PARTICLE ARRANGEMENTS IN KAOLIN

by

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ABSTRACT

AN AMBIGUITY in interpretation of electron micrographs of replicas of fracture surfaces of dry clay samples is illustrated, and a method of making ultra-thin sections described. Micrographs of ultra-thin sections of remoulded, consolidated, and sheared kaolin specimens are shown. In unconsolidated specimens random arrangements of particles, or of small groups of particles, were found. In failure zones, in sheared samples, the preferred orientation was parallel to the zone, with, in some cases, a subsidiary zone in which particles were inclined to the main zone. Interparticle contacts were mainly edge-to-edge or face-to-face.

INTRODUCTION

REMOULDED kaolin specimens, treated differently, were examined by electron-microscopy using ultra-thin sections and replicas (see Bibliography). Attention was paid to the arrangement of the particles, that is to the geometrical relationship between two particles, which either touch or approach closely, to the structures built by small numbers of particles, and to preferred orientation of tabular particles.

Adjacent particles may be expected to associate in the following arrangements: face-to-face (Lambe, 1953), edge-to-edge (Emerson, 1962), oblique edge-to-edge, perpendicular edge-to-face, oblique edge-to-edge (Trollope and Chan, 1960; Ashbee, 1961; Mungan and Jessen, 1962), corner-to-face, etc.

The structures to be expected are: cardhouse, turbostratic, and salt-flocculated (Hofmann, 1942; Aylemore and Quirk, 1960; Schofield and Samson, 1953; respectively).

Various hypotheses have been advanced to describe the structural changes resulting from mechanical disturbances, for example the development of preferred orientation during consolidation (Sorby, 1908, pp. 189–90), and orientation of tabular particles parallel to failure zones during shearing (Graecen, 1959).

Sloane and Kell (1965) give a brief review of work in this field.

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SPECIMENS

Spestone Kaolin, an English china clay, was received from the quarry as a fine dry powder. After being mixed in a Z-mill with an equal weight of distilled water for 1 hr, it was divided into three specimens. One specimen was consolidated in a shear box to 4 kg/cm^2, allowed to rebound to 2 kg/cm^2, sheared at 1 mm/min and finally unloaded completely. For the second specimen shearing was omitted. The third specimen was left in the remoulded unconsolidated state. Undisturbed samples were cut from the sheared and consolidated specimens with a cheese-wire; the remoulded specimen was subdivided with a spatula.

Another portion of Spestone Kaolin, mixed at 136% mc, was consolidated in a triaxial shear test machine to 80 psi (5.6 kg/cm^2) and strained, undrained, at 0.1 mm/min until failure and thereafter, by manual drive, more rapidly. This specimen was then oven-dried.

REPLICAS

Samples of the specimens, other than the triaxial test specimen, were dried and fractured in the vertical direction. The fracture surface was shadowed with platinum–carbon at 45°, and two further coats of carbon were added. The sample was pushed down an incline into hydrofluoric acid so slowly that the replica floated clear. After 4 hr the replicas were washed on 10% alcohol in distilled water, freshly mixed, and mounted on AEI-type specimen support grids.

The triaxial test specimen was broken open along a well-developed failure plane; and replicas were made of this plane in exactly the same way.

These replicas were triply selected: some of the samples collapsed during fracturing; the fracture probably passed through the weaker parts of the sample, rather than through average parts; and replicas of the rougher parts of the fracture surface probably disintegrated.

Different interpretations of the same surface are illustrated in Fig. 1. In these sketches, each rectangle represents the cross-section of a tabular particle; and these particles are arranged in two-dimensional analogues of three-dimensional situations. The dotted lines represent the cross-section of

