APPENDIX I

DIALYSIS OF COLLOIDAL PARTICLES

The tubing used for dialysis was of the type Gallenkamp DL/620 with an average pore diameter of 4.8 nm. The tubing allows low molecular weight compounds in the aqueous solution to diffuse whilst preventing the passage of higher molecular weight material.

Before use, the tubing was conditioned according to the following procedure recommended by the manufacturers.

(i) Boiling in 0.5 per cent acetic acid for 30 min.

(ii) Washing thoroughly in tap water and then in distilled water.

(iii) Boiling in 0.5 per cent sodium carbonate solution for 30 min.

(iv) Washing thoroughly in distilled water.

(v) Storage in a refrigerator, in distilled water.

One of the open ends of the tubing was first closed by tying the end into a knot. After transferring the colloidal suspension of cellulose particles the other end was sealed in a similar manner. The tubes were suspended in a tank of distilled water which was constantly replenished by a distilled water still. The syphoned outflow of water was controlled to maintain a regular water-level in the tank. A mechanical stirrir continuously agitated the water in the tank. All the equipment inside the tank (apart from the dialysis tubing), and the tank itself, was made of glass.

APPENDIX II

PREPARATION OF SPECIMEN SUPPORTING FILMS

Supporting films for mounting specimens, for subsequent examination in the electron microscope, should satisfy the following requirements.

(i) *Material* - The films should be made from materials of high transparency to electrons, i.e. substances of low atomic number (see section 4.1).

(ii) *Thickness* - In the 80 - 100 kV range of accelerating potential, the energy of the incident electrons involved require the use of thin films (< 20 nm, cf. section 4.1).

(iii) *Strength* - The films should be self-supporting and stable to a wide range of temperatures (cf. section 1.4) in order to withstand rapid temperature fluctuations.

Evaporated *carbon* films, satisfy the above general conditions and hence, were used in the present work.

A quick and efficient method for the production of supporting films on glass slides has been described by Johnson[20] and Dobb[276]. However, in the present investigation, thin sheets of freshly cleaved (i.e. atomically smoth) mica (approximately 2.5 x 7.5 cm^2) were used instead of glass slides, to deposit the carbon films. The mica sheets were placed in an Edward's coating unit (model 12E6/716), and the chamber evacuated to less than 1.333×10^{-2} Pa. A current of 60A (at 10V) was passed for about 0.8s, through two carbon electrodes in (initial) point-contact. Since the precision of the timing control on the coating unit was found inadequate for the production of suitably thin films, the following

technique (developed in this laboratory) was adopted with success. A piece of white glazed porcelain (opal glass) was placed inside the chamber beside the target. The thickness of the carbon film was judged by the brown coloration of the indicator which was visible only on the area not covered with oil. When the light brown colour is just detectable, compared with the white appearance of the oil covered area, the thickness of the film has been found to be about 5 nm[277]. In the present work, films were obtained when the indicator showed a light chocolate coloured deposit corresponding to an approximate thickness of 10 nm[277]. Carbon films prepared in the above manner were found to be of adequate mechanical strength and suitable thickness.

The next stage of the operation, namely the transference of the thin carbon film deposited on the surface of the mica sheet on to the specimen supporting grids, was accomplished in the following manner. Several grids were placed on a piece of copper gauze kept under distilled water in a special container[277]. The carbon film was carefully floated off on to the liquid surface and the water then gently drained off, thus depositing the film on the grids. After drying at room temperature the grids were ready to receive the specimens.

APPENDIX III

METAL SHADOW-CASTING

The shadow casting process, introduced by Williams and Wyckoff[278] has long been recognized as a useful method for enhancing contrast in the electron microscope. Subsequently, Hall[279] extended this method to measure the diameter of spherical and tubular shaped macromolecules. However, a series of technical difficulties have been encountered in attempts to exploit the potentialities of the technique. These include, granularity of the shadowing metal, problems encountered in stripping metallized deposits from the substrate in the case of replication, presence of contaminating deposits, and difficulties in finding sufficiently smooth and flat substrates. Some of the obstacles have been overcome by such refinements and improvements as discussed below. For instance, freshly cleaved mica has been used as a substrate, the surface of which is not only very smooth, but also highly hydrophilic which is an additional desirable property.

