

The Bakelite Resin—Impregnation and the Microstructures of Kaolinite

by
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Introduction

THE natural soils contain a variety of constituents such as the clay minerals, fragments of minerals, iron oxides and organic materials. However, as the majority of the constituents have particle sizes less than 2 microns, physical methods such as X-ray diffraction and differential thermal analysis can be employed to identify the constituents easily, but these methods do not present us with clear pictures of the mutual arrangements of the finely divided particles of soils, called fabric. To obtain a clear idea of what goes on due to the consolidation of soils and the fabric change due to shearing action, an electron microscopic approach to the investigation appears to be the answer, as the resolution and the magnification of the optical microscopes is limited for such an investigation.

As the electrons in an electron microscope have a low penetrating power, if it is necessary that the specimen for such an investigation should be ultra-thin, preferably of a thickness not more than 1100°A . To obtain an ultra-thin section of such thickness it is necessary to impregnate the clay soils by some suitable binding materials. The primary object of the impregnation is to replace the pore fluid and air of a clay mass by a suitable binding material which can hold the clay particles strongly together and at the same time does not affect appreciably the original clay fabric after impregnation. It has a clear advantage over drying which makes the clay samples more fragile and difficult to work. Moreover, drying of saturated clay samples is associated with high volume shrinkage, resulting in a considerable amount of fabric disruptions.

This paper is primarily concerned in establishing Bakelite resin as a very promising clay impregnating material for electron microscopic investigation, a suitable technique for proper impregnation and the microstructural change within the shear discontinuities of sheared specimens subjected to external stresses and strains. Different types of resin impregnating media were used for the present research programme in order to establish the advantages and disadvantages of each impregnating medium and because comparisons seemed to be desirable. They were Araldites AY 18 and AZ 15, Vestopal W and Bakelites (1970), (1971). Although the resins

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have different impregnating properties and different recipes, the impregnation procedure for all of them was similar. It is not intended to go into details and discuss the complete resin impregnation and the properties of all the resins as this would be lengthy, thus outside the scope of this paper and may be seen elsewhere (1970), (1971). The Bakelite resin impregnation was chosen to discuss fully, as it has some advantages over the other resins from the point of view of ultra-thin sectioning.

The ultra-thin sectioning of the consolidated unsheared specimens followed the conventional procedure (1965), (1966). The electron microscope used for this research was Siemens Elmiskop II.

Sample Preparation and Slicing

The clay used for this investigation was 'supreme' kaolinite. It was obtained from English Clay Lovering Pochin & Company Limited and fractionated by sedimentation to give a close range of particle size between one to two microns (1970), (1971). It was found that the particle size of the kaolinite, after fractionation was primarily between 1 and 2 microns. A very small number of kaolinite particles finer than 1 micron and coarser than 2 microns were present within the mass of clay and could not be eliminated (1972). A 500 percent dilute suspension of the fractionated kaolinite was prepared, using distilled water only, stirred in a high speed mechanical stirrer for an hour after increasing the pH of the suspension to 8 by adding 0.1 N sodium hydroxide. After allowing the suspension to age for one or two days, it was poured into an oedometer cylinder of 65 cm high and compressed to a maximum load of 45 kg/cm². The material was subjected to an unloading and reloading cycle before being finally unloaded prior to removal from the consolidometer. The load increments were usually at an interval of one day and double the previous one.

From the consolidated kaolinite, 60 mm × 60 mm × 25 mm block samples were taken out and further trimmed to accept the brass wedges of the modified direct shear box (1971). The orientation of the block samples was varied at the time of extracting from the consolidated kaolinite. Thus the horizontal, vertical and inclined at 45° block samples were obtained from a cake of consolidated kaolinite. The terms 'horizontal', 'vertical' and 'inclined at 45°' refer to the directions of the preferred orientation of the particles of the block samples relative to the direction of the shearing action. Shear loading was carried out under normal effective stress (σ'_N) of 2.4 kg/cm² and the rate of shear deformation was 3.56×10^{-3} mm/min.

