of Canada, representative of four different compaction zones. They showed a predominance of pores in the 4–10 nm size range where depth of burial was >2000 m, but where depth of burial was <2000 m, pore sizes showed a wider range and often peaked in the 0.1 to 1.0  $\mu\text{m}$  range. It is of interest that all shales found to be over-pressured in this study were dominated by pore sizes in the 4 to 10 nm range.

#### *Nature of water in clays and shales*

The location and state of water within clays and shales is an important consideration in the context of instability problems. For clay minerals deposited

in an aqueous environment it would be anticipated that they would be fully hydrated. Tardy & Touret (1987) found that the water content of smectitic clays increased exponentially as relative vapour pressure was increased from 0.96 to 1.0 and that not all the water at high relative humidities (above  $p/p_0 = 0.69$ ) could be accounted for in terms of interlamellar hydrates. This excess of water and its exponential increase upon saturation of the clay at  $p/p_0 = 1.0$ , regardless of the interlayer cations, was explained by the slow diffusion of “free” water into pores within the clay fabric. High-resolution TEM and small angle X-ray scattering studies by Touret *et al.* (1990) documented the partition of water over three kinds of pore spaces as inter-aggregate, intra-aggregate and inter-lamellar. Inter-aggregate pores range in size from 0.2  $\mu\text{m}$  to  $\sim 3 \mu\text{m}$ , whereas intra-aggregate pores are much smaller (0.01–0.2  $\mu\text{m}$ ) and are delineated by the boundaries of primary particles within the aggregates. Interlamellar pores occur between the silicate sheets of expandable clays, their size depending on the hydration of the interlayer cation. A tentative partition of water over these pores in several fully-saturated (at  $p/p_0 = 1.0$ ), powdered, Mg-saturated expandable clays (Table 8) shows a predominance of inter-aggregate and intra-aggregate water over that within the interlamellar spaces of all expandable clay minerals. Touret *et al.* (1990) considered that the water in inter-aggregate pores could be described as “free”, that within the intra-aggregate pores as “bound”, and that within the interlamellar space as “crystalline”.

In considering what happens to these different types of water when sedimented clay materials are deeply buried, compressed and lithified, it should be borne in mind that such materials are nearly always deposited as aggregates rather than as free particles (Bennett *et al.*, 1991). In general, it would be anticipated that during burial and compression, the more loosely bound inter-aggregate water would be squeezed out and lost, whilst the intra-aggregate water would be likely to be retained in the smallest pores and to show a relative increase in the sediment as a whole. The relative amount of intra-aggregate water compared with interlamellar water in a shale will clearly be highly variable even under conditions of uniform stress, depending on texture, structure and fabric, which will all affect porosity and permeability, as well as the relative amounts of expanding and non-expanding clays.

There is a dearth of information on the stability of interlamellar water in smectitic clays at the

TABLE 8. Partition (wt.%) of water over different types of pores in saturated, powdered, expandable clays where  $Mg^{2+}$  is the exchangeable cation (after Touret *et al.*, 1990).

Clay mineral	Inter-aggregate	Intra-aggregate	Inter-lamellar	Total amount of water (g) per gram of clay
Hectorite	25	40	35	1.46
Montmorillonite Wyoming	33	42	25	1.79
Montmorillonite Camp-Berteaux	58	20	22	2.08
Nontronite	52	24	24	1.72
Vermiculite	69	12	19	1.12

