PARTICLE ARRANGEMENTS AND DIFFERENTIAL IMBIBITIONAL SWELLING IN DEFORMED OR DEPOSITED KAOLINITE–ILLITE CLAY*

by

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ABSTRACT

A FINE-GRAINED kaolinite-illite clay, in which planar particle orientation resulted from filtration or plastic deformation, swelled differentially when liquid water was imbibed by test pieces already in the plastic state. Swelling was greater perpendicular than parallel to the plane in which the clay platelets tended to be arranged. Filtration, in contrast to deformation, gave test pieces that swelled more rapidly and extensively. The slipbands, present only in plastically deformed test pieces, restricted the swelling. They were arranged diagonally to the plane in which the clay platelets tended to lie. The plastically deformed test pieces showed enhanced expansion perpendicular to this plane when they were made successively thinner by compression under a cylindrical roller. This was attributed to improved planar orientation of the clay platelets.

INTRODUCTION

BOLT (1956) has concluded that the swelling of clay-mineral aggregates in water is enhanced when the particles are thin and their specific surfaces large, and that interlamellar swelling of the individual crystals is not necessary for its occurrence. Thus Mielenz and King (1955), de Bruyn, Collins and Williams (1957), Aylmore and Quirk (1962) and others have reported the imbibitional swelling of illite or of kaolinite–illite clays. In particular, Williamson (1955a) showed that planar particle-orientation in kaolinite–illite clays induced a differential imbibitional swelling, in which the expansion was greater perpendicular to the plane of preferred orientation. He had some evidence that the swelling was less when the orientation had been produced by plastic deformation rather than by filtration. The diminished swelling, if substantiated, may result from shear-hardening, to which Macey (1940; 1942) ascribed the retarded expansion of clay brought to equilibrium moisture content between porous pistons, and subsequently decompressed. Shear-hardening may result from a better-developed face-to-face arrangement of the clay platelets (Williamson, 1947), and energy considerations accord with this idea (Williamson, 1955b;

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1960) Later work, however, suggests that more complex particle arrangements may be involved in the development of shear-hardening. Thus Weymouth and Williamson (1953) showed that slipbands, in which the clay platelets have a specific orientation, develop during plastic deformation and cut diagonally across the planar arrangements of platelets caused by laminar flow.

The progressive thinning of plastic kaolinite–illite slabs by pressure from a cylindrical roller tended to enhance differential drying-shrinkage (Williamson, 1962). The shrinkage was greater perpendicular, than parallel, to the surface over which the roller had passed, because the clay platelets tended to be parallel to this surface. The enhanced differential drying-shrinkage resulted from an intensification of the planar orientation, caused by progressive deformation. The present study was designed to determine if differential imbibitional swelling was similarly enhanced or if, on the contrary, progressive deformation limited such swelling by causing greater shear-hardening. The study was designed also to investigate the suggestion by Williamson (1955a) that plastically made test pieces swelled less than those prepared by filtration.

MATERIALS AND METHODS

The Clay Investigated

The water contents quoted for this clay were calculated on the dry weight, and the percentage imbibitional linear expansions refer to the original length of the plastic clay.

The material was a "ball clay" from Dorset, England, having the approximate composition kaolinite 70, illite 10–15, and quartz 15–20 per cent by weight (Kilgore, 1963). A chemical analysis, the exchangeable cations present, and the particle-size distribution appear in Tables 1, 2 and 3 respectively.

TABLE 1.—ANALYSIS OF BALL CLAY FROM DORSET, ENGLAND

SiO_2	53.50
Al_2O_3	31.11
Fe_2O_3	1.10
TiO_2	1.68
MgO	0.24
CaO	0.24
K_2O	2.59
Na_2O	0.49
Ignition loss	8.95
	99.90

Plots of the cubical drying-shrinkage against the corresponding water content, for test pieces made plastically by various techniques, indicated "first critical points" at 16.1–18.6 per cent of water, the percentages varying

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TABLE 2.—EXCHANGEABLE CATIONS IN BALL CLAY

Cation exchange capacity	15.36 meq/100 g
Ca	4.65
Mg	3.17
Al	3 84
Na	0.16
K	0.52
(By difference) H	3.02

TABLE 3.—PARTICLE-SIZE ANALYSIS OF BALL CLAY (CENTRIFUGE METHOD)

Equivalent spherical diameter	% Weight (Finer)
2.0 μ	90
1.5	86
1.0	80
0.5	68
0.2	49
0.1	21

with the techniques (Kilgore, 1963). These percentages corresponded to where the plot, linear for larger water contents, showed an inflection caused by a relatively sudden diminution in drying-shrinkage, or by a change to a slight drying-expansion. The inflection was presumed to indicate the water content at which the clay particles, separated by water films in the earlier stages of drying, first achieved virtual contact.

