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THE GENERAL STRUCTURE OF FIBRES

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PLANT fibres are elongated cells, that is, grown objects whose structure can only be fully understood from a development point of view. The young differentiating fibre cell has a very thin wall, consisting of an amorphous matrix of pectic and hemicellulosic material reinforced by only a few per cent. of cellulosic microfibrils. Curiously enough, this percentage (say, 5 per cent. by volume) corresponds approximately to the amount of iron rods in reinforced concrete! The microfibrils with a diameter of about 250 Å are arranged in a dispersed interwoven texture (Fig. 6). This so-called primary wall contains in the living cell some 90 per cent. of water and, of the technically important dry matter, only less than half is cellulose, which alone is left over in the macerated preparations for the electron microscope. The growth of the primary wall consists in a widening of the existent texture combined with a continuous neoformation of new wall lamellae (multi-net growth). The differentiation of pit fields and bordered pits occurs during this growth. The pit areas no longer increase their surface, though their distance may still considerably increase (mosaic growth, Fig. 5).

Towards the end of the described extension growth, more and more microfibrils of cellulose are produced, so that a wall layer results which has no longer the same aspect as the primary wall. We have called it the transition lamella (*Übergangslamelle*, Fig. 7). It is possible that it is identical with the outer layer of the secondary wall according to the terminology of Bailey (Fig. 1).

The bulk of the fibre is formed by the secondary wall, which grows by apposition and, therefore, appears to be lamellated. In these lamellae, the microfibrils of cellulose are so densely packed that they must be arranged in parallel (parallel texture — typical features of this texture are the so-called slip planes (Fig. 8) and 'geometrical cavities'). They touch one another

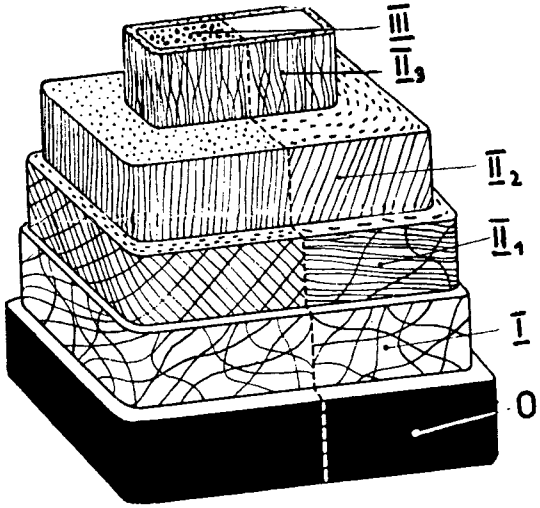


Fig. 1 — Organisation of the fibre cell wall.
Left half — wood fibre of birch; right half — tracheid of spruce (from H. Meier's doctorate thesis 1955)

0	—	middle lamella	II_2	—	bulk of secondary wall
I	—	primary wall	II_3	—	inner layer of secondary wall
II_1	—	transition layer	III	—	tertiary lamella

laterally and form characteristic fasciations. As a result, the microfibrils, which in the primary wall are clearly individualised, fuse laterally and lose their individuality. For this reason, it is difficult to find substantial values for the diameter of cellulose fibrils in the secondary wall; as a matter of fact, their width depends on the method by which the fibre is disintegrated. If only a mechanical fibrillation is applied, microscopic macrofibrils result (Fig. 2); if chemical maceration is made use of, sub-microscopic microfibrils are observed (Fig. 3); but these can be further split into micellar strands or elementary fibrils by ultrasonic treatment (Fig. 4). In Table 1, the dimensions of these disintegration products are listed for cotton.

