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Prepared contributions

Dr P. J. Houen, Technical University of Norway, Trondheim

IN A continuing investigation on the tensile rupture of paper being performed in the Pulp and Paper Technology Department of the Technical University of Norway, some effects of deliberate fibre weakening have been studied. The results seem to support the new theory for the load/elongation properties of paper represented by Kallmes & Perez.

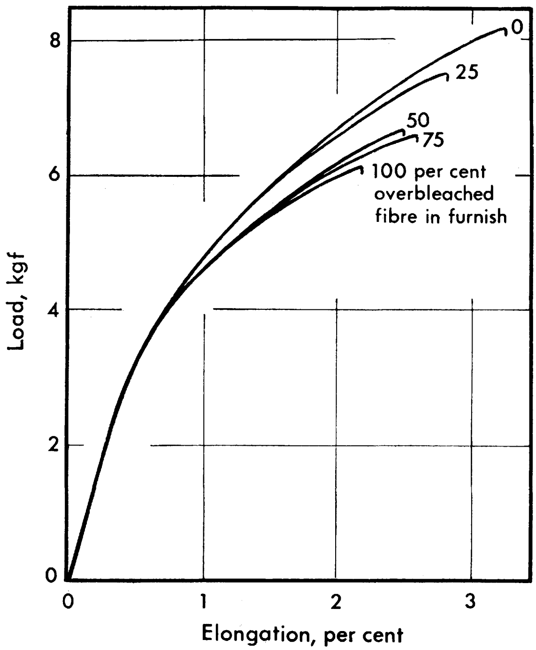

Fig. 1—Experimental load/elongation curves of paper made from blends of the +35 mesh fraction of commercial, bleached southern pine kraft pulp and the corresponding pulp after having been subjected to an oxidative treatment with sodium hypochlorite: pulp beaten 8 000 rev PFI before fractionation and chemical treatment

A commercial, bleached southern pine kraft pulp was beaten to different levels in a PFI mill and the beaten pulps then fractioned over a 35 mesh screen. At each beating level, the +35 fraction was divided into two portions, one of which was overbleached by a rather drastic, oxidative treatment with sodium hypochlorite. At each level of beating, handsheets were made from blends of the normal and the overbleached pulp; then 0.75 per cent guar gum was added before sheetmaking to overcome the strong tendency of the over-bleached pulp to flocculate.

Papers made from 0 and 100 per cent overbleached pulp, respectively, did not show any significant difference in tensile strength for beating times less than approximately 3 000 rev. For pulps beaten 4 000 rev and more, however,

TABLE 1

Strength and other properties of paper made from blends of the +35 mesh fraction of a commercial, bleached southern pine kraft pulp and the corresponding pulp after having been subjected to an oxidative treatment with sodium hypochlorite
Pulp beaten 8 000 rev PFI before fractionation and chemical treatment

Property	Composition of paper—overbleached pulp, per cent				
	0	25	50	75	100
Breaking length, km	7.7	7.3	7.0	6.6	6.4
Tear factor	139	119	103	83	65
Burst factor	61.0	52.5	43.5	41.0	38.0
Apparent density, g/cm^3	.701	.687	.687	.691	.709
Scattering coefficient	190	199	194	199	190
Rupture elongation, per cent	3.11	2.93	2.53	2.42	2.27

load/elongation curves revealed that rupture load, rupture elongation and post-yield slope decrease with increasing amounts of overbleached fibre in the furnish. Primary slope was unaffected, suggesting that the same amount of material is initially engaged in all cases and that the elastic properties of the fibre material do not change during the drastic sodium hypochlorite treatment. Fig. 1 shows a set of experimental curves for paper made from pulp beaten 8 000 rev PFI.

A tentative explanation of these results can be based upon the concept that fibre breakage is the determining factor in tensile rupture of paper sheets having a degree of bonding above a certain level—in this case, that developed by beating for 3 000 rev. The tacit assumption here that overbleaching mainly acts by decreasing the strength of the fibres, leaving their bonding capacity and sheet structure more or less unchanged is supported by the constant primary slope of the load/elongation curves and the approximately constant values for apparent density and scattering coefficient (Table 1).