On completion of the shear tests (Figure 7) the samples were taken out of the shear boxes, the brass wedges being removed and the samples were sliced parallel to the direction of shearing action to produce thin sections of 60 mm × 25 mm × 2-3 mm thick. A razor blade was found to be the best tool for slicing. The 2-3 mm thickness was chosen as it was found that the dissection to a thickness thinner than 2 mm of a saturated sample destroys the original fabric of the sample. A thicker specimen of 4-5 mm usually resulted in only partial impregnation. The size of the ambient (consolidated unsheared) slices were usually 25 mm × 12 mm. Immediately after slicing, the dissected slices were transferred into a glass petri dish and distilled water was poured in until the slices were fully immersed.

The Bakelite Impregnation Technique

The kaolinite slices, submerged in distilled water were placed in a bath of 25 percent methanol—75 percent distilled water solution for one day and the concentration of methanol was increased subsequently by 25 percent each day until the slices were in 100 percent methanol. This process would ensure the gradual replacement of pore fluids of the kaolinite slices by methanol. After 100 percent methanol, the methanol was replaced by acetone in a similar manner, *i.e.*, acetone concentration was increased by 25 percent each day until the clay slices were saturated in 100 percent acetone. The 100 percent acetone stage was repeated twice to ensure the 'complete' removal of methanol and water, if any. The two stages will commonly be referred to as the methanol and the acetone stages of impregnation, and were carried out at room temperature (20°C—22°C).

After the acetone stage, Bakelite-hardener mixture was prepared, the recipe was :

- 100 gm of Bakelite R. 18,774/1,
- 30 gm of Hardener DQ 19,262 and
- 20 gm of acetone or 10 gm of styrene.

The acetone or styrene was used as a diluting agent. The choice of acetone or styrene depends upon the required pot-life of the resin-hardener system and the working room temperature. At the room temperature of 22°C, the setting time of the resin-hardener mixture with acetone as a diluting agent is at least double that of the resin-hardener system with styrene as a diluting agent, and almost equal to that of the resin-hardener system with styrene at a low temperature of 15°C. The advantage with the styrene is that unlike acetone, it does not act as a primary cause for volume shrinkage. The only disadvantage with styrene is that it makes the impregnated specimen more brittle, a disadvantage for ultratomey and it is advisable not to use more than 10 percent of styrene when it is going to be used as a diluting agent for clay impregnation.

The resin-hardener system with acetone as a diluting agent was of low reactivity, the pot-life of the mixture was not measured accurately, but believed to be about 10—12 hours at 22°C. The pot-life of the mixture increases with decreasing temperature; thus impregnation at 10°C—15°C *may be advantageous in certain cases.*

The acetone was drained off at the end of the acetone stage and resin-hardener-diluting agent mixture was poured into the petri dishes until it topped the clay specimens. The petri dishes were covered and the impregnation continued for at least three days. In the end the curing was done with gradual increase in temperature from 35°C to 105°C with two intermediate steps at 45°C and 65°C.

The impregnated and cured kaolinite samples were of dark golden colour, tough and flexible. Sometimes large cracks appeared at the time of hot-curing, particularly at high temperature, but usually not through the impregnated slices. As the cracks did not appear through the impregnated kaolinite slices, possibly the cracks did not induce fabric changes; any fabric change was essentially due to the volume change of the resin. It was found that the cracks appeared mainly due to the lack of mixing of the resin-hardener system. The buckling of the impregnated samples

was observed, and avoided by applying a dead weight at the time of curing and allowing the slices to cool at room temperature under pressure. Although the amount of disruption of soil microstructure due to high temperature curing at 105°C is difficult to estimate, it has been found that the effect can be insignificant (1972).