Preparation of Test Pieces

The finely ground clay was tempered with 30 per cent of water, and kept for 1 month in a closed " plastic " bag stored over water in a sealed vessel.

A lump of the clay was thrown onto a flat surface in such a way that a 2-in. cubical block was formed. The block was then flattened to thicknesses of $\frac{3}{4}$, $\frac{1}{2}$ and $\frac{1}{4}$ in. by passing a wooden roller repeatedly across one face. It rested on a jig board, which prevented the roller from being depressed beyond the

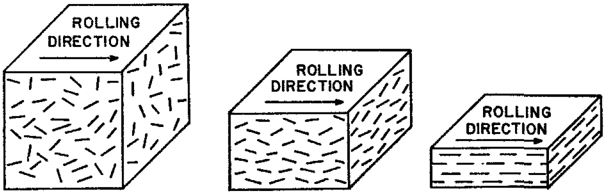

FIG. 1. Schematic diagram showing the progressive rearrangement of clay plates into parallelism with the surface under the roller. If the left-hand block represents an unrolled test piece, those in the middle and on the right are imagined to be cut from similar blocks progressively flattened and spread by the roller.

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limits corresponding to the predetermined thickness. The technique resembled the flattening of pastry under a rolling pin (Figs. 1 and 2). Waxed paper beneath the clay minimized the loss of water to the underlying jig board.

Rectangular prisms 2-in. long, ¼-in. broad, and ¾-, ½- or ¼-in. thick were cut from slabs of corresponding thicknesses.

Less than 0.1 per cent of water was lost during rolling and cutting the clay.

Test pieces were made also by filtration. An aqueous suspension containing about 35 per cent by weight of clay, with no added dispersant, was poured into a filter paper supported in a 11.1-cm diameter Buchner funnel connected with a water-aspiration pump. The resulting disks contained about 40 per cent of water when removed from the funnel. Prisms were cut from them, similar in dimensions to those dissected from the plastically formed slabs. These were dried slowly at room temperature until about 30 per cent of water remained.

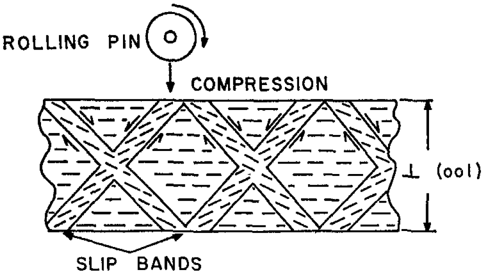

Fig. 2. More detailed schematic diagram showing the disposition of the clay platelets in the right-hand block of Fig. 1. Compression under a conventional " rolling pin " rearranged the clay-platelets parallel to the rolled surface, but also produced slipbands diagonal to this surface. In these, the clay platelets have a specific orientation, differing from that in matrix, and associated with differential movement of the opposing walls of the band (cf. Weymouth and Williamson, 1955, for photomicrographs and diagrams of comparable slipbands).

The various prismatic test pieces were wrapped in waxed paper and stored in saturated air at room temperature before use.

Promotion of Swelling

The test pieces rested on horizontal filter papers supported by glass microslides lying on the bottom of a Petri dish 100 mm in diameter and 20-mm deep. The dish was covered with a glass plate and contained a pool of water that kept the filter papers saturated but did not come in contact with the test pieces.

The experiments were made at 24.5 ± 1.0°C.

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Measurement of Swelling

The prismatic test piece was arranged so that the plane in which the clay platelets were preferentially oriented, termed (001) for brevity (Weymouth and Williamson, 1953), was parallel to the axis of a measuring microscope (Fig. 3). The microscope focused through the glass cover of the Petri dish and measured to 0.01 mm. Minute, appropriately disposed fiducial marks on the faces of the test pieces permitted linear measurements parallel or perpendicular to (001). Two or three measurements were made as quickly as possible along independent lines of traverse in each direction, and the results were averaged. As the test piece continued to swell during the measurements, the averaging of measurements along more numerous lines of traverse would have diminished rather than increased the precision.