The interesting observation of decreased post-yield slope can hardly be accounted for from the concept of weaker fibres alone, provided fibre rupture does not occur in prerupture flow. To explain this, overbleaching must also be assumed to affect the bonding capacity of the fibres and/or their flow characteristics. Because the introduction of overbleached fibres seems not to be most important.

Dr D. L. Taylor, Monsanto Co., St Louis, U.S.A.

We have found that the sonic modulus of paper undergoes characteristic changes during and after straining. Relative to the initial value, the sonic modulus of paper under load is increased. After release of the load, we find the sonic modulus to have decreased to below its original value for the unstrained paper.

The modulus loss after release of the stress begins to appear only at stress levels beyond the linear elastic region for the sample. Apparently, the modulus loss is a reflection of structural damage in the sheet, probably breakage of interfibre bonds.

We have measured the modulus loss caused by straining as a function of angle θ to the stressed direction. The results presented in Table 1 (below) should be of interest to paper physics theorists. (The stress level was 80 per cent of the ultimate tensile strength.)

TABLE 1

Paper type	*Stress direction*	*Sonic modulus,* 10^{10}*dyn/cm*^{2}	*Modulus loss at angle θ to stress, per cent*				
			0°	22°	45°	67°	90°
1. Bleached kraft soft-wood, laboratory sheet, 500 CSF	—	8.44	26	11.3	5.8	10.5	24.7
2a. Unbleached kraft wrapping, Fourdrinier sheet	MD	14.9	2.4	—	3.3	—	8.5
2b. Ditto	CD	9.67	9.3	—	3.7	—	1.4

Transcription of Discussion

Discussion

Dr J. Kubát—The theory of stress/strain behaviour proposed by Kallmes is based on the concept of bond breaking as a flow mechanism. An experimental fact that should be mentioned in this connection is the variation of the modulus of elasticity with strain. It is always found that, on straining, the modulus either increases or remains unchanged. This phenomenon, known as strain hardening, occurs in paper as well as in other materials. Under exceptional conditions—as on repeated loading of paper with high strain amplitudes—a softening may take place, that is, a breakdown of the sheet structure. This effect is of no importance, however, when ordinary stress/strain curves are discussed. The increase in modulus with strain can be demonstrated both in creep and stress/strain processes on which a sinusoidal deformation of comparatively small amplitude is superimposed.* Such a procedure permits an evaluation of the elastic response of the sheet during the various phases of the flow process to be made. Needless to say, the slope of the plastic region of stress/strain curves does not represent any measure of the elastic response of the sheet.

The theory presented here is based on the assumption that the number of fibres engaged in the loading process of the stress/strain curve decreases. Such a theory of plastic flow could formally be correct, if any one of the following additional assumptions were to be taken into account—for instance, compensation for the effect of bond breaking by strain hardening; the formation of new bonds; even redistribution of the stress in the sheet. Such assumptions are not contained in the theory being discussed. The question is how can the decreasing number of fibres engaged in the straining process be brought into agreement with the constancy of the modulus or, what is more likely to occur, its increase with strain. The second question is more general and relates simply to the fact that no attention has been paid to the accepted theories of solid state flow.

* Kubát, J., Nyborg, L. and Steenberg, B., *Svensk Papperstidn.*, 1963, **66** (19), 754–764
Kubát, J. and Lindbergson, B., *Svensk Papperstidn.*, 1965, **68** (21), 743–756

Dr O. J. Kallmes—In order to answer these questions, we have to describe the theory in more detail than we did in the paper. To begin with, Fig. 5 illustrates the way in which the stress/strain curve ought to be constructed according to the basic concepts of the theory; in the paper, we used the approximate method indicated on page 785.