TABLE 1

Fibrillar elements observed in cotton

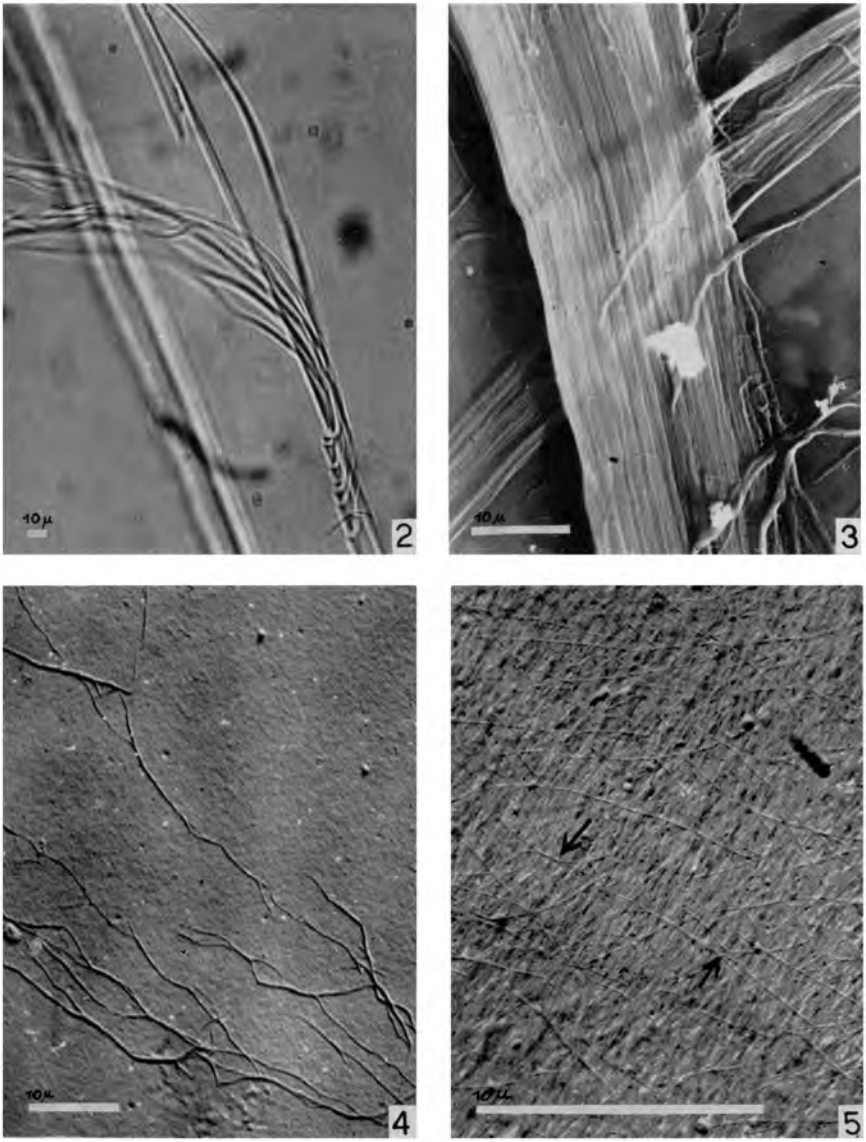
	Area of cross-section	*Number of cellulose chains on cross-section*
Cotton hair	$\pi(10\mu)^2 = 314\mu^2$	1,000,000,000
Macrofibril (Fig. 2)	$(0.4\mu)^2 = 0.16\mu^2$	500,000
Microfibril (Fig. 3)	$(250\ \text{Å})^2 = 62{,}500\text{Å}^2$	2,000
Elementary fibril (Fig. 4 and 5)	$\left.\begin{matrix}50\text{Å}\times60\text{Å}\\30\text{Å}\times100\text{Å}\end{matrix}\right\} = 3{,}000\text{Å}^2$	100
Cellulose molecule	$4\text{Å}\times8\text{Å} = 32\text{Å}^2$	1

The elementary fibrils seem to be ideally crystallised, while the spaces between them are filled partly with paracrystalline cellulose and partly with non-cellulosic material, which can be removed by alkali treatment. The specific weight of bleached ramie (1.39) is considerably less than that of crystallised cellulose (1.59). Consequently, the secondary wall is porous by submicroscopic capillaries. These are of varying dimensions. There are coarser pores where lignin is located and colloidal dyestuffs can penetrate and much finer heterogeneities where only micromolecules such as water and iodine can penetrate (heterocapillary system). The problem of topochemical reactions and transformation of fibres is discussed.