A fundamental requirement of the shadowing metal is that it should be a good electron scatterer. Metals such as gold, platinum, palladium, uranium and chromium have been used with varying degrees of success.

The granular size of the metal is obviously the limiting factor of the shadowing technique. The error involved in particle size measurements becomes worse with increasing shadow length to height ratio. Granularity can also enhance anomalous results related to phase effects in electron optics (cf. section 2.2).

When used for measurements of isolated single units, such as particles of cellulose, the shadowing technique suffers from further, serious disadvantages. The metal coating can, in many cases, obscure

the shape of such small particles, thus preventing reliable measurements. This difficulty has been studied extensively by Ohad, Danon and Hestrin[81,82]. They have reported that when the thickness of the film is between 1.0 and 2.0 nm, and the height/shadow ratio is between 0.2 - 0.1, the added width will be between 5 and 20 nm, which can be a value considerably larger than the structure studied. Further, they worked out a method for the adjustment and conversion of measurements obtained from metal shadowed, isolated particles. Colvin[79], however, has criticized some of these conclusions and stated that metal shadowing as commonly practised need not necessarily lead to such erroneous results as suggested by Ohad *et al.* There is yet another difficulty, common to both shadowing and staining techniques, in that it is not always possible to obtain particles appropriately distributed on the surface of the supporting membrane, free of impurities and drying artifacts. In such aggregates of particles, the shadowing method would, if the coating is relatively thick, no longer resolve the closely packed constituents. Some of the above mentioned anomalies encountered in the shadowing technique, may well be responsible to some extent for the different values quoted for the dimensions of the "ultimate cellulose unit".

In the present study, metal shadow casting was used on the particles of cellulose, in the preliminary stages but was later abandoned in preference of negative staining methods.