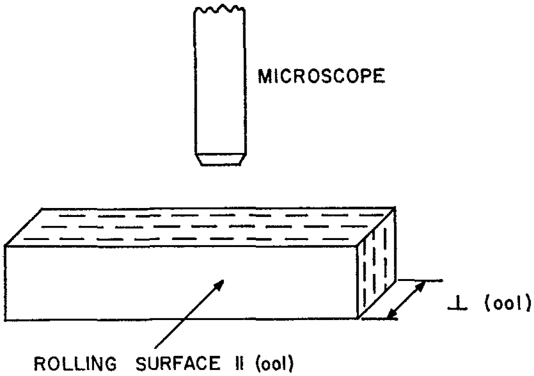

FIG. 3. Arrangement of test piece under the measuring microscope so that the plane of preferred particle orientation is parallel to the axis of the microscope. Vertical edges measure ¼ in., and the shorter horizontal edges ¾, ½, or ¼ in., according to the predetermined thickness produced by rolling or filtration.

Water Content of Clay

The percentage water contents of the original clay, the swollen test pieces, etc. were found by measuring the loss in weight of specimens maintained for about 48 hr at 105°–110°C.

Investigation of Microstructures

The microstructures of relevant dried test pieces were examined. Drying was unlikely to effect significant rearrangements of the clay platelets (Williamson, 1955b), although Aylmore and Quirk (1960) reported distortions of illite crystals and aggregates that may have been caused by desiccation. Peels (Williamson, 1956) or thin sections were made corresponding to selected

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planes in the test pieces. The thin sections were prepared in a commercial laboratory specializing in such work. Here the test pieces were filled with a transparent hardening material and then cut and ground in oil to give sections of standard thickness. The polarizing-microscope methods used to study the textures of the test pieces and to assess the intensity of the clay–platelet orientation have been discussed elsewhere (Williamson, 1956).

EXPERIMENTAL RESULTS

Expansion of Plastically Deformed Test Pieces

Test pieces made successively thinner under the roller tended to expand more rapidly perpendicular to (001) and to show a greater final expansion in

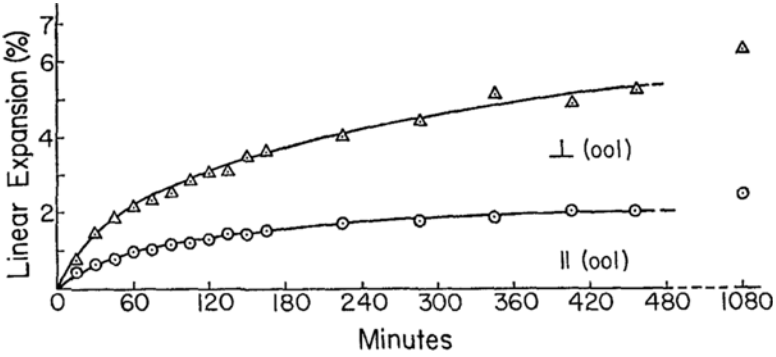

FIG. 4. Differential swelling of test-piece made by rolling, initially $\frac{3}{4}$ in. thick.

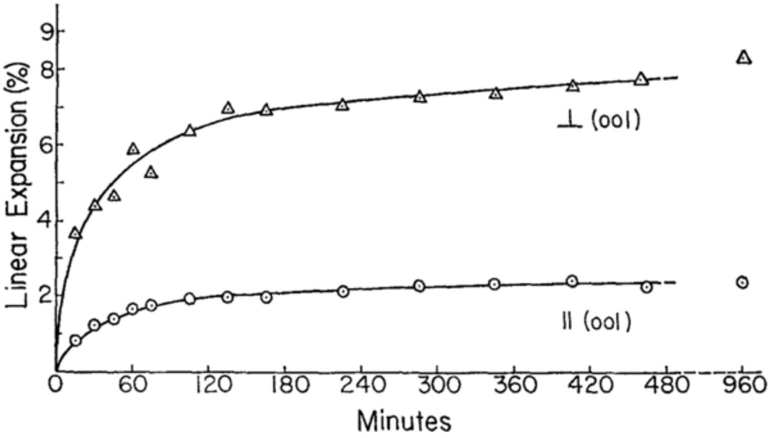

FIG. 5. As Fig. 4, but $\frac{1}{2}$-in. thick. Final water content, 43.2 per cent.