The first step in constructing the curve is the calculation of the elastic modulus of the unstrained structure by equation (*21*); note that bond failure *per se* —in the form of the term $(1 - f_e)$ of equation (*9*)—does not enter into the picture. After a small finite elongation, the first modulus curve is terminated.

As a result of the first elongation (no matter how small, even infinitesimal), a few fibres are eliminated as load-bearing elements because of bond failure. Thus, we have a new structure, one that is slightly different from the original unstrained one. The load developed per unit of elongation e, as $e \rightarrow 0$, of this new structure is lower than what would have been developed by the original one at the end of the first elongation if no bond failure had occurred; for the new structure, f_c of equation (*21*) is increased slightly. Thus, the elastic modulus curve of the second structure is displaced downwards and to the left (in Fig. 5) of the first curve. The two modulus curves are connected by a vertical line from the termination point of the first curve to the second; the latter intersection is the beginning of the second curve. The vertical line represents the stress drop caused by the bond failures that occurred during the first elongation.

The second modulus curve is terminated after an elongation equal to the first. The modulus of the third structure developed as a result of bond failure in the second elongation is then calculated. The second and third modulus curves are connected in the same way as the first pair. By proceeding in the same manner, the sawtooth stress/strain curve depicted in Fig. 5 is constructed. The entire load/elongation curve is terminated when the conditions representing sheet failure, equations (*18*) and (*19*), are met.

If it is assumed that the modulus of the fibres remains constant during straining, the slope of every modulus curve is lower than that of the preceding one, because of the reduction in the number of load-bearing fibres (that is, an increase in f_c). We know, however, from the work of Taylor & Craver presented at this symposium that the modulus of the fibres increases during the straining of the sheet, owing to a strain-hardening effect. Therefore, the small loss in sheet modulus from the removal of fibres as load-bearing elements is probably compensated for by an increase in the modulus of the fibres still taking load. Hence, the elastic modulus of the sheet remains constant during straining.

The above concept is supported further by another observation of Taylor & Craver. They found that the modulus of a sheet after straining is lower than that before straining and attribute this loss to bond failure. We concur with this concept in the sense that we believe that the number of fibres participating in the modulus measurement of the relaxed sheet is fewer than in the original because of bond failure. It should be noted, however, that the drop in sonic modulus is not only a measure of the number of fibres removed as load-bearing elements, but also represents the small irreversible change in the fibres' modulus caused by straining and relaxing (see pp. 518–519 of our first contribution).

In the case of thin 2-D sheets, the picture is slightly different than for an ordinary sheet. The fraction of fibres acting as load-bearing elements is reduced markedly by the failure of every crossing. Thus, the increase modulus of the strain-hardened fibres still developing a load after a given strain cannot compensate for the loss in the number of fibres participating. As yet, we have not attempted to include creep phenomena in the theory. All we have done is to calculate the modulus of structures resulting from given strains, though there is no reason that you could not substitute time for elongation and develop a picture for creep.

Mr D. H. Page—My point is complementary to that made by Kubát. Tydeman and I did an experiment some time ago when we had ideas more or less along the lines expressed by Kallmes. As a result of our thoughts following this experiment, we abandoned those ideas.

We made some idealised models of paper using strips of a clear plastic (cellulose acetate I believe). The strips, several inches long, were laid down at random positions, but with two preferred orientations for convenience. The strips were glued together at points where they crossed. We then put this structure under tensile strain. The stress/strain curve had an initial elastic linear region. Then, as the bonds began to break, just as Kallmes has indicated, the stress/strain curve turned over exactly as the stress/strain curve of paper. When we had broken about half the bonds, we lowered the stress and found that the curve returned more or less linearly *to the origin*. There is no permanent set in such a structure—and there cannot be, because all our elements are elastic; while there is still one elastic path remaining between the jaws of the tester, there will be a restoring force returning the structure to zero strain.