Throughout the growth of the fibre cell there exists a tertiary lamella at the boundary between the living cytoplasm and the fully differentiated secondary wall. It is chemically different from the secondary wall and displays in certain cases special morphological features (warts, Fig. 9).

Artificial fibres do not show such a complicated morphology and, therefore, have a simpler and more uniform structure.

Full particulars of the problems discussed and an extensive bibliography will be found in the forthcoming monograph *Die pflanzsliche Zellwand*, A. Frey-Wyssling (Springer Verlag, Berlin, 1958).



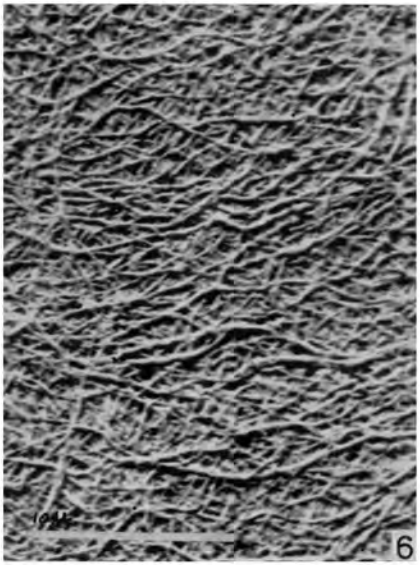
Fibrillation of the secondary wall

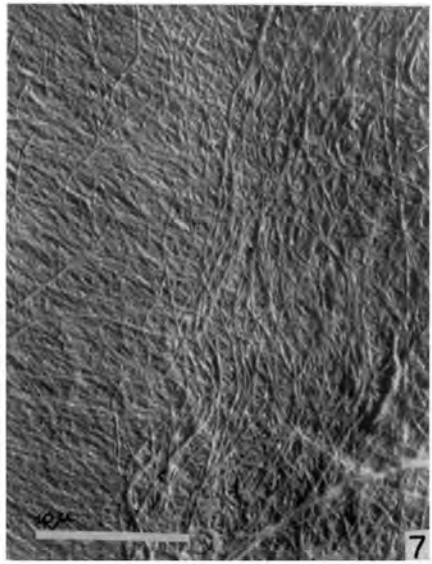
Fig. 2 — Macrofibrils of a fir tracheid (light microscope)

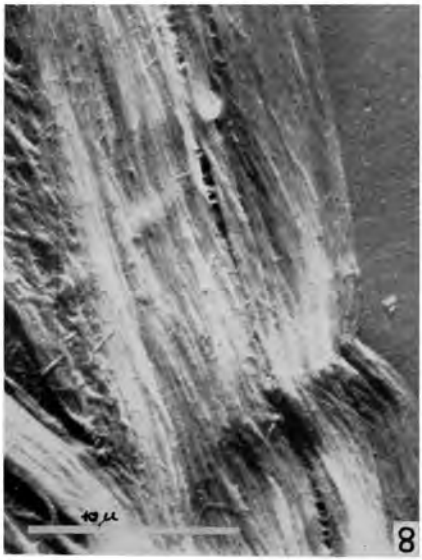
Fig. 3 — Microfibrils of a ramie fibre } (photographed by

Fig. 4 — Disintegration of microfibrils by ultrasonics } K. Mühlethaler)

Fig. 5 — High resolution of the microfibrils of the primary wall discloses their composition of elementary fibrils, as shown by arrows (photographed by H. Moor)







Sub-microscopic features of the different wall layers

Fig. 6 — Primary wall of the cambium-dispersed texture (photographed by H. H. Bosshard)

Fig. 7 — Transition layer-crossed texture } (photographed by

Fig. 8 — Secondary wall-parallel texture with 'slip plane' } K. Mühlethaler)

Fig. 9 — Warts on tertiary wall

DISCUSSION

DR. W. GALLAY: May I ask Prof. Frey-Wyssling why he starts with the elementary fibril as an entity? After having stated that the so-called macrofibril and the microfibril can be observed over a wide range of diameters depending on the method of preparation and therefore are not distinct entities, why not confine ourselves to the individual cellulose molecule as the only true entity?