An LKB ultratome was used to obtain the ultra-thin sections. The procedure for adopting ultra-thin sections were conventional (1965) except for the micrographs (Figures 2—6) which were obtained using selected-area ultra-thin sectioning (1970), (1971). A diamond knife was used for obtaining all these ultra-thin sections, the floating fluid was distilled water only and the ultra-thin sections were collected on mainly 100 and 200 mesh copper grids. The electron microscopic magnifications were

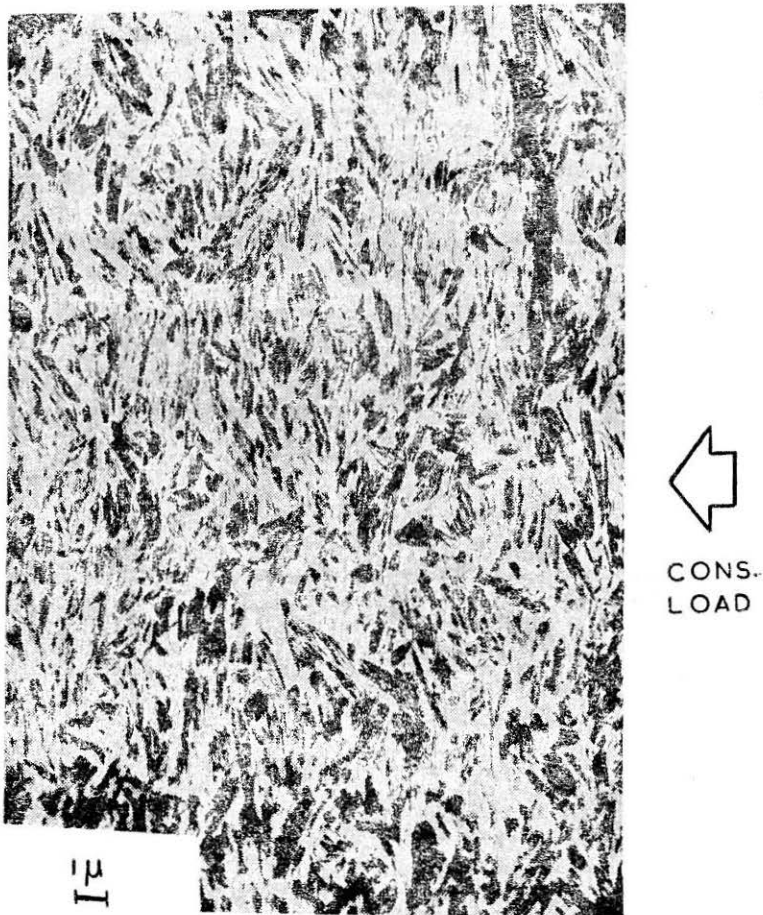
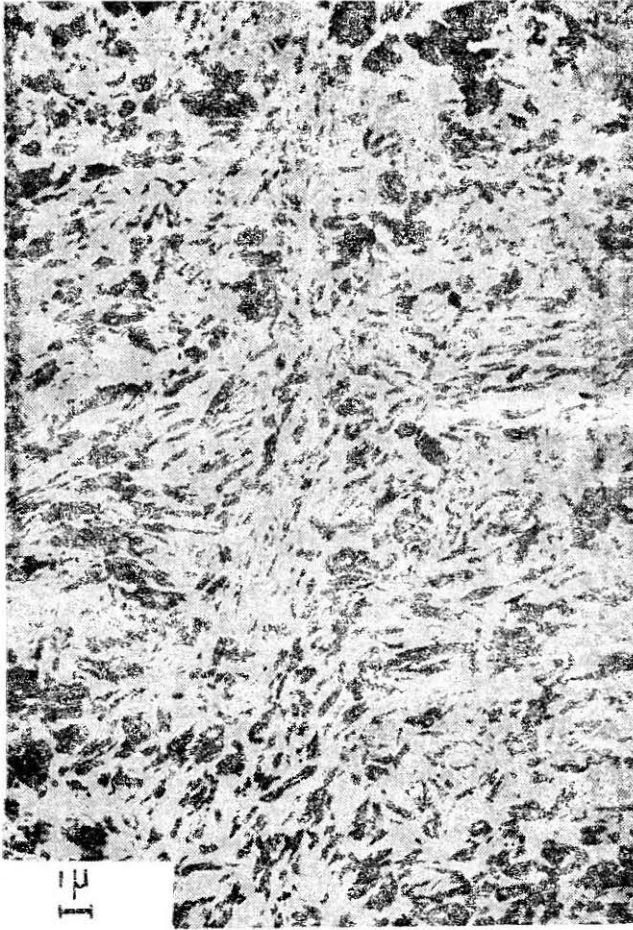
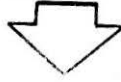