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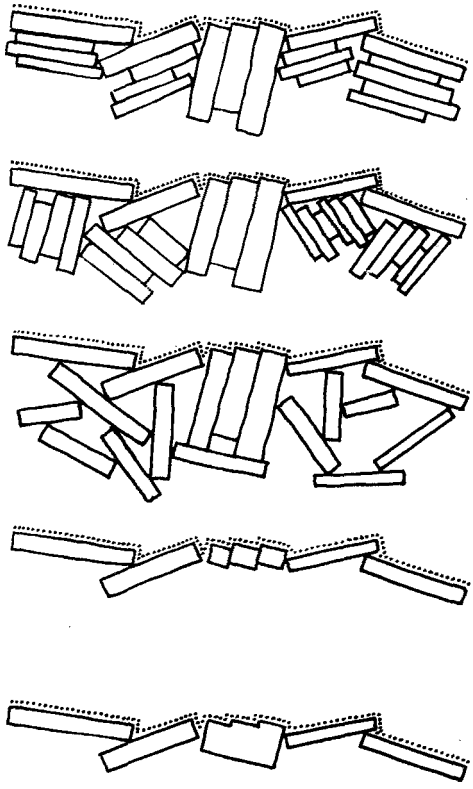


FIG. 1

a replica; the same replica is shown in each sketch. In the two upper sketches, the tabular particles are arranged in packets. In the uppermost sketch, the fracture surface has passed between packets; in the second sketch, through them. Even were it known that the particles were arranged in packets, the real distribution of the orientation of the packets would be difficult to ascertain. The central sketch shows the same surface hiding cardhouse structure. The same surface could also hide salt-flocculated structure (not illustrated). In the two lower sketches, the central feature of the replica, which had been interpreted as a packet in all the other sketches, is shown as: three laths side by side, and a single tabular particle with growth steps on

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its surface. Whilst, in practice, it may be possible to obtain clues, which help to resolve these difficulties, it is obvious from these sketches that great care must be used when interpreting micrographs.

The holes in the support grids were nominally circular and 40 μ diameter. Several holes in each grid were examined. These holes were usually chosen to form two rows, one perpendicular to the other, crossing the replica in arbitrary positions. There were holes in the replica; and, for each grid-hole, the proportion of the area filled with replica was judged by eye. Holes having more than 75% of their area were noted as "filled". The number of packets seen in each filled hole was counted. The results are shown in Tables 1 and 2. If the packets were distributed randomly, the frequency distributions would follow Poisson distributions, but too many holes showed few packets. The author assumes that the packets were distributed randomly in part of the area and that, in other parts, the fracture has passed through a domain and shows only face-on particles. As might be expected, least packets were seen in the failure plane. The difference between the unconsolidated and the consolidated samples is not significant. The sheared sample showed twice as many packets as the consolidated. The author assumes that shearing has resulted in the elimination of some of the domains in which the particles were still orientated vertically after consolidation.

TABLE 1.—PACKET COUNTS

n	f				n	f			
	U	C	S	P		U	C	S	P
0	7	4	0	30	17	1	0	4	
1	10	7	1	8	18	2	2	2	
2	5	17	2	7	19	0	1	2	
3	6	8	3	8	20	2	1	0	
4	5	9	5	2	21	1		0	
5	5	13	3	3	22	1		1	
6	3	3	1		23			0	
7	4	6	1		24			1	
8	3	4	2		25			1	
9	2	7	4		28			1	
10	3	6	1		29			1	
11	2	2	5		37			1	
12	2	5	2		39			1	
13	3	2	3						
14	2	3	1						
15	0	0	1						
16	0	0	2						

n = the number of packets per hole.
f = the frequency with which n was observed.
U = unconsolidated specimen.
C = consolidated specimen.
S = sheared specimen.
P = failure plane.

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TABLE 2.—PACKET DENSITY

Specimen	Average number of packets per support grid hole
Unconsolidated	6.52
Consolidated	6.06
Sheared	12.48
Failure plane	1.19

ULTRA-THIN SECTIONS

In order to make ultra-thin sections, samples were impregnated with Vestopal or Araldite, after first replacing the pore-water by acetone or alcohol. In the basic procedure, these samples were not dried, and care was taken to avoid osmotic shocks, surface tension shocks and mechanical shocks. However, the triaxial test specimen was dried, and, after it had been broken open along the failure plane, a sample containing part of this plane was cut out and immersed in acetone.