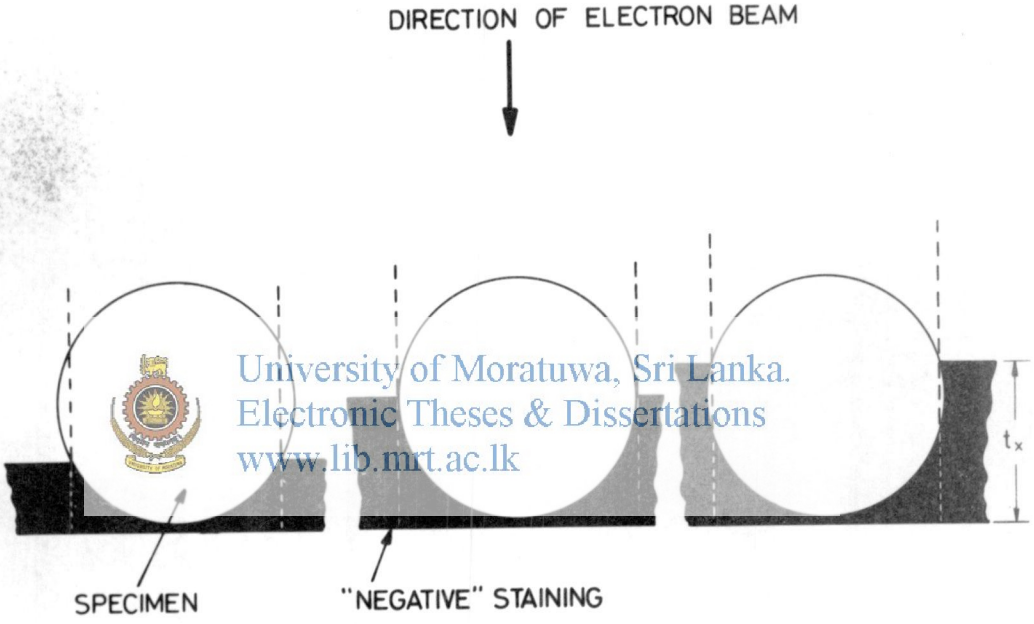


FIG. A1 DISTRIBUTION OF "NEGATIVE" STAINING MATERIAL

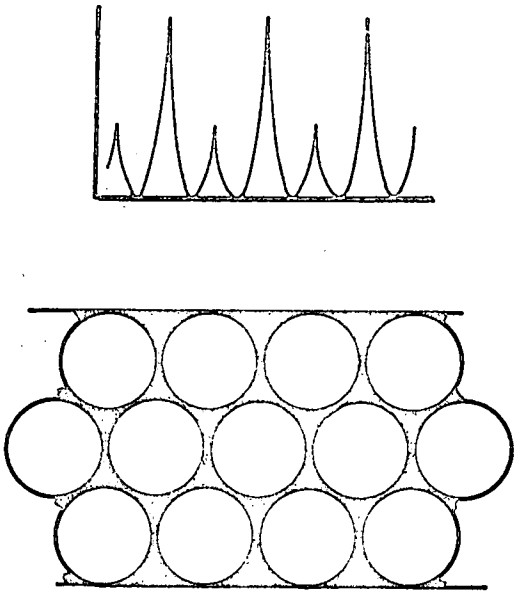
APPENDIX IV

NEGATIVE STAINING

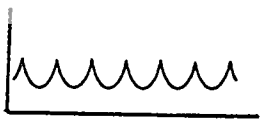
In electron microscope studies of biological material, it is often necessary to use some type of non-specific staining procedure, in order to achieve adequate specimen contrast. The method has also been extended to measure the dimensions of macromolecules, but unless extreme caution is excercised in the interpretation it may lead to erroneous results as discussed below.

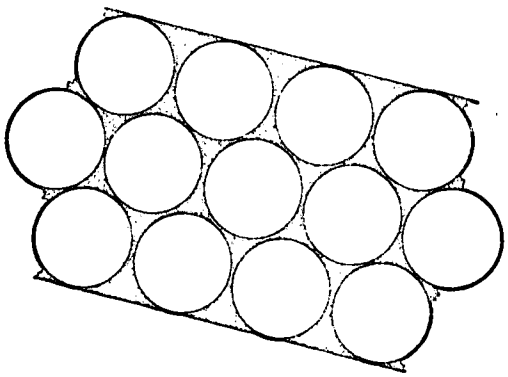
Negative staining involves the surrounding of a relatively "transparent" specimen by an electron "opaque" matrix. The resultant image need not necessarily be a true replica of the specimen; in fact it is a derivative structure which is observed as a shadowgraph of those parts of the specimen not penetrated by stain. However, penetration of the stain greatly complicates the interpretation of negatively stained specimens.

Negative staining assumes little or no adsorption of the electron opaque material by the object under examination. Silver nitrate, phosphotungstic acid (PTA), osmic acid, and uranyl acetate have been used in the past to stain cellulose. However, Colvin[79] has reported that on occasions (for unexplained reasons), PTA was found to be adsorbed strongly to native cellulose microfibrils. According to Preston[93], the cellulose microfibrils in plant cell walls possess a crystalline core surrounded by a "cortex" of paracrystalline material and therefore, it is conceivable that the stain would permeate the outer region of such a system and reveal unstained crystalline core. If Preston's proposition is correct, then negative staining will always yield an underestimate of the fibrillar dimensions of cellulose.



(a) End view of parallel microfibrils (white circles) with the interstices filled by the negative stain, together with the intensity distribution when viewed from above.





(b) The intensity distribution when the system in (a) is tilted through 12.5°.

FIG. A-2 NEGATIVE STAINING AND DISTRIBUTION OF INTENSITY (ref. 93)

Another factor, which would lead to similar anomalous results, has been pointed out by Colvin[79] and is illustrated in FIG. A-1. Since the electron scattering material is used to outline the specimen, if the shape, size and distribution of the specimen permits encroachment of the stain, (not applicable for objects with square or rectangular cross-sections) the apparent width will depend on the relative thickness (t_x) of the layer of stain. Colvin[79] has suggested that such erroneous results may be avoided by comparison of the shape of a microdensitometer trace across the images of the objects with known morphology, but in practice the situation can be further complicated by other factors as discussed below.

Millward and Sikorski[94] highlighted one of the most significant causes leading to misinterpretation of high resolution electron micrographs of thin sections (transverse and longitudinal) of mammalian keratin. Using an idealized diagram they were able to show clearly one of the main reasons for the variation of the apparent width of a single unit in a regularly packed system of aggregates. Subsequently, Preston[93] has extended the same argument for the case of cellulose particles as shown in FIG. A2. If the particles are not regularly packed and are not uniformly straight and parallel (as in the real case) the smallest centre to centre distance would still be less than the real value. Thus, it would appear that the results from the use of "negative" stains in the case of *isolated* particles would not suffer from the above discrepancy.