PROF. A. FREY-WYSSLING: It is not possible to disintegrate elementary fibrils by mechanical means. You must treat them with cellulose solvents that destroy all the hydrogen bonds in order to obtain individual cellulose chains. Morphologically, therefore, it is not really the chain that is the element; it is the elementary fibril.

MR. L. G. COTTRALL: How does Prof. Frey-Wyssling envisage that the strength of a lignin-free fibre is derived? We are given to understand that the cell wall is made up of masses of microfibrils oriented in different directions in the various layers. There must be some attachment between these small elements, either a mechanical attachment (an entanglement) or else a bonding — alternatively, a combination of both. How does Prof. Frey-Wyssling conceive that this attachment between the microfibrils occurs? If there is any bonding, has he any idea of the proportions to which these elements take part in the bonding? — what is the number of bonds per unit area?

PROF. FREY-WYSSLING: The situation is this. We do not really know the length of the elementary fibrils, but, compared with their diameter, they are practically of indefinite length. If they are side by side and there is mutually only very weak bonding — sometimes a hydrogen bond here and there — this very long surface makes an even stronger bonding than the sum of the covalent bonds of the molecule chain in the cross-section of the elementary fibril. The longitudinal area of contact is indefinitely much higher than the cross-section, so the number of bondings per unit area can be small. Then, of course, if there is hemicellulose left between the fibrils, much stronger lateral bonding results, exceeding many times the tensile strength of the elementary fibrils.

DR. B. G. RÅNBY: Last time we met (in 1954), we discussed the dimensions of the sub-microscopic fibrils and debated the existence of the 250 Å fibrils, already in question at that time. Today, we find that a famous member

of the Royal Society has joined the '100 Å school'; we are pleased therefore not to have to discuss these matters any more. We agree that the fibrils are about 100 Å wide and frequently aggregated. We must be a little cautious about how elementary the fibrils are and how well defined they are, because there are indications that the crystallisation takes place after the chains have been formed and are deposited. At least, we have been able to show that in the case of a *Dictyosteleum* slime mould. The reason the dimension 100 Å is preferred seems to be related to an interplay between different (thermodynamic, kinetic) forces, which make the cellulose chains crystallise to fibrils that happen to be about 100 Å wide for wood fibre cell walls. Somewhat wider fibrils have been found in animal and algal cellulose (in tunicates and *Valonia* algae, respectively).

I should like Prof. Frey-Wyssling to discuss the difference between the 100 Å fibrils in the primary wall and the fibrils of the same dimension in the secondary wall, because they do not seem to be identical, according to some reports I recall.

Another point is that the slip planes in the cell walls which were first discussed by Prof. Frey-Wyssling and later by Dr. Bucher, seem to me to be related to slip planes in the cellulose lattice of the fibrils. Such slip planes in the lattice would cause irregularities (disorder in the hydrogen bonding) and thus make the cellulose chains more susceptible to chemical attack — for instance, to heterogeneous hydrolysis or oxidation.

My third point is what proof is there of an 'interwoven' fabric of fibrils in the primary wall? I am not convinced by the evidence put forward in support of it. It would be interesting to have it discussed further.

PROF. FREY-WYSSLING: I will start with the last item first — the question of weaving. If you look carefully at the electron micrographs, you will find an arrangement like that in Fig. A. I would like to discuss this situation in terms of a familiar game — Pick-up-sticks. You have a bundle of long sticks (the microfibrils), you let them fall and then the game is to take away one stick after another. The man who wins can take all of them away without disturbing the others. Those who play this game know very well that, if they get to the position where the sticks are entangled as in Fig. A, they have lost the game. In one respect, I think the criticism of Dr. Rånby is right. The weaving is not like the warp and weft weaving of a cloth, as we thought in the very beginning, but it is an entanglement. Its origin can be explained as follows. If the layer where the microfibrils grow is not just of the dimension of the diameter of the microfibrils, but is somewhat thicker,

they will not grow strictly parallel to the surface, but at a slight angle to it and an entanglement, as illustrated is possible.