FIGURE 1: Electron-micrograph of thin section of 500°A through consolidated kaolinite impregnated with Bakelite, consolidated from 500 percent moisture content, pH 8, under 45 kg/cm².

CONSOLIDATION LOAD



1E

SHEAR
DISCON.

FIGURE 2: Electron-micrograph showing a major shear discontinuity in 'Vertical' material after peak stress. Thin section of Bakelite impregnated material, 800 \AA thick.

deliberately kept low so as to accommodate all salient features together in one micrograph.

Comparison and the Application of the Technique on Natural Soil

It is felt this method should be compared with other methods and the suitability and the effectiveness of this method on natural soil should be discussed.

Although Kay (1965) has compiled a few resin impregnation techniques for biological specimens, these are often unsuitable for the resin impregnation of clays, for the purpose of ultra-thin sectioning; the techniques may be suitable for replication of clays where a high degree of impregnation is not required. Another common method of impregnation is to immerse the clay in a bath of liquid Carbowax 6,000 for five to seven days at 65°C, so that liquid Carbowax can diffuse into the clay pores. This method of impregnation is suitable for optical microscopic investigation (1966), (1970), (1971), but is too soft for ultra-thin sectioning for electron microscopic investigation.

For the purpose of electron microscopic investigation of clay microstructures, an ideal clay impregnating material should have at least the