higher temperatures and pressures associated with increasing depth of burial (Anderson *et al.*, 2010). Early studies indicated that such clays are often converted to mixed-layer I/S during sediment burial, and it was generally considered that this transformation involved the ready loss of interlamellar water. However, work based both on modelling and experimentation has shown that the interlamellar water of smectites could be surprisingly stable under such conditions. Thus, Odriozola & Guevara-Rodríguez (2004), using Monte Carlo (MC) and molecular dynamics (MD) computer simulations, concluded that where the number of water molecules per clay sheet was  $\sim 40$  in the simulation box, then the one-layer hydrate of Na-montmorillonite yielding a basal spacing of 12.5 Å was stable to a depth of 6 km, corresponding to a temperature and pressure of 180°C and 90 MPa respectively. Similar results for Na-montmorillonite were also obtained by de Pablo *et al.* (2005) who also investigated the stability of the hydrates of K- and Ca-montmorillonite using MC simulation. It was found that the one-layer hydrates of Na-, K- and Ca-montmorillonites were stable at all temperatures (up to  $\sim 200^\circ\text{C}$ ) and pressures (up to  $\sim 100$  MPa) encountered to a depth of  $\sim 6.7$  km, assuming a normal geothermal ( $30^\circ\text{C}/\text{km}$ ) and lithostatic (15 MPa/km) gradient. Transient two-layer hydrates may exist at greater depths but will revert to the one-layer hydrate with any sudden change in temperature and pressure.

Experimental work has also shown that in K-poor systems the stability of montmorillonite in general, and its interlamellar water in particular, can be remarkably high. For example, Wu *et al.* (1997) studied the *in situ* dehydration of Ca- and Mg-montmorillonite at high pressures in a hydrothermal diamond anvil cell. They found that at pressures of

1000 MPa, dehydration of montmorillonite from a fully expanded 19 Å hydration state to a 15 Å hydration state occurred over a temperature range of 260–350°C for Ca-montmorillonite and at 200–250°C for Mg-montmorillonite. At higher pressures the conversion from a 15 Å hydrate to a 12.5 Å form took place at 590–605°C, leading Wu *et al.* (1997) to conclude that hydrated montmorillonite can be transported down into the subduction zone before its water is released.

#### *The diffuse double layer and clay minerals*

The diffuse double electrical layer theory is widely accepted as a realistic representation of the interaction of forces between clay and water/electrolyte systems. When a negatively charged clay particle is in contact with a fluid such as an aqueous electrolyte, the distribution of cations in the fluid is conceived as being relatively concentrated at the surface of the clay particle so as to maintain overall electrical neutrality. In an electrolyte the concentration of the counter-ions gradually decreases with distance from the charged clay particle until it becomes the same as that in the bulk fluid (Fig. 2). This distribution of cations and anions is referred to as the electrical or diffuse double layer (DDL). The thickness of the DDL depends on the magnitude and distribution of the charge at the clay particle surface and on the salinity of the fluid. All other things being equal, the higher the charge at the clay surface the thicker the DDL and the higher the salt concentration in the fluid the thinner the DDL.

Various estimates have been made of the thickness of the DDL in different chemical environments. Thus, Johnston & Tombác (2002) determined that for montmorillonite with a CEC of

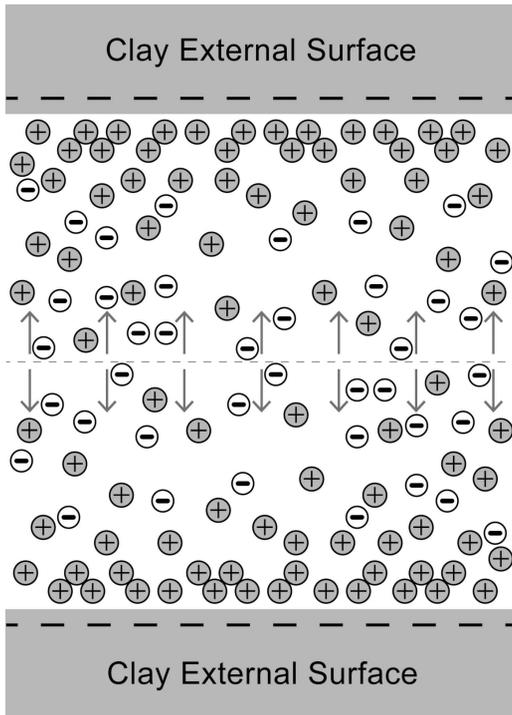


FIG. 2. Simplified sketch of the diffuse double layer (DDL) associated with the surface of clay minerals in shale pores indicating pressure generated by forced overlap of the DDL.