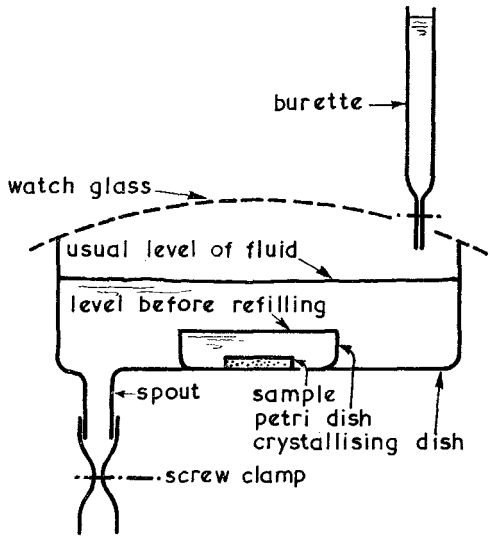


FIG. 2. Apparatus for impregnation.

Samples measuring $10 \times 5 \times 2$ mm were cut from the other specimens and placed in apparati similar to that shown in Fig. 2. This figure is not to scale; the dimensions must be chosen to suit the samples. The sample is in a petri dish. The petri dish is placed in a crystallizing dish, which is fitted with a

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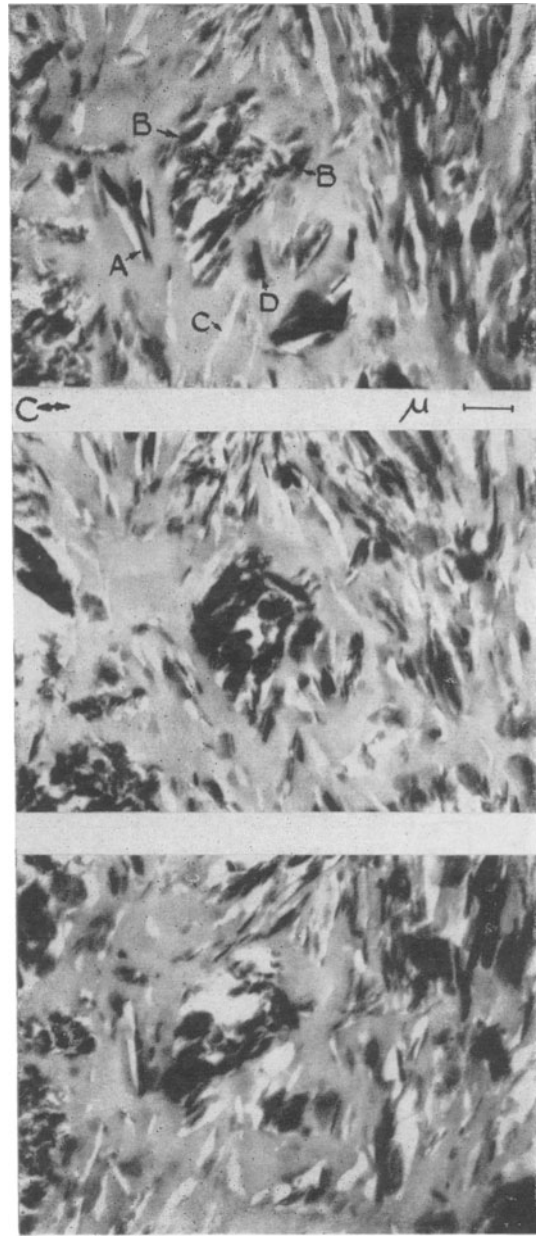


PLATE 1. Electron micrographs of corresponding areas of three ultra-thin sections. Beside C and μ are the direction of cut and the length of one micron, respectively.

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drain, consisting of a glass spout, rubber tube, and screw clamp. The flexibility of the rubber tube insulates the samples from any clumsiness when operating the clamp. The crystallizing dish is filled gently from a burette held steadily by a clamp; care must be taken to avoid adding fluid directly



PLATE 2. Remoulded unconsolidated kaolin.

to the petri dish, and care is needed at the moment at which the petri dish is overtopped. The crystallizing dish is filled first with 25% alcohol in distilled water. After time for diffusion, 1 day, the crystallizing dish is emptied by unscrewing the clamp, and refilled; the petri dish is not emptied. At successive changes the strengths of the alcohol solutions are 50%, 75%, 100%, 100%, and 100%. For some samples, the process is continued with mixtures of acetone in alcohol. Except whilst refilling, the crystallizing dish must be

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covered with a well-fitting watch glass; otherwise the solutions evaporate with disastrous rapidity.