At the discussion on microfibrils at the Cell Biology Symposium in St. Andrews some weeks ago, evidence was put forward that there is bipolar tip growth of the elementary fibrils in bacterial cellulose. If it is permitted to transpose this to the cytoplasm, these fibrils will grow into an entanglement.

In the electron microscope you find a distortion in the fibrils (*see* Fig. B). My argument on these slip planes is the following. Solubility, staining and so on are functions of the fibre's density. If the fibrils are separated by such distortion, a place exists where easier penetration is possible for chemical agents. I cannot imagine that the grid has been broken or attacked by the mechanical forces involved. It is more important that the surface of the fibrils, formerly densely packed, has been freed.

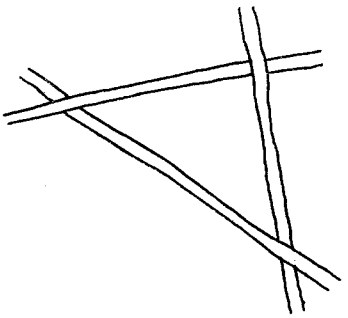


Fig. A

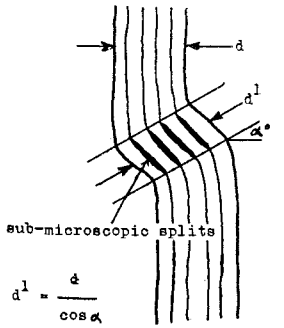


Fig. B

DR. RÅNBY: I think that is one effect, but the question is — how much bending can a lattice take before the lattice itself slips?

PROF. FREY-WYSSLING: We have measured these angles; they are about 30°. In an X-ray pattern of such a bend, you would find no difference from a straight fibre. On the other hand, the loosening of the parallel texture is quite obvious.

There is only a small chemical difference between the fibrils in the primary and secondary walls. The cellulose of the parallel-textured secondary wall may be crystalline to 70 per cent., that of the disperse-textured primary wall to 40 per cent. or 50 per cent. — that is, a similar crystallinity as in bacterial cellulose. In the first case, degrees of polymerisation of 3 000 – 5 000 are found; in the second case, it is slightly less — 1 000 – 2 000. Thus, there is no real difference in the chemical properties of these two celluloses.

As to the diameter of the sub-microscopic fibrils, they are rather coarse in the primary wall. As a rule, they range about 250 Å and are well individualised: this is why I insisted on the existence of microfibrils of this size. It was only recently that we could disintegrate the primary fibrils into somewhat twisted elementary fibrils.

DR. F. MULLER: Would Prof. Frey-Wyssling tell us how to identify the elementary fibrils with the crystalline regions assumed by so many authors in a cellulose structure?

PROF. FREY-WYSSLING: This is difficult to say. With the electron microscope, we cannot distinguish between crystalline and paracrystalline cellulose — that is, between the crystalline core of the elementary fibrils and its less orderly arranged surface layers. Formerly, this was known as amorphous cellulose; now we call it paracrystalline cellulose. The thickness of this layer can be calculated, taking 70 per cent. crystallinity, estimating how the cross-sections of this core would be shaped and then deducing the thickness of the layer of the non-crystalline cellulose. I think Dr. Rånby agrees that what is seen with the electron microscope is the border of the paracrystalline cellulose. This layer is very important, because it can make contact with neighbouring elementary fibrils and form hydrogen bonds.

DR. MULLER: I assume we must accept the concept of chain molecules running from one elementary fibril over to the other?

PROF. FREY-WYSSLING: That is difficult to say, because all our discussion bears on the cross-section; therefore, we cannot see on our diagram whether such a pair of elementary fibrils will merge into one another in other planes. Therefore, the question you have put forward cannot be answered in a positive or a negative sense. Have you ever seen branching elementary fibrils, Dr. Rånby?