85 meq/100 g the thickness of the DDL in a solution of 0.001 M NaCl would be  $\sim 10$  nm ( $\sim 0.01$   $\mu\text{m}$ ). Mojid & Cho (2006) determined the thickness of the DDL of Na bentonite in distilled water, 0.001 M NaCl and 0.01 M NaCl to be 183, 19 and 6 nm respectively. For Ca bentonite in distilled water and the same NaCl concentrations the DDL thickness was estimated at 95, 10 and 3 nm respectively. DDL thicknesses for montmorillonite, illite and kaolinite in 0.0001 M KCl and LiCl were found to be similar at  $\sim 15$  nm by Sridharan & Satyamurty (1996).

It seems clear therefore that the thickness of the DDL associated with clay minerals could be of the same order or even thicker than a significant proportion of the pores found in shales. This being so it would be anticipated that external stresses following from sediment burial and compaction could often bring about a situation where the DDLs of clay minerals exposed at opposite sides of pores could be forced to overlap, thus leading to increased pore pressures as illustrated in Fig. 2.

## DISCUSSION

The above review has illustrated that the general hydrophilic nature of clays, in combination with the overall texture, structure and fabric of shales, plays a major role in causing instability of wellbores when these are exposed to aqueous fluids during drilling operations. The role played by the individual clay minerals in the context of instability mechanisms will now be discussed in further detail.

### *Smectitic shales*

The well known instability of smectitic shales during drilling operations appears in many cases to be convincingly related to the volume expansion consequent on the interlamellar swelling of the smectite minerals. It is not at all clear, however, that the principal cause of shale instability in general, and even of smectitic types in particular, is necessarily related to osmotic swelling of  $\text{Na}^+$ -saturated smectites as recently maintained by Anderson *et al.* (2010). Evidently, this mechanism cannot account for the instability of non-smectitic shales, and for smectitic shales there is a dearth of data relating to the stability of interlayer water in smectites following deep burial, as well as to the swelling capacity of these clays under basin conditions. The detailed nature of osmotic swelling, if it occurs in these circumstances, is certainly not known nor is it clear to what extent such swelling is able to spread throughout the body of the shale. Most swelling data on smectites have been obtained when the clay is in an aqueous colloidal suspension or has been deposited on glass slides as thin films. Here the charged sites within and on the clay mineral are readily accessible to exchangeable cations, but it is doubtful that such sites would be so accessible in a compacted, relatively impermeable shale and at the higher temperatures and pressures encountered at depth.

It may be argued though that even if osmotic swelling does not occur under basin conditions, the more limited crystalline swelling of smectites may still be a major mechanism in shale instability. If, however, the computer simulations of de Pablo *et al.* (2005) concerning the arrangement of water and Na, Ca and K cations in the interlayer space of montmorillonite under basin conditions represent reality, then the one-layer hydrate for all these cations will persist stably at the temperatures and pressures prevailing to a depth of  $\sim 6.7$  km. It is

difficult to see therefore that exchange of interlayer Na cations in the interlayer space of smectitic clays for K or Ca cations in the drilling fluid will necessarily involve a volumetric decrease of the smectite and a reduction of swelling pressure. Interlamellar swelling of highly smectitic shale may certainly occur in the way envisaged by Van Oort (2003), when the impermeable shale comes into direct contact with the drilling fluid at the exposed wellbore face, but in many cases a shale becomes relatively more permeable through the existence of micro-fractures, laminations, higher proportions of non-clay minerals and other heterogeneities, thus enabling the drilling fluid to gain access to the intra-aggregate pores more rapidly than to the inter-lamellar region of the smectites.