After the final change of acetone, the crystallizing dish was emptied, and most of the acetone in the petri dish was removed, leaving the sample submerged. A mixture of Vestopal and acetone was gently added directly

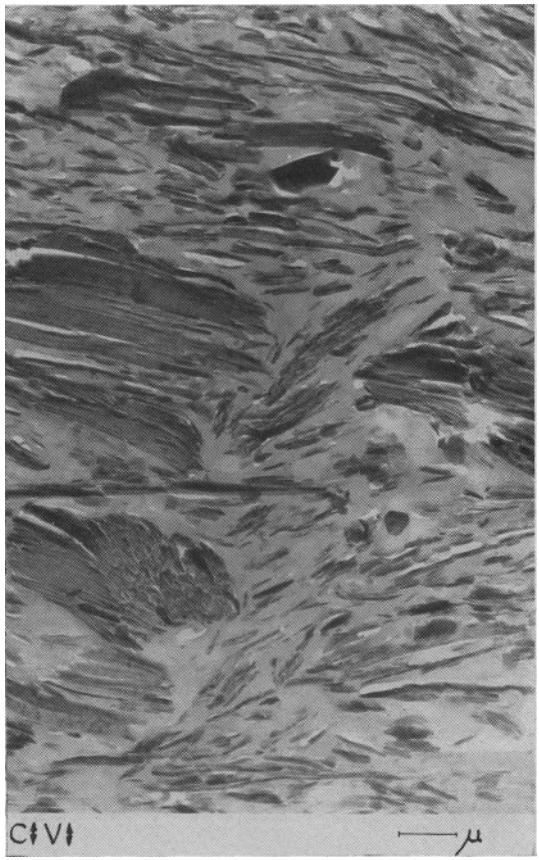


PLATE 3. Consolidated kaolin. Beside V is the vertical direction.

to the petri dish. A typical recipe is: Vestopal W, 100; initiator, 1; activator, 1; acetone, 300 parts by weight. (Vestopal is obtained from Martin Jaeger, Vesanez, Geneva, Switzerland.) The acetone was allowed to evaporate. The Vestopal impregnated the sample and hardened, then the Vestopal was cured at 60°C for 24 hr. (The author allows two months from adding Vestopal to starting microtomy.)

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The other samples were lifted on the brass foil out of the alcohol and submerged in Araldite; a typical recipe is: Araldite AY18, 100; Hardener HZ18, 75; dibutyl phthalate, 0 to 10 parts by weight. (Araldite is obtained from CIBA (A.R.L.) Ltd., Duxford, Cambridge, England.) After 2 weeks impregnation was complete, and the Araldite had solidified.



PLATE 4. Consolidated kaolin, a vertical section cut vertically.

Other resins were tried with limited success.

After curing, all the samples were subdivided, mounted in Araldite in gelatine capsules and sectioned as described in books on electron microscopy (Kay, 1961). Huxley, Porter–Blum MT2, LKB Ultratome I, and LKB Ultratome III microtomes were used with diamond knives. Typical data are: knife angle 45°, clearance angle 4°, cutting speed 2 mm/sec, nominal thickness of section 700 Å.

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Electron micrographs of ultra-thin sections are large-scale shadow graphs. The particles appear dark grey, the plastic matrix light grey, and holes in the section white.

About thirty types of artifacts were found; these will be discussed elsewhere. Apart from distortion of the section during cutting and mounting,