DR. RÅNBY: No, in no case was it clearly branching, unless it was an aggregated thicker string. In these cases, you could see a twist of the strings, indicating a fibril bundle, but branching does not seem to be very frequent. The differences between the fibrils in the primary and secondary wall as described by Prof. Frey-Wyssling are significant and very interesting to me.

DR. MULLER: I have another question, this time for Dr. Bucher. Is anything known about the differences in chemical composition between the tertiary and the secondary wall, because the differences in their behaviour should, I think, be caused by differences in chemical composition and not only by differences in structure?

DR. H. BUCHER: I think there is a difference in chemical character. Victoria Blue has the property of an indicator — blue in acid solution, red in alkaline solution. That the tertiary wall remains blue with this stain even in alkaline swelling agent could be explained by the reaction of the dyestuff with the substance of the tertiary wall.

What is this substance? It has a special affinity for the dyestuff, but I think this is not a very exact interpretation. It probably consists of pure cellulose and hemicelluloses with acid groups. From their investigations, Meier and Yllner in Stockholm conclude that the tertiary wall might consist of xylans.

DR. H. MEIER: Do the tertiary walls with flat helices in spruce belong to springwood fibres and the tertiary walls with steeper helices to latewood fibres?

DR. BUCHER: It is difficult to identify swollen fibres. Wide-lumened tertiary walls that have windings at an angle of 65° may originate from springwood fibres, the narrow-lumened walls with 30° windings from latewood fibres.

MR. H. W. EMERTON: May I show two slides that bear on the subject discussed by Dr. Bucher? These are of the freshly cut surface of spruce (*P. excelsa*) and viewed in Fig. C across the lumen at the inner surface of the cell wall. Rather more than the width of three tracheids is shown. In the lefthand cell, the inner secondary (or tertiary) wall, S3, is present almost throughout; towards the bottom of the picture, it has probably been disturbed by the knife. The righthand cell (with the bordered pits) shows similar features. In the central tracheid, the striations of the middle secondary wall only are evident, almost axial in this case: it cannot be said whether S3 has been removed by the knife or whether it was never present. We have observed S3 in this way many times in spruce and the results obtained are supported by the electron micrographs taken by my colleague D. H. Page (Fig. D). There appears in this species to be a fibrillar helix making an angle, with respect to an axial direction, typically of approximately 65°. This helix is of the S-form. Surrounding this in the cell wall (and therefore underlying it in pictures of this kind) is at least one other layer of S3, the microfibrils of which are often observed to be almost transverse. Has Dr. Bucher any evidence of two fibril systems being present in this wall?

DR. BUCHER: Fig. 22 of our first publication in 1953 on the tertiary lamella was taken before we did our measurements on helical inclination and it shows typical striations on a spruce fibre with slight sinoid form, indicating that it is a helix. We found transverse orientation visible in one part and