Now, the stress/strain behaviour of paper is not like this. Permanent set is an intrinsic part of it. In an attempt to create a structure that would exhibit set, we remade the model putting in a number of kinked segments. We hoped that during straining bonds would break and kinks would be pulled out, but again this did not happen. The result was very similar to that obtained previously. The same argument holds. To achieve permanent set, every original elastic path must be broken and this would require a degree of bond breakage that would imply complete disruption of the sheet.

Discussion

The experiments here are not proofs, of course, but rather guides to thought. They lead to the important principle that it is impossible to produce a theory that incorporates permanent set simply on the assumption of bond breakage in a random structure of elastic fibres, even if some of these are kinked.

These thoughts led us to the view that the stress/strain curve was not so much a property of the gross sheet structure, but was more the property of the fibres in the regions of the fibre-to-fibre crossings.

I query the suggestion that the stress/strain curve of microcompressed fibres is concave towards the stress axis. Under the usual testing procedure, the rather gross curls and kinks that are usually present on microcompressed fibres may produce this effect, but the microcompressed regions themselves may not display such a curve. I have been able to produce sheets of paper in which every fibre is microcompressed down its entire length and they exhibit quite normal stress/strain curves.

Dr Kallmes—This is a model of plastic that contains many features not in paper. When a bond fails in this thin, rigid model, there is no opportunity for the type of stress redistribution inherent in our theory. For example, crossing failures do not eliminate fibres as load-bearing elements. As you well know, a microcompressed part of a fibre often does not develop a load after bond failure.

As far as the stress/strain curve of microcompressed fibres is concerned, I do not know its exact shape any better than you do; we have never measured

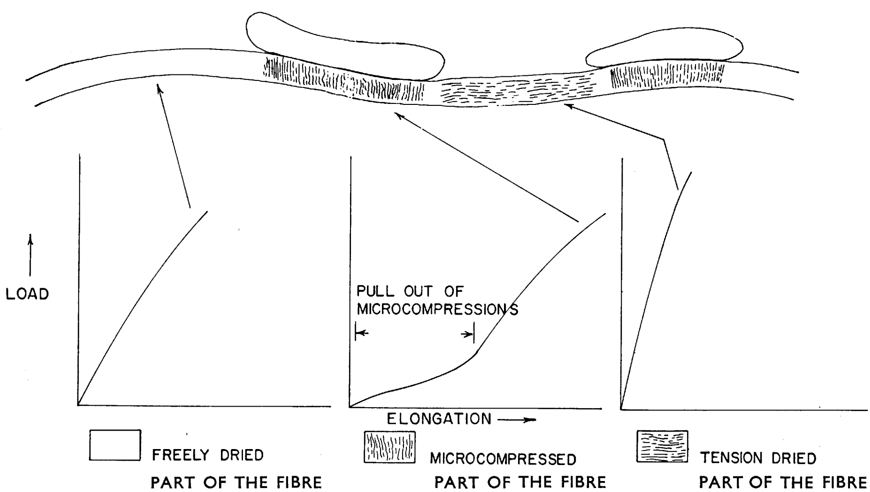

Fig. N—Load/elongation properties of parts of fibres under different conditions of restraint within a sheet it successfully. The picture I presented yesterday (illustrated in Fig. N) is purely speculative.

Mr P. A. Tydeman—It is of further interest to note that, in this plastic model, a fibre broke very early on in the stress/strain curve by shear, yet this still did not give the structure any significant permanent set.

Some points about the theory and its derivation and, firstly, on the basic assumptions. It is assumed that the strain in fibre segments can be based on the average or sheet strain, hence the paper ruptures when its extension is the same as the extensibility of a fibre. This must be seen in the light of the variability of heterogeneity of paper on a macroscopic scale. We have observed appreciable variation from point to point of the strain of paper under tension. This variability should be taken into account when calculating the value of the average strain when rupture will occur.

On another assumption, that the transverse modulus of fibres is negligible, I should like to see this more firmly established. I discussed one of your pieces of evidence after presentation