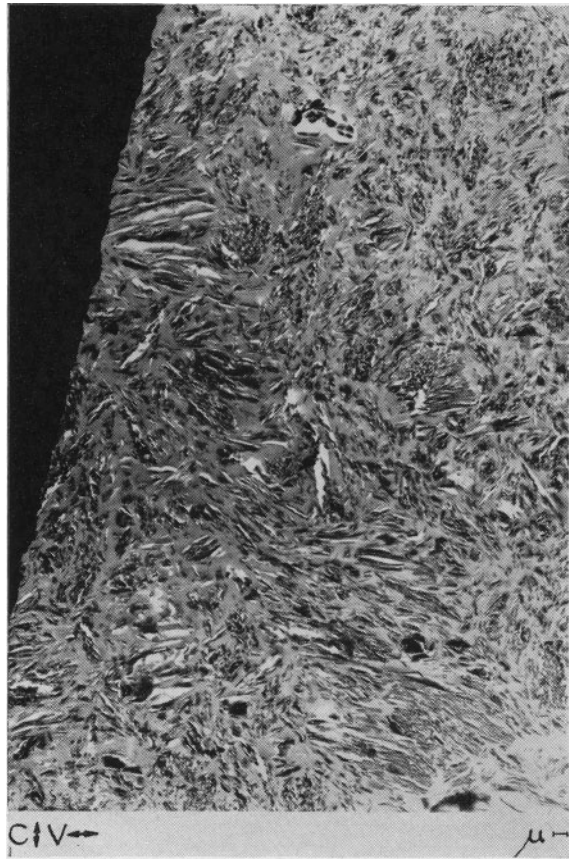


PLATE 5. Consolidated kaolin, a vertical section cut horizontally.

the most important effect was that many particles appeared as though they were not in contact with other particles. In some cases it was thought that the interparticle contacts had been broken during embedding, and that the particles had moved slightly. Another cause of this effect is illustrated in Plate 1.

The three micrographs in Plate 1 show corresponding areas of three sections taken in order, not necessarily consecutively, from a string of sections from

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a sample of sheared kaolin in Vestopal. At A, see upper micrograph, is seen a group of particles (and holes) which can be traced through the series of micrographs. In the central micrograph they appear as though they are not in contact with other particles, but contact appears to be established in the lower micrograph. Thus, a deficiency of contacts in one ultra-thin section does not necessarily indicate a deficiency of contacts in the sample. At BB is a larger group, in which holes in one micrograph frequently correspond with particles in another. At C is a hole corresponding with a particle in the central micrograph. At D is a particle with a dark patch to the left. At the corresponding position in the central micrograph is a hole, whose shape suggests that the dark patch may result from uneven cutting of the plastic matrix.

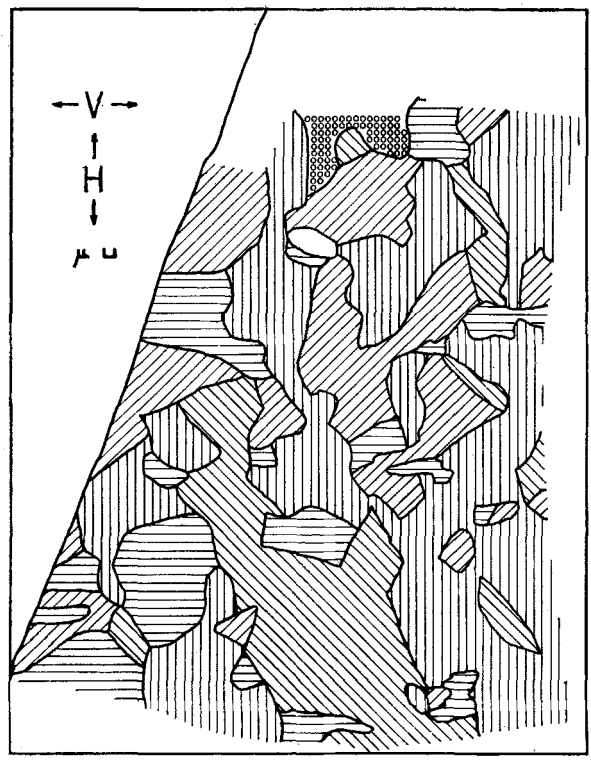


FIG. 3. Domain trace diagram corresponding to Plate 5.

Plate 2 is an ultra-thin section of unconsolidated remoulded kaolin impregnated with Vestopal. There are many small holes beside particles in the section; these holes are cutting artifacts. In the top right-hand corner, at S, are the remains of two particles, which were shattered during microtomy,