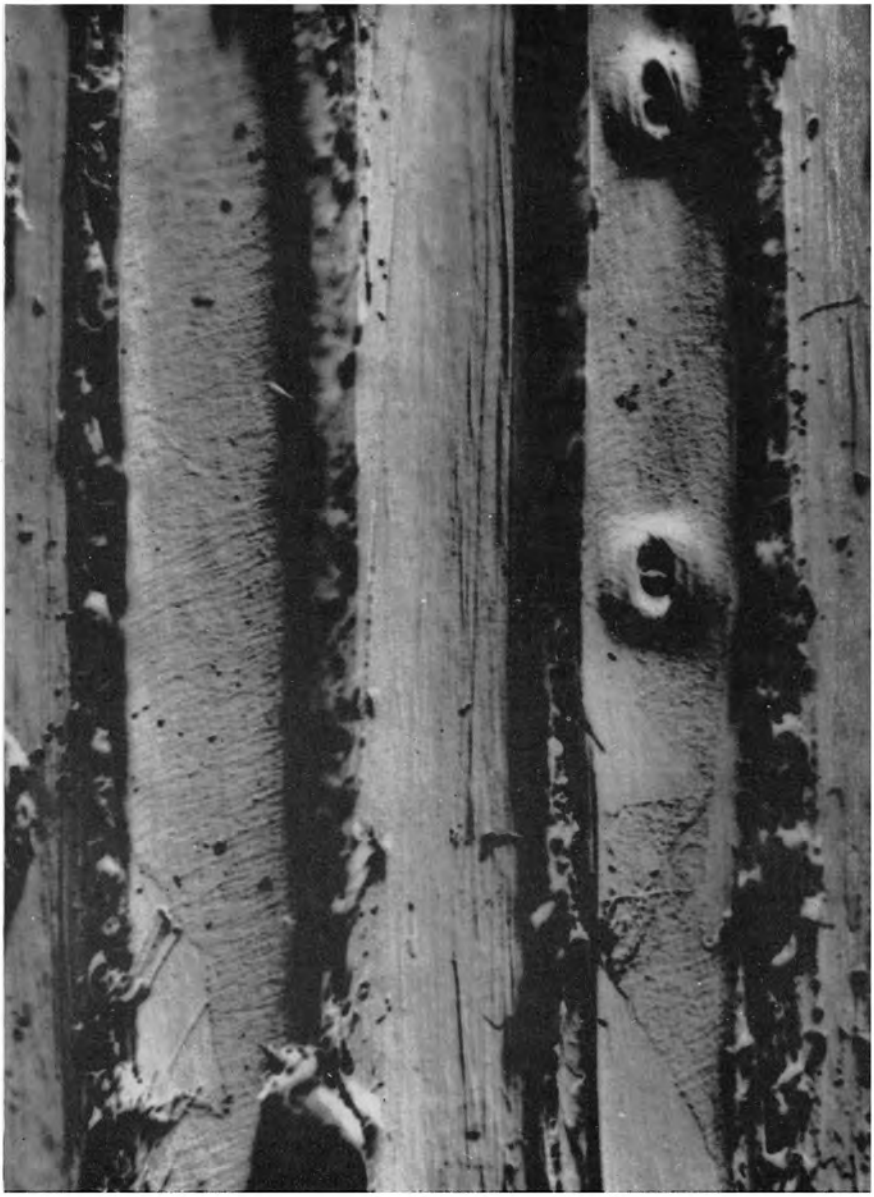


Fig. C — A radial surface of Norway spruce — the cut has passed through the lumina (light micrograph × 1020)

steeper orientation in another (65°). I showed the picture to a mathematician to have him interpret the helix that must be present. Although he had no idea what the subject was, he found that there must be *two* different helices. There exists no possibility of a *single* helical system producing winding features as seen in this picture (Fig. 17, page 26).

DR. D. ATACK: The so-called slip planes are very interesting features, since they appear to be points of initiation of failure in wood. The initiation of failure in metals is supposedly controlled by defects in the structure — is there any defect in the wood structure to give rise to the localised folding (unfortunately called slip planes) under compression, which may be identified microscopically? In this example, I do not wish to imply that the slip planes in wood are analogous to those observed in some metals. Robinson some 30 years ago called slip planes the dislocation lines that appear in wood under stress and observed them very frequently in wood from which lignin had not been removed.