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this direction. A smaller expansion occurred parallel to (001) but the rapidity and magnitude of this were not obviously influenced by the degree to which the clay had been flattened under the roller (Figs. 4–6).

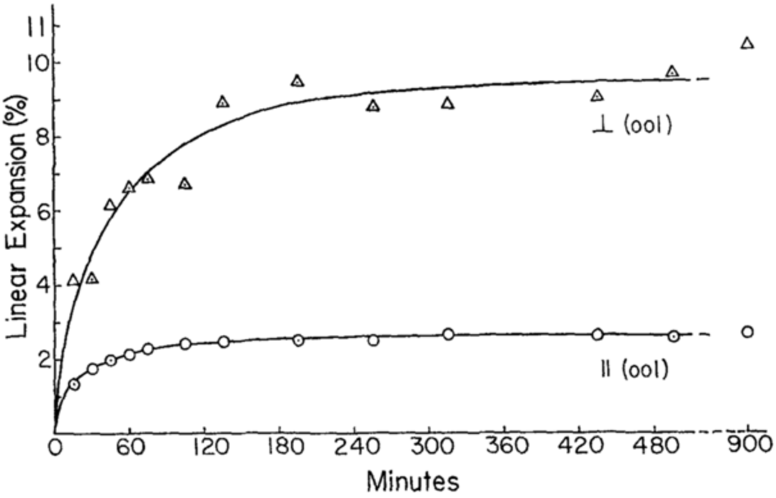

FIG. 6. As Fig. 4, but $\frac{1}{4}$-in. thick. Final water content, 42.9 per cent.

There was also a small differential expansion in the plane of (001) itself, i.e. in the surface to which the roller was applied. The expansion was greater parallel than perpendicular to the direction in which the roller had passed. Thus a test piece, $\frac{3}{4}$-in. thick, showed a difference of about 0.75 between the relevant percentage linear expansions. Thinner test pieces showed smaller differences.

Expansion of Test Pieces Made by Filtration

These test pieces also tended to expand more rapidly and to attain a greater final expansion perpendicular to (001) when they were initially thin rather than thick. Here the thickness of the test piece was determined by that of the parent filter cake. The rate and magnitude of the expansion parallel to (001) did not vary obviously with the thickness of the test piece, but both were markedly greater than those of corresponding plastically deformed test pieces (Figs. 7 and 8).

Figs. 4–8 are regarded as typical and have been selected from more numerous graphs.

Microstructures of Plastically Deformed Test Pieces

There were numerous slipbands, sometimes narrow and sharply bounded but more often broad and diffusely outlined, which were arranged diagonally

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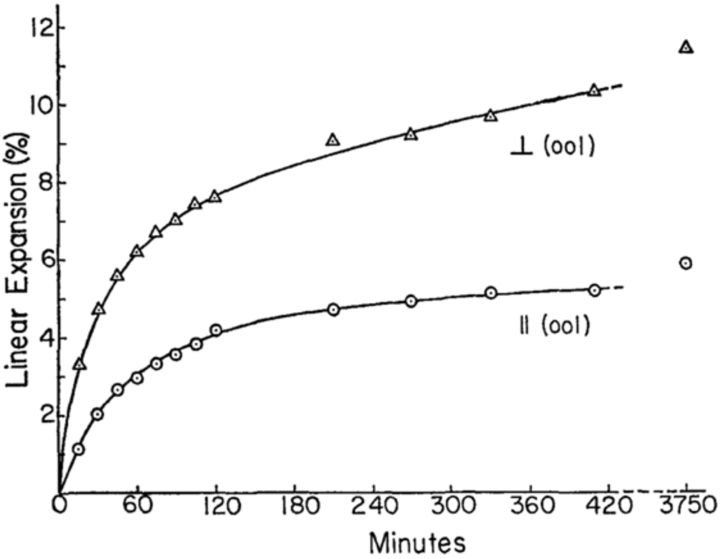

FIG. 7. Differential swelling of test piece made by filtration, initially $\frac{3}{4}$-in. thick.