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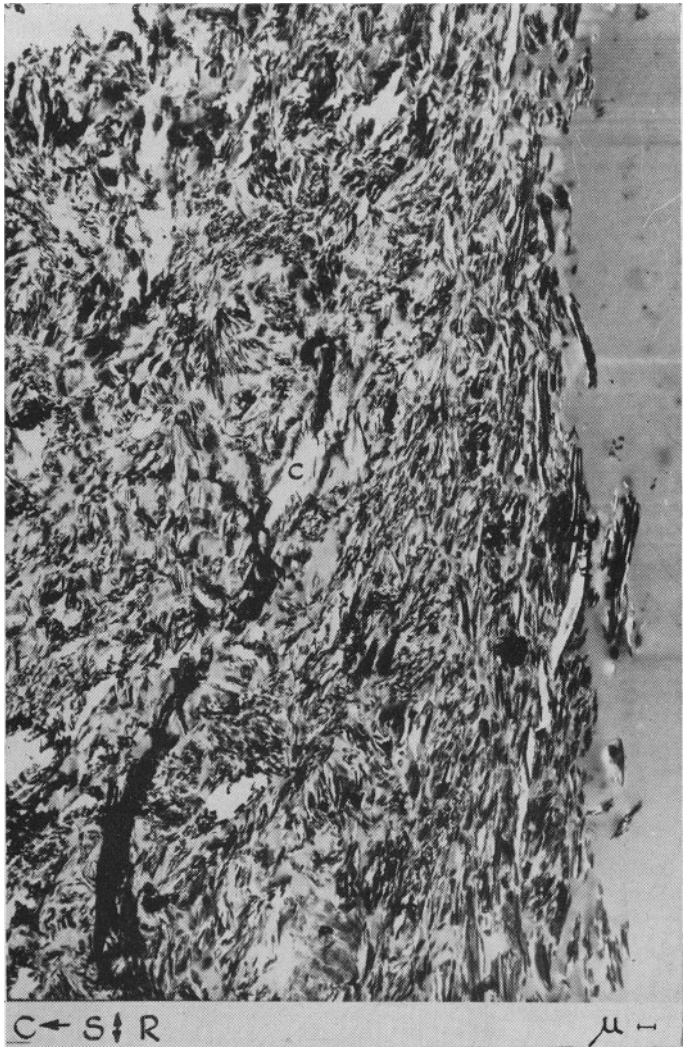


PLATE 6. Ultra-thin section of a failure zone. Beside S is the direction of shear.

another common artifact. Most of the particles are arranged face-to-face in very small domains; the domains are arranged randomly. The voids are commensurate with the particles. Although there is no preferred orientation, it is difficult to describe this as cardhouse structure. In other parts of this specimen, turbostratic structure was found. These observations illustrate the difficulty of preparing remoulded specimens with truly random structure.

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Plate 3 is consolidated kaolin in Vestopal. The interparticle contacts, or approaches, are mostly edge-to-edge or face-to-face.

Plate 4 is a vertical section of consolidated kaolin in Vestopal cut vertically. There are two knife marks, indicated by arrows, running from side to side across the micrograph. The particles are arranged in well-developed domains with pronounced preferred horizontal orientation.

Plate 5 is a vertical section of consolidated kaolin in Vestopal cut horizontally. Many of the particles with preferred horizontal orientation were shattered during microtomy; and the visual effect is misleading.

The domains on the original of Plate 5 were mapped and classified:

H particles approximately horizontal;
I particles approximately at 45° to the horizontal;
U particles approximately vertical.

The resulting domain trace diagram is shown in Fig. 3. The ruled lines indicate the domain class. The circles indicate a domain in which most of the particles were seen face-on; this domain was included in class U. Areal measures, as percentages, of these domains were: H, 41; I, 40; U, 19. The preferred horizontal orientation is evident from these percentages.

Plate 6 is a section of the failure zone of the triaxial test specimen. There is a rumple from R, at the bottom, to C, at the centre of the micrograph. The failure zone extends from C to the right. On the extreme right is some Vestopal containing a few particles detached from the main sample. Near C, the failure zone is divided into a primary zone, on the right, and a secondary zone, nearer C. The primary zone is continuous, and the particles in it are parallel to it. The secondary zone is discontinuous, and the particles are at about 30° to the primary zone. To the left of C is an unfailed part with a more random structure.

SUMMARY

Virtually nothing like either cardhouse or salt-flocculated structure has been found in the kaolin samples examined. The ultra-thin sections have shown: the development of turbostratic structure with preferred orientation during consolidation, and the orientation of particle in failure zones.

The deviation of the packet-counting data from Poisson distributions suggests that the replicas did not give a true representation of the sample, probably because the fracture surface is not representative.

Ultra-microtomy, whilst by no means perfect, is a most useful method; and improved techniques and auxiliary tests should increase this usefulness.

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