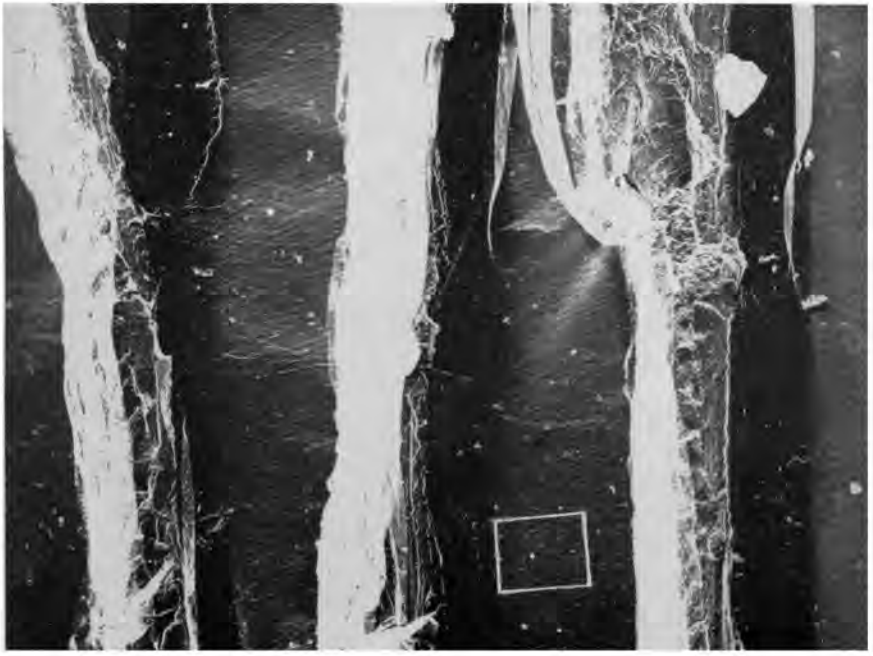


Fig. D (*i*) — As Fig. C (electron micrograph × 900)

Could either speaker suggest any particular morphological feature in the wood that may be associated with the formation or the initiation of these slip planes? Furthermore, what determines the angle of the slip planes?

PROF. FREY-WYSSLING: In my experience, the angle depends on how much the loosening takes place. If the texture is loosened very much, the angle is steeper; if the loosening is small, it is flatter. If there are two adjacent tracheids, dislocation will go through to the next wall at the same angle. As shown in the illustrations, even if there are several dislocation planes, they are all at the same angle. It is interesting that the compression necessary to produce them is about one tenth of the crushing pressure of wood. I should like to insist that these dislocation lines have nothing whatsoever to do with slip planes. In lignified fibres, they are rather rare, because it is much more difficult to produce the necessary distortions.

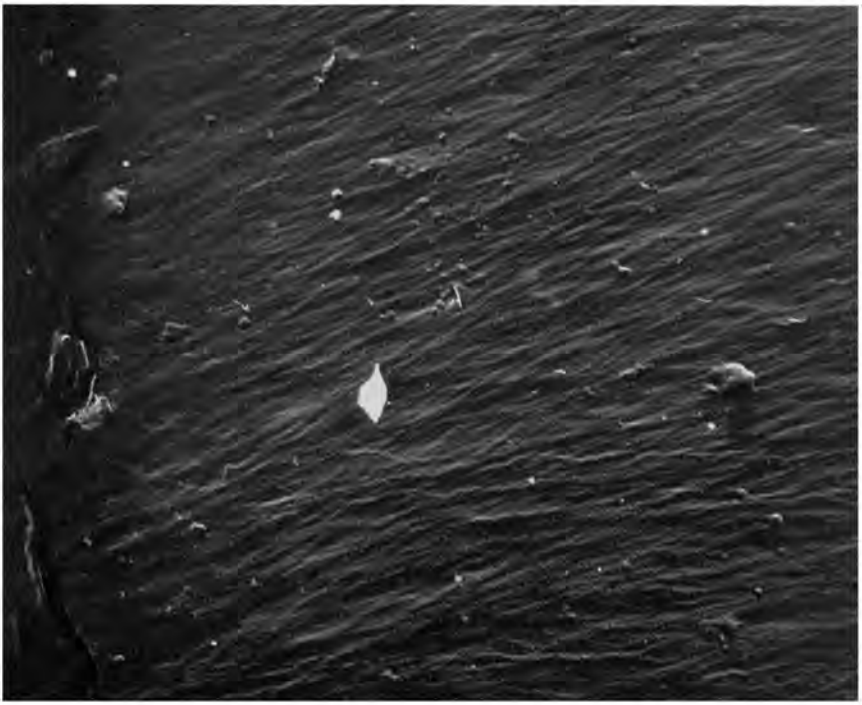


Fig. D (*ii*) — Enlargement of the inset area of Fig. D (*i*)
(electron micrograph × 9 000)