Nevertheless, if shale swelling through the interlamellar expansion of smectite clays does not occur at depth in sedimentary basins, then it is reasonable to ask why treatment with mixed KCl/polymer solutions has such an inhibiting effect on shale instability. It is widely accepted at present that such treatment is effective because the interlayer Na or Ca cations in the smectite exchange with the  $K^+$  cations in the drilling fluid, so forming a one-layer water structure in the interlamellar space and shrinking the basal spacing of the smectite from 15 Å to 12.5 Å. It is further thought that cationic polymers are able to penetrate into the interlayer space, effectively binding the 2:1 silicate layers together and thus further inhibiting the volume expansion which leads to shale instability (Anderson *et al.*, 2010).

An alternative scenario is that the inhibiting effect of mixed KCl/polymer solutions on smectitic shales is due to the effect of the shrinkage in the thickness of the DDL in micro- and meso-sized pores (<2 nm and 2–50 nm respectively), brought about by the more concentrated brine solutions, combined with the encapsulating effect of the polymer on the external hydrophilic clay surfaces exposed in the pore walls. This mechanism could be more important, especially where the shale is relatively permeable as described above. In this scenario, the locus of inhibiting activity with respect to unstable smectitic shales is moved from the interlamellar space in the smectite to the external surfaces exposed in micro- and mesopores within the shale.

Where a relatively impermeable smectitic shale is exposed at the wellbore face, however, then even a small volume increase in the interlayer may well

bring about rapid dispersion of the clay and loss of integrity of the shale, as well as having a disintegrating effect on cuttings as they pass up the annulus, as advocated by van Oort (2003).

### *Illitic shales*

By definition illites are non-swelling minerals, so that the instability of illitic shales cannot be related to interlamellar expansion in the same way as it has been for smectites. It should be noted, however, that the higher layer charge of illite compared with smectite will mean that the charge density on its external basal surfaces will be correspondingly higher than that of the external surfaces of smectite. This would in turn mean that the DDL associated with illite basal surfaces exposed in the pores of illitic shales would tend to be thicker than the DDL of smectite basal surfaces similarly exposed, all other factors being equal. Compression by lithostatic pressure of the DDL associated with illite, which would consist of loosely bound, hydrated ions on the basal surfaces of the clay mineral exposed in the shale micro- and meso-pores, is thus envisaged as leading to over-pressure and shale instability in the same way as described above for smectitic shales. Inhibitive treatments involving KCl/polymer solutions would also be effective in stabilizing illitic shales through the same mechanism as envisaged for smectitic shales, namely contraction of the DDL and encapsulation of the hydrophilic clay surface.

It is important to note that the nature of illite, particularly with respect to its crystallinity, particle size and thickness, in addition to associated chemical and physical properties such as cation exchange capacity, morphology, surface area and porosity, can be extremely variable. In soils and rocks, many illitic clays seem to be mixtures of comminuted detrital micas combined with more poorly-ordered 10 Å material. Thus, Lanson & Besson (1992) using a decomposition procedure for X-ray diffraction powder patterns concluded that the 10 Å material found in sediments of the Paris Basin actually consisted of three components, namely detrital mica, illite and a mixed-layer mineral. Meunier & Velde (2004) using the profile fitting procedure of Lanson (1997) similarly decomposed the broad 10 Å reflection occurring in some French illitic soil clays into separate components, which were interpreted as well crystallized illite, poorly crystallized illite and an illitic mixed-layer mineral. The (001) surfaces even of

coarse-grained well crystallized muscovite are capable of adsorbing structurally ordered aqueous films with associated hydrated cations up to 30 Å in thickness according to molecular dynamics simulations and experimental X-ray reflectivity data (Wang *et al.*, 2005). Such effects will become greater with decreasing particle size and increasing surface area so the activity of illitic shales with respect to instability should not be surprising.