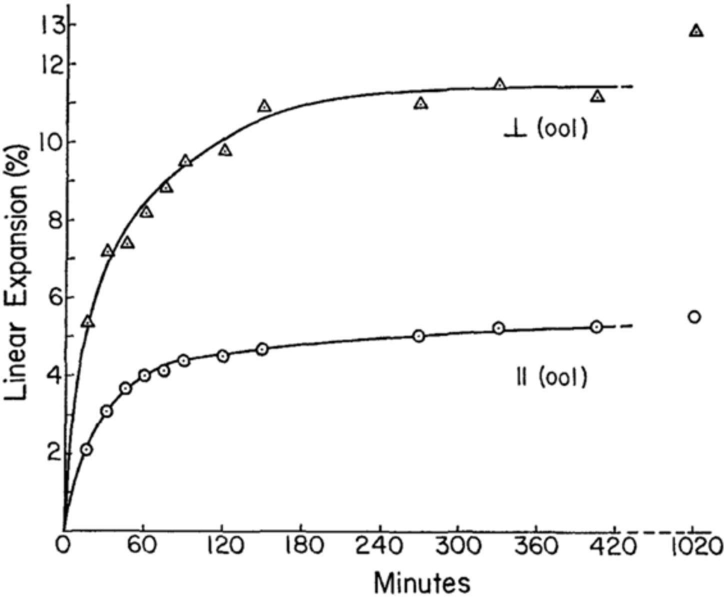

FIG. 8. As Fig 7, but initially $\frac{1}{4}$-in. thick. Final water content, 51.4 per cent.

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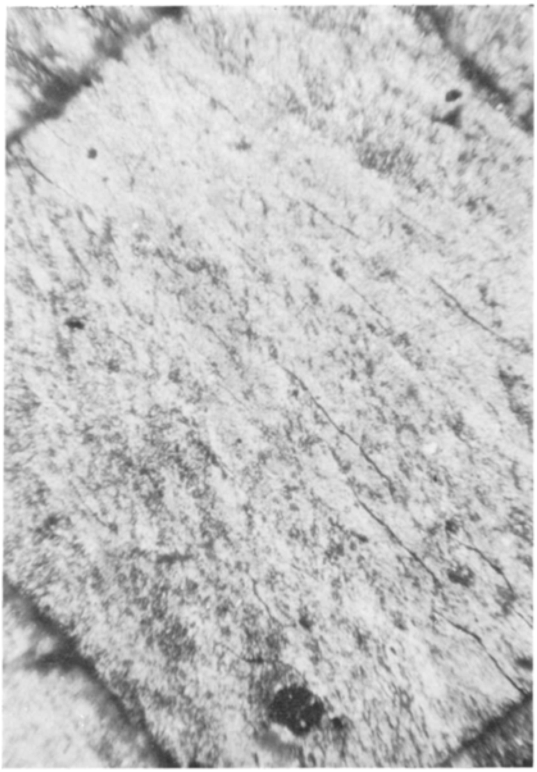

Plate 1. Rolled test piece, section perpendicular to the plane of preferred particle orientation. Crossed polars, 45° position, maximum illumination (×35), (001) N.W.–S.E.

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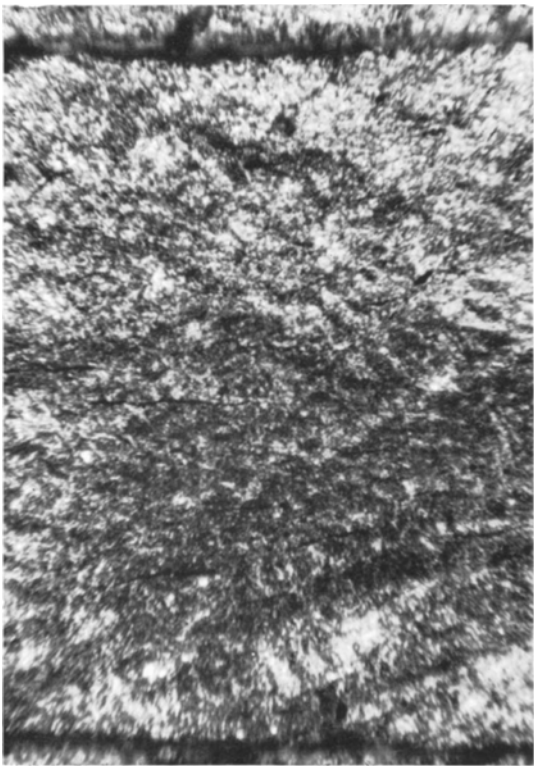

Plate 2. As in Plate 1, but position of maximum extinction. The difference in illumination between Plates 1 and 2 is consequent upon particle orientation, but the extinction in Plate 2 is patchy or incomplete, mostly because diffusely bounded slipbands contain clay platelets oriented differently from those in their matrix (×35), (001) E.–W.