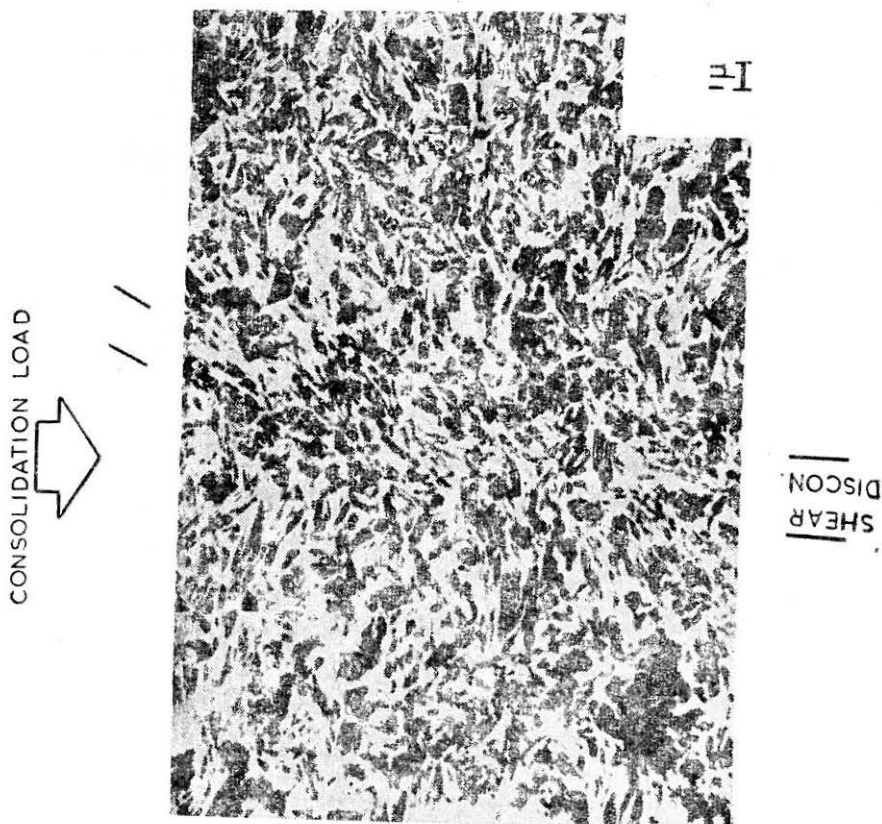


FIGURE 3 : Electron-micrograph showing microstructure of a major shear discontinuity in 'Vertical' material after peak stress, 1000°A thick.

following properties :

- (a) Low viscosity and flexibility, so that it can penetrate into the clay pores easily, and can hold the clay particles strongly together at the time of ultra-thin sectioning. This will allow the thin-sections to bend easily at the diamond knife edge and would cause few tears and holes.
- (b) Linear strain in any direction should be minimal.
- (c) The chemicals and the resins should not react with the clay particles.

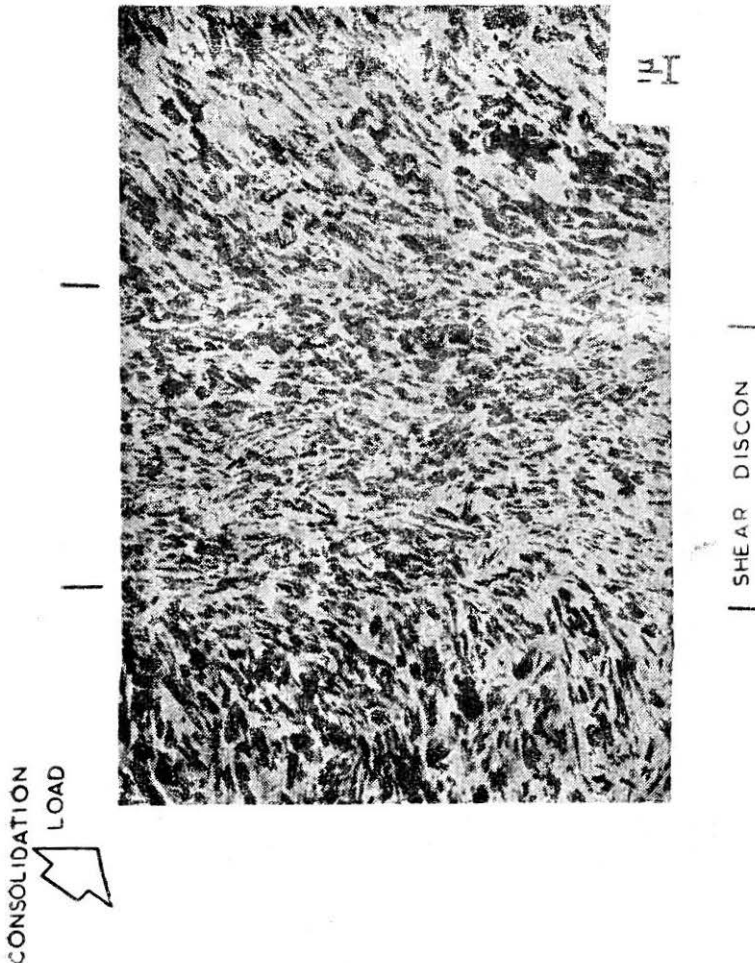


FIGURE 4 : Electron-micrograph showing micro-structural change, i.e., change of particle orientation and degradation, within a major shear discontinuity in 45° inclined material after peak stress. This section of Bakelite impregnated material, 700 \AA thick.

- (d) It must be either transparent or translucent to electrons so that the clay particles can be observed easily in the electron microscope.
- (e) It must be hard enough for ultra-thin sectioning and strong enough to withstand the electron bombardments.