In the present study, a 2 per cent aqueous solution of uranyl acetate was used for staining cellulose particles and the measurements were confined to isolated units (cf. section 2.3.3).

APPENDIX V

WIDE ANGLE X-RAY DIFFRACTION PROCEDURES AND ANALYSIS

(a) Specimen Preparation

Cotton fibres cleaned following standard procedure (cf. section 2.3.1) were parallelized as far as possible by careful combing and made into a thin compact cylindrical bundle of 1 - 1.5 nm diameter. The fibre bundle was consolidated by tying the ends with a sewing thread and clamped taut in a frame.

(b) X-ray Generator and Camera

A commercial "Hilger and Watts Y90" X-ray generator was used incorporating a Philips 1kW, permanently sealed tube with a copper anode, operating at 36 kV, and a tube current of 10 mA. The radiation generated is *mainly* copper K_α, having a wavelength of 0.1542 nm together with some "white" radiation.

The flat-film camera was fitted with a glass-capillary collimator (D ≈ 0.5 mm), over which a nickel foil was fixed, in order to absorb most of the copper K_β and white radiation. The specimen was clamped over the end of the collimator, and the film held in a casette with a mask of aluminium foil. The specimen to film distance was kept at about 3 cm (see (d) below- calibration) and an exposure time of 3.5 hr.

(c) Recording of X-ray Diffraction Patterns.

The diffraction patterns were recorded on a "slow" film (Crystallex, Kodak Ltd.) in order to obtain clear patterns with a minimum of background darkening. The film was developed for 3 minutes at 24°C in 4:1 (V/V) aqueous DX-80 developer and, subsequently, washed and treated with 4:1 (V/V) aqueous FX-40 fixer.

(d) Calibration of Specimen to Film Distance

A silver foil[280] placed in the middle of the fibre bundle was used to calibrate the specimen to film distance.

The relevant crystallographic data for silver obtained from *American Society for Testing Materials*[281] is as follows.

TABLE 30

CRYSTALLOGRAPHIC DATA FOR SILVER

d_{hkl} (nm)	I/I_1
0.236	1.00
0.204	0.53
0.144	0.27
0.123	0.53
0.118	0.05

d_{hkl} silver = 0.236 nm, for the most intense reflection.

$\sin\theta = \frac{\lambda}{2d}$, Bragg's law (n=1)

λ = 0.154 for Copper K_α

$\therefore \sin\theta = \frac{0.154}{2 \times 0.236}$

$= 0.326$

$\theta = 19^\circ 3'$

$2\theta = 38^\circ 6'$

$\tan 2\theta = 0.784$

If the specimen to film distance = x and distance of the diffracted beam from the undeviated beam, measured along the film = r, then tan $2\theta = \frac{r}{x}$

Radius of silver ring measured on the film = r = 2.46 cm

$$\therefore \quad x = r \tan^{-1} 2\theta$$

$$= \underline{3.14 \text{ cm}}$$

(e) Regression Analysis of Crystallite Orientation and Some Properties of Native Cotton Fibres.

A regression analysis of the following data was carried out using a built in programme for multiple linear regression analysis in the Leeds University ICL 1906A computer[282] (cf. § 3.4.3, and Table 23).

TABLE 31

DATA FOR REGRESSION ANALYSIS

TYPE OF COTTON	SEMI-AZIMUTHAL ANGLE OF 002 REFLECTION MEASURED AT HEIGHT			ELONGATION AT BREAK %	TENSILE STRENGTH (g/tex)	NO. OF CONVOLUTIONS (cm^{-1})
	40%	50%	75%			
Acala	26.6	22.9	15.2	5.8	45.4	77
Lankart	36.5	33.7	26.5	8.2	33.8	80
Menoufi	27.2	22.0	18.7	5.7	51.9	53
Tanguis	31.0	27.7	20.0	6.5	40.9	70
Lambert	25.5	22.6	15.4	6.3	48.4	62
Uganda	29.4	26.5	20.6	5.6	41.3	75

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