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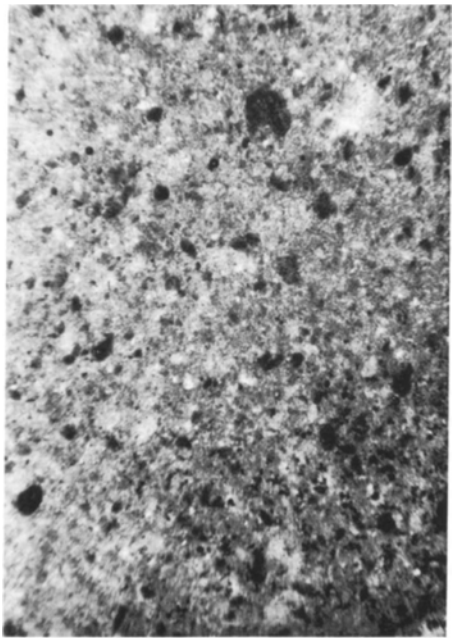

Plate 3 Test piece made by filtration, showing flocs. Section perpendicular to plane of preferred particle orientation. Lower polar inserted (×35).

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to the plane of preferred orientation of the clay platelets (Plates 1 and 2). The clay platelets in the slipbands were at various angles to this plane, which was parallel to the surface over which the roller had passed. More detailed study showed that the slipbands separated domains in which the clay platelets tended to a common parallelism in planes that might be arranged at angles of a few degrees to the corresponding planes in neighboring domains. The planes remained, however, approximately parallel to the rolled surface.

The microstructure resembled that seen in clay cylinders made progressively thinner by rolling between parallel plates (Williamson, 1954). In these cylinders or in the present test pieces, slipbands developed at angles to the direction of compression (Fig. 2). The test pieces were becoming thinner in this direction by plastic flow and, in consequence, the angle of the slipbands to the direction of compression changed, slip became inoperative, and the bands were obliterated by the flow of their matrix. Simultaneously, however, new generations of slipbands were formed, only to be obliterated later. Thus the slipbands still visible in the microsections were of late formation, and the ill-defined examples were those already partially obliterated.

In general, the microstructures resembled those postulated for rolled polycrystalline metals (Gay, Hirsch and Kelly, 1954, Fig. 3).

Test Pieces Made by Filtration.—Variously sized flocs of undispersed clay were abundant and broke the continuity of the planar particle orientation, which tended to be parallel to the original filtering surface. There were no slipbands (Plate 3).

DISCUSSION

The interpretation of the results depends on the particular structure ascribed to the plastic clay. For instance, the clay platelets may be regarded as being in mutual contact, thus forming a meshwork with water in its interstices. Alternatively, they may be assumed to be essentially independent and separated by water films. The latter hypothesis is adopted, because it is in better accord with the behavior of plastic clay that has been subjected to fabricating operations (Williamson, 1955b).

The test pieces contained about 30 per cent of water before imbibition commenced, and thus the swelling was not caused by the initial hydration of the clay-mineral surfaces, but was of the type associated with double-layer repulsion, which can be interpreted also as osmotic swelling (Bolt, 1956; van Olphen, 1962 and 1963, pp. 149–52).

The imbibed water increased the thicknesses of the intercrystalline films already present, and the swelling was greater perpendicular than parallel to (001) because, in the former direction, there were more films per unit length. This simple model resembles that used to explain differential drying-shrinkage, although here progressive thinning, not thickening, of the water films occurs (Williamson, 1947). In fact, the present test pieces, after max-

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mum swelling, showed a marked differential drying-shrinkage, greater perpendicular than parallel to (001). Thus the imbibition or subsequent loss of water had produced no substantial change in the arrangements of the clay platelets. This was supported by the appearance of a " memory " effect during drying, which caused some test pieces to develop a slight curvature concave to the surface originally compressed by the roller. Williamson (1955a) has already shown that " memory " effects, resulting from arrangements of particles produced during a prior deformation, still appeared after test pieces were allowed to imbibe water, or to dry partially, under a light load sufficient to prevent recovery and from which they were later released.