The author has not come across any resin impregnating method for clays, other than the methods developed by himself (1971) which can

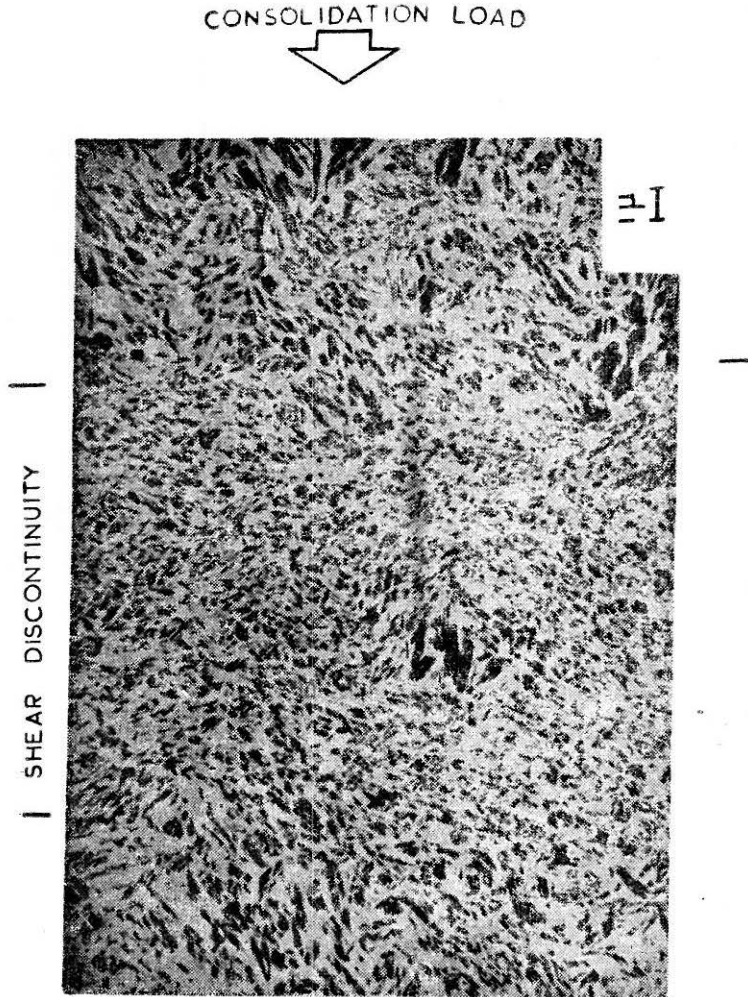


FIGURE 5 : Electron-micrograph showing micro-structural change of particle orientation and degradation, within a major shear discontinuity, in horizontal material after peak stress. The Bakelite impregnated material, 700°A thick.