#### *Illitic shales with mixed-layer I/S*

Many illites when examined by XRD after treatment with ethylene glycol show evidence that they contain expandable layers and these have been interpreted as smectite layers within the illite structure, so forming a mixed-layer illite-smectite (I/S). In very many cases the X-ray evidence on which this interpretation is based is a narrowing towards the low angle side of the first-order 10 Å basal reflection of illite after glycolation, compared with the width of this reflection in the air-dried state. Computer simulations of I/S such as those of Reynolds (1980), where the amount of smectite ranges between 5 to 10% in the mixed-layer structure, show that such a peak profile is indicative of a specific type of ordering in which every three illite layers is followed by a smectite layer. Thus, an IIS structure is formed which, using an ordering parameter from the German literature termed "Reichweite", is described as R3. The instability of illitic shales containing R3 I/S could therefore be accounted for by the occurrence of smectitic layers within the illite structure, the principal mechanism being volume expansion following cation exchange and hydration of the smectitic interlayers in the mixed-layer structure. This mechanism would, therefore, be essentially the same as that currently accepted for the instability of smectitic shales where the clay mineral occurs in discrete form.

There is, however, another interpretation of R3 I/S which views its diffraction effect as being due to the occurrence of extremely thin illite particles. These particles range in thickness from ~20 to 50 Å and are thought to precipitate in shales as a result of the progressive dissolution of smectite with increasing depth of burial. This reaction is involved in an alternative concept of the principal mechanism of the illitization of smectites during burial diagenesis of argillaceous sediments, referred to currently as dissolution/precipitation (D/P), and was pioneered by Nadeau *et al.* (1985).

McHardy *et al.* (1982) and Nadeau *et al.* (1984a,b) provided evidence that very thin illite particles (3 to 5 nm in thickness) dried down onto a glass slide and glycolated will diffract as though they contain smectite layers because the glycol is adsorbed between the thin illite particles. In other words the diffraction is an inter-particle effect and does not necessarily denote the existence of smectite layers.

It may be anticipated that the illite which yields an R3 I/S-type XRD pattern will be an extremely reactive component of the shale by virtue of its fine grain size as well as the nature of its external surfaces. Accepting that the layer charge in illite is symmetrically distributed around both sides of the 2:1 layer then it would seem inevitable that the external surface of an illite particle will be highly charged and that its charge density will be higher than that of the external surfaces of smectite. R3 I/S type illite exposed on the walls of micropores will therefore attract hydrated cations to its external surface and will have a thicker DDL than smectite, all other things being equal, and may cause shale instability due to hydration and swelling in micropores in the same way as smectite described above.

#### *Kaolinitic shales*

For many years kaolinite has been perceived as a non-reactive clay mineral with respect to shale instability problems, but more recent investigations suggest that kaolinitic shales do cause problems. Thus, Cliffe & Young (2008) considered that kaolinite was implicated in the bit-balling and accretion tendencies of a variety of North Sea shales. They found that the kaolinite content of these shales was positively correlated with the percent of solids accreted after 10 minutes interaction with a steel surface ( $R^2 = 0.7972$ ), whereas the I/S content of the shales was negatively correlated after a similar period of interaction ( $R^2 = -0.9655$ ). On the basis of accretion rates in the field and laboratory studies, they concluded that shales containing a higher percentage of illite and kaolinite clays are more susceptible to accretion and bit-balling when exposed to drilling in water-based fluids than shales dominated by mixed-layer I/S. The amount of shale accretion to a steel surface was time-dependent with the stronger correlations being found after 10 minutes compared with a 5 min exposure (Table 9). With increasing exposure time to 60 minutes, the correlation of accretion with

TABLE 9. Clay mineral content (%) in bulk shales and accretion to a steel surface after 5 and 10 minutes (after Cliffe &amp; Young, 2008).

Shale	Kaolinite	Illite	Illite/smectite	% A-5*	%A -10 <sup>#</sup>
Green Clay	8	2	65	20	72
London Clay	9	9	41	73	90
Oxford Clay	13	10	39	48	90
Foss Eikeland	1	18	21	76	101
Arne	34	25	19	72	110

\* % A-5 –per cent of accreted shale after 5 min test.