If progressive thinning under the roller had caused increased shear-hardening, swelling might have become more limited. However, the initial rate of expansion and the final extent of the swelling perpendicular to (001) were greater for thinner test pieces, indicating that the degree of parallelism of the clay platelets had a major influence on the swelling behavior. A greater degree of parallelism would be expected in the thinner test pieces.

Shear-hardening may limit the swelling of test pieces that are too dry to be effectively fabricated by the present technique, especially where the water contents were less than those appropriate to the " first critical point ". This possibility was not investigated.

Test pieces cut from filter cakes tended to show a greater expansion perpendicular to (001) when the original filter cake had been relatively thinner (Figs. 7, 8). This was because the poorer particle orientation, which occurred away from the filtering surface, contributed least to the expansion behavior of such test pieces.

Comparison of Figs. 4–6 with Figs. 7 and 8 shows that there was initially a more rapid, and ultimately a greater, expansion parallel to (001) when the test pieces had been made by filtration rather than by plastic deformation. The phenomenon could be due to the poorer particle orientation associated with the presence of distinct flocs in the test pieces made by filtration (Plate 3). However, this is unlikely to be the entire explanation, since the final expansion perpendicular to (001) was greater for test pieces made by filtration than for the corresponding test pieces made by plastic deformation, which could imply a better, not a worse, particle orientation in the former. If it is argued that the ratio [linear expansion perpendicular to (001)] : [linear expansion parallel to (001)] is a more valid basis for comparison, the appropriate values calculated on the final expansions shown in Figs. 4 and 8, for instance, are 2.5 and 2.3 respectively. These ratios are not very different, yet the test piece made by filtration (Fig. 8) showed markedly greater expansions either perpendicular or parallel to (001).

It may be suggested that the slipbands, found only in test pieces made by plastic deformation, tended to restrict swelling perpendicular or parallel to (001), although the particle orientation, which was often better in these test pieces, might have been expected to enhance the swelling, especially that perpendicular to (001). The apparent marked restriction of swelling parallel

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to (001), obvious when Figs. 4–6 are compared with Figs. 7 and 8, may be partly a consequence of the experimental methods. Lines of traverse in this direction were up to seven times longer than those perpendicular to (001). Thus the total restrictive effect of a greater number of slipbands contributed to the calculated percentage expansion and may be more than additive.

The clay platelets in the slipbands made relatively large angles with the planes of particle orientation in their matrix. Thus an edge-to-face association of the platelets at the boundaries of the bands was feasible, having a restrictive effect similar to the crosslinking that, according to van Olphen (1962), can limit the interlamellar swelling of montmorillonite.

The restrictive effect of the slipbands is, however, thought to be caused mostly by their own differential imbibitional swelling, which may oppose that of the matrix, because the plane of parallelism of the clay plates and the associated direction of maximum swelling are different in the two regions.

The slightly greater swelling in the plane of (001), parallel, as contrasted to perpendicular, to the direction of rolling, may indicate that the slipbands do, in fact, swell differentially. It is explicable if the bands are visualized as packets of clay flakes arranged at angles to this plane (cf. Fig. 2), with the line of maximum slope on the surface of each flake tending to lie in a plane that includes also the direction in which the roller passed.

CONCLUSIONS

Plastic fine-grained kaolinite–illite test pieces, in which the clay platelets tended to be arranged in a common plane, imbibed liquid water spontaneously at 24.5 ± 1.0°C. Swelling was greater perpendicular than parallel to this plane. Plastically made test pieces swelled more, perpendicularly to this plane, when progressively flattened under a cylindrical roller, because flattening improved the common orientation of the platelets. The roller produced slipbands diagonal to the plane of orientation, containing clay platelets arranged at angles to the latter. These slipbands induced a swelling parallel to the rolled surface, which was slightly greater along the direction of movement of the roller.

Swelling of the slipbands opposed that of their matrix, because both swellings were differential, with maxima in different directions. Thus plastically formed test pieces, despite the better planar particle orientation in this matrix, swelled less rapidly and extensively than those made by filtration, because the latter lacked slipbands.

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