CONSOLIDATION

LOAD

SHEAR DISCONTINUITY

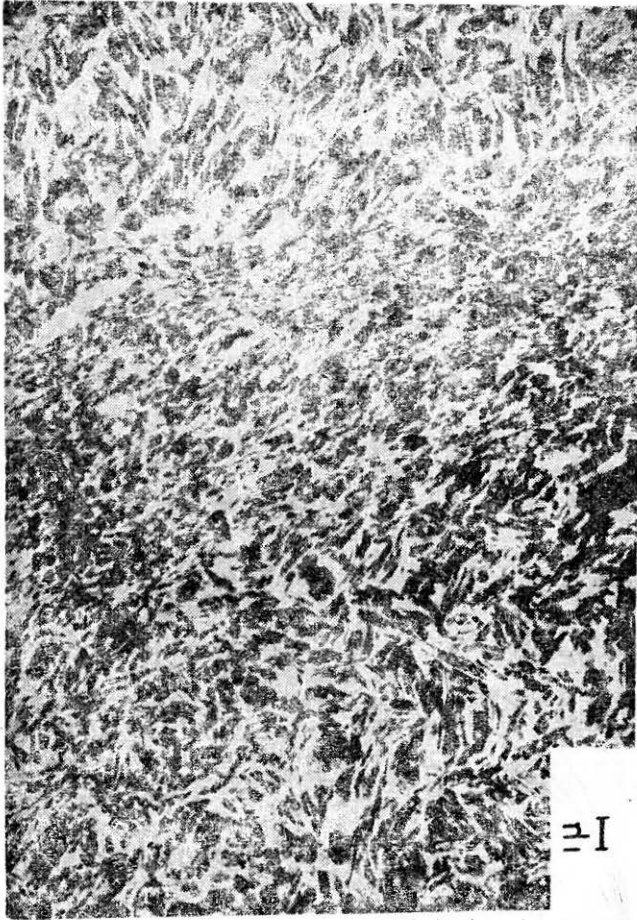


FIGURE 6 : Electron-micrograph showing micro-structural change of particle orientation and degradation, within a major shear discontinuity, in horizontal material after peak stress. Thin section of Bakelite impregnated material, 900Å thick.

satisfy all the above conditions, and hence a direct comparison is difficult.

The application of the above technique on natural soil was made and may be seen elsewhere (1971); the resin used for this purpose was Araldite AZ 15. However, it should be borne in mind that the replacement of acetone by the resin-hardener-acetone mixture is usually very slow for natural soil; for some natural soil; it may be too slow to impregnate, *i.e.*, the resin-hardener-acetone mixture may solidify itself before impregnating the soil specimen. The problem can be overcome by 'vacuum impregnation' or 'dynamic impregnation' (1970). A detailed study of all the methods and their application may be published later in this journal.

The Microstructure of Kaolinite

The microstructure of the consolidated unsheared kaolinite could be seen in Figure 1. The micrograph was taken from an ultra-thin section of 500 \AA thick and the clay particles appeared to have suffered some damage from the diamond knife at the time of ultra-thin sectioning. Comparing the particle damage in Figure 1 with that of Figures 2 to 6 (thickness of these ultra-thin sections was more than 700 \AA) it could be concluded that, although a higher resolution was obtained in the thinner section, Figure 1, thicker sections, in fact, present a more meaningful representation of the clay microfibrils. Again from Figure 1, in which the resolution is high, it should be observed that the 'larger and thicker particles' in Figures 2 to 6 consist, in majority of the cases, of several particles in face-to-face association. A strong overall particle orientation normal to the direction of consolidation could be seen in Figure 1.

The microstructural change due to the shearing action in a direct shear box, within a shear discontinuity is shown in Figures 2 to 6. The shear discontinuities and its neighbourhood areas of three samples whose directions of the shearing action were parallel, inclined at 45 $^{\circ}$ and normal to the direction of consolidation in the oedometer are shown in Figures 2 & 3, Figure 4 and Figures 5 & 6, respectively. Thus the average clay particle orientation outside the shear discontinuities in Figures 2 & 3, Figure 4 and Figures 5 & 6, could be seen as approximately normal, inclined at 45 $^{\circ}$ and parallel to the directions of the shear discontinuities, and the particle orientation within the boundaries of the shear discontinuities follows the direction of it.