<sup>#</sup> % A-10 – per cent of accreted shale after 10 min test.

% kaolinite markedly improved whilst correlation with % illite deteriorated.

Van Oort (2003) assessed the potential of shales to bit-ball by considering in a general way the liquid/plastic limits, the so-called Atterberg limits, of the shale materials. Increasing the water content of an initially dry clay will first lead to a zone below the plastic limit where the material is too dry to have a significant tendency to stick. Above the plastic limit at higher water contents, however, the sticking tendency rapidly increases until at still higher water contents the liquid limit is reached such that the material has very limited inherent strength and will readily disperse (Mitchell, 1993). On this basis, therefore, the zone between the plastic and liquid limits of a clay material represents a clear “Danger Zone” for bit-balling. Determination of the plastic and liquid limits of individual clay minerals, as well as their plasticity indices (PI) (the difference between liquid and plastic limits) shows a wide range both within and between these minerals (Table 10).

The formations known as ball clays are of interest in the context of this review because they are highly plastic, form ball-shaped masses and

their clay mineralogy is dominated by kaolinite and illite (Shen, 1993). Although they occur relatively rarely in the stratigraphic column, it may be confidently predicted that their plastic properties are such that they would certainly give rise to accretion and other problems during drilling. The clay mineral content of the bulk materials ranged from 40 to 80% with kaolinite ranging from ~30 to 80% and illite from ~3 to 30% (Table 11). There was no discrete smectite in the ball clays, although ethylene glycol solvation sometimes caused a slight shift in the profile of the 10 Å illite reflection, suggesting a small amount of expandable interlayers. TEM examination of the clay fractions showed that they consisted primarily of thick subhedral to anhedral kaolinite particles, sometimes approaching a hexagonal morphology, accompanied by exceedingly fine-grained and thin lath-like illite (Fig. 3a,b). SEM examination of the ball clays showed in every case a distinctly randomly oriented fabric in which the isometric and subhedral particles of kaolinite and the tiny lath-like particles of illite could be easily observed (Fig. 4a,b). These mineral elements are so loosely packed that voids can be readily seen, which would enable external

TABLE 10. Liquid limits, plastic limits and plasticity indices of individual clay minerals (after Mitchell, 1993).

Mineral	Plastic limit (%)	Liquid limit (%)	Plasticity index
Montmorillonite	50–100	100–900	50–800
Illite	35–60	60–120	25–60
Kaolinite	25–40	30–110	5–70
Palygorskite	100–120	160–230	60–110
Chlorite	36–40	44–47	7–8

TABLE 11. Quantitative normative mineralogy of ball clays from England, China and Ukraine (after Shen, 1993).

Country	Sample	Kaolinite	Quartz	Illite	K-feldspar	Plagioclase feldspar	Total
England	97-2014	35.70	19.29	27.99	12.26	4.67	100.0
	97-2016	42.50	21.25	22.20	10.51	3.54	100.0
	97-2020	29.60	51.00	9.83	7.81	1.76	100.0
	97-2027	28.18	25.80	31.39	10.94	3.69	100.0
	97-2050	26.45	41.78	19.41	8.53	3.83	100.0
	97-2500	51.77	9.91	20.32	16.16	1.84	100.0
	97-2501	67.63	7.27	16.12	6.87	2.11	100.0
China	95-1964	82.92	11.05	0.00	4.64	1.39	100.0
	97-1065	35.48	30.02	18.99	11.16	4.35	100.0
	97-2234	42.53	26.15	27.74	2.50	1.62	100.0
	97-2193	57.94	25.47	3.22	9.59	3.78	100.0
Ukraine	DBX	61.48	12.01	17.14	7.87	1.50	100.0
	DBY	38.70	27.13	24.45	6.46	3.26	100.0

fluids to penetrate and interact with the clay particles with ease. The highly plastic properties of these ball clays are, therefore, quite understandable.