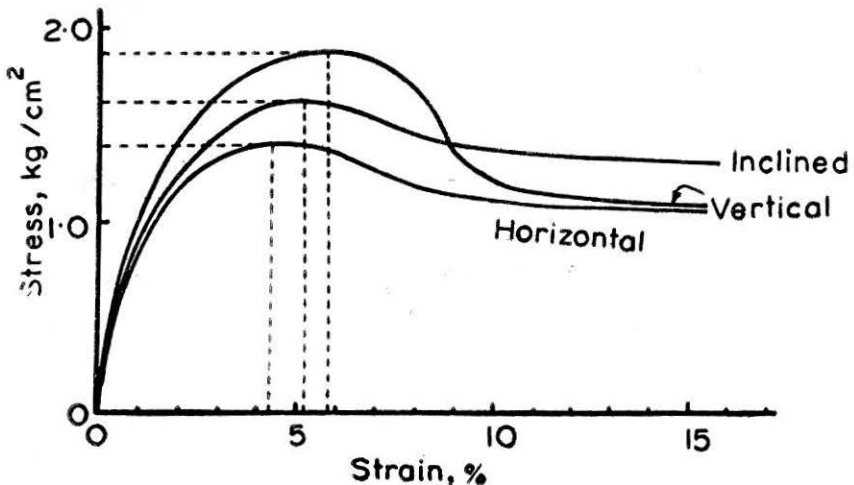


FIGURE 7 : Comparative shearing resistance graphs for horizontal, vertical and inclined specimens.

The degradation of the individual and 'flocks' of kaolinite particles within the shear discontinuities may be seen in all micrographs, particularly in Figures 4, 5 & 6. This degradation is primarily due to the interlocking of particles caused by the shear strain (1971). Although the degree of degradation mainly depends upon the amount of strain, it is

particularly noticeable that the degree of degradation of particles is highest in Figures 5 & 6 and least in Figures 2 & 3, whereas Figure 4 shows a considerable amount of degradation, all being subjected to an approximately equal amount of strain (Figure 7). Also Figures 5 & 6 show wider shear discontinuities compared to that of Figure 4 and much wider when compared to those of Figures 2 and 3. Thus higher degradation was found in the wider and least in the narrower shear discontinuities and this phenomenon is presumed to be related to the initial orientation of the kaolinite particles relative to the direction of shearing strain. As the initial orientation of the kaolinite particles in Figures 5 & 6 was parallel to the direction of the shear discontinuity, one would expect more slidings of the clay particles than the rotations of them compared to that of inclined or vertical specimens subjected to an equal amount of strain.

Let us now see how an external shear strain is engaged in the formation of a shear discontinuity. In case of a vertical specimen, whose direction of initial particle orientation is normal to the direction of the applied shear strain, initial consolidation resulted in front of the advancing face of a shear box specimen, causing a relative displacement and an uneven crack, on a microstructural scale. Further strain will rotate the clay particles within the close vicinity of the crack, along the direction of the crack. Thus a fair amount of strain and energy is absorbed in the rotation of particles. Any further strain will cause sliding and displacement of particles, causing interlocking of particles and degradation of them. In the case of horizontal specimens, whose direction of the initial particle orientation is parallel to the direction of the applied shear strain, a negligible amount of energy or strain is absorbed in the rotation of the particles, as most of them are in a favourable position for sliding over, and interlocking and degradation of particles start much earlier. In the case of an inclined specimen at 45° less energy is absorbed in rotation of the particles and the degradation of the particles starts earlier than that of the vertical specimens. Figure 7 shows the comparative shearing resistances of the three specimens. A peak stress occurred in the horizontal specimen at an early strain compared to the other two, the vertical specimen being the latest. Thus subjected to an equal amount of external strain in all specimens, the clay particle within the shear discontinuity of the horizontal specimen would be subjected to more sliding, as *sliding of particles takes place after the formation of a shear discontinuity*. Hence, all other factors affecting the shear tests being equal, it can be concluded that the width of a shear discontinuity primarily depends upon the initial orientation of the microstructures of clay and it is the amount of sliding of the clay particles within the shear discontinuity that primarily govern the width and the amount of degradation of particles within it.

Conclusions

The Bakelite resin was found to be an excellent one for impregnation of kaolinite. After curing, the Bakelite impregnated slices were tough and flexible, resulting in good ultra-thin sections for transmission electron microscopic observations. The microstructural investigation of the shear discontinuities revealed that it was the amount of sliding of kaolinite particles within the shear discontinuities that primarily governs the width of the shear discontinuity, and the amount of degradation of particles within it.

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