Carpacho *et al.* (2004) found that kaolinitic shales in South America were highly unstable and that the use of K-based drilling fluids increased that

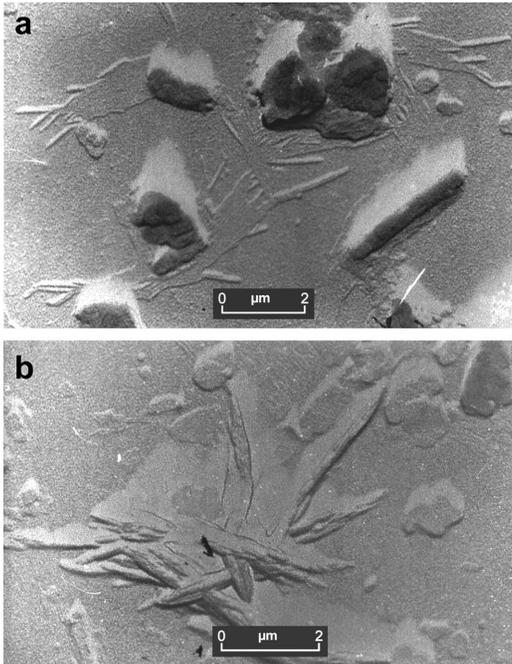


FIG. 3. TEMs of ball clays from (a) Devon and (b) Ukraine showing thick, chunky morphology of kaolinite and thin stubby, lath-like form of illite.

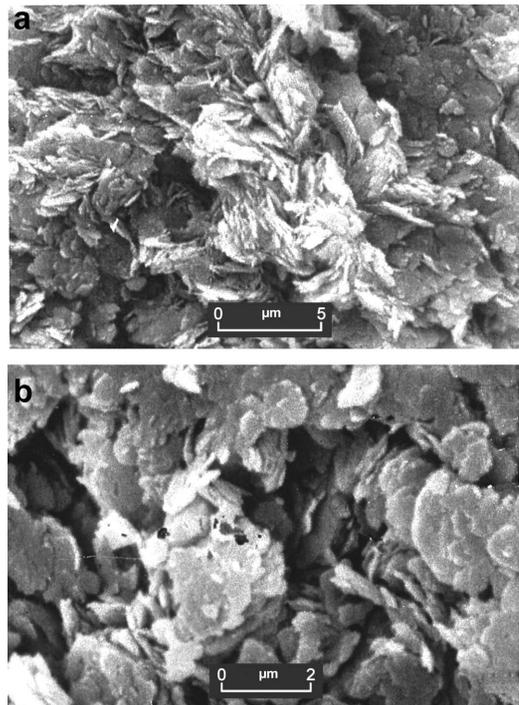


FIG. 4. SEMs of ball clays from (a) Devon and (b) Ukraine showing an open randomly oriented fabric with many voids.

instability. They suggested that the failure mechanism involved was through fluid invasion through micro-pores and micro-fractures, combined with mineralogical transformation of kaolinite to highly dispersible illite. Such a rapid transformation is possible as demonstrated by the synthesis experiments of Bauer *et al.* (1998), but only under such highly alkaline conditions (pH >11) as to bring about the complete dissolution of kaolinite. Such a reaction seems unlikely in a borehole environment.

The very low cation exchange capacity of kaolinite suggests that its basal surfaces are essentially neutral so that it would seem that the failure mechanism cannot be related to compression of the DDL in micropores as suggested above for smectitic and illitic shales, nor indeed to inter-lamellar swelling and hydration. However, kaolinite interacts strongly with water by virtue of its exposed hydroxide basal surface, as well as through edge sites. The edge surface may form a not inconsiderable proportion of the total surface area where kaolinite occurs in small particles, which it often does when the mineral occurs in a poorly crystallized form. In addition, Newman (1987) pointed out that experimental work has shown that kaolinite does have a small permanent negative charge, and that as the total specific surface area of the clay mineral is relatively small (compared with smectite), its charge density is similar to that of moderate- to high-charge smectite. Thus, Newman (1987) calculated a charge density  $0.127 \text{ Cm}^{-2}$  for montmorillonite compared with a charge density for kaolinite in the range 0.15 to  $0.2 \text{ Cm}^{-2}$ . This means that in order to balance this charge hydrated cations are adsorbed to both the siloxane and hydroxide basal surfaces of kaolinite. A variety of studies reviewed by Newman (1987) indicate that the influence of the kaolinite surface on the arrangement of the adsorbed water extends to between two to four molecular layers of water. It seems reasonable therefore to consider that the mechanism for the instability of kaolinitic shales may be similar to that of illitic shales, and relates to the ease of hydration of the cations adsorbed at the basal surfaces of the clay mineral combined with increasing pore pressure as a result of compression of the DDL.

Another possible mechanism for the instability of kaolinite in wellbore conditions may be found in the ability of this clay mineral to intercalate salts between its constituent layers. In particular, this may relate to the ineffectiveness of K-based fluids

at inhibiting instability as found by Carpacho *et al.* (2004). They were able to stabilize these kaolinitic shales by adding an aluminium complex to the drilling fluid which *a priori* would be unlikely to form an intercalation complex with kaolinite. Wada (1961) showed that following a period of grinding it was possible to form an interlayer complex between kaolinite and K-acetate whereby the basal spacing of the clay mineral expanded from  $\sim 7$  to  $14 \text{ \AA}$ . It was later found that other salts of organic acids of low molecular weight with large monovalent cations of low hydration energy could also form complexes with kaolinite. Reaction rates for complex formation generally increased with increasing grain size, temperature and water content (Rausel-Colom & Serratos, 1987). It is not known whether such complexes would form in kaolinitic shales in wellbores but it may be speculated that even the incipient stages of complex formation following interactions with K-based fluids could lead to a delaminating and dispersive effect on the kaolinite clay mineral. Evidently this is a subject that merits further investigation.

## CONCLUSIONS

Current explanations of the relationship between clay mineralogy and shale instability emphasize volume expansion following osmotic swelling of the interlamellar space of Na-smectitic clays as the primary failure mechanism. Evidently, this mechanism cannot account for the instability of illitic or kaolinitic shales and it is not at all certain that it is necessarily applicable to all smectitic shales, bearing in mind the lack of data showing the reality of osmotic swelling under basin conditions, as well as the apparent stability of the one-layer hydrates of Na, K and Ca smectites to a depth of  $\sim 6.7 \text{ km}$  as adduced by MD computer simulations (de Pablo *et al.*, 2005). An alternative scenario for the failure of smectitic and illitic shales envisages a mechanism related to overlap of the DDL associated with the charged external surfaces of the clay minerals exposed in opposing walls of micro- and meso-pores of the shale, thus leading to a build up of pore/hydration pressure. The inhibiting effect on shale instability of the more concentrated K-based, aqueous drilling fluids would then be related to contraction of the thickness of the DDL and not to the collapse of the interlayer space of the smectites. Similarly, the inhibiting effect of

polymers would relate to their encapsulation of the external clay mineral surfaces so inhibiting the hydration of adsorbed cations. It is certainly possible that reactions involving expansion/contraction of the interlayer space of smectites in impermeable shales may occur at the immediate wellbore face. For many shales, however, it is suggested that invasion of fluids through heterogeneous features such as micro-fractures and sedimentary laminations leads to increased pore/hydration pressure in micro- and meso-pores where the charged external faces of the clay minerals are exposed. In this scenario the principal reason for shale instability would be the forced overlap of the DDLs associated with the clay minerals. Kaolinitic shales may be destabilized by the same mechanism but it is also surmised that the ability of the clay mineral to form intercalation complexes of increased volume with low molecular weight salts, where the monovalent cation is large and of low hydration energy, may delaminate and disperse the clay mineral and so have a destabilizing effect on the integrity of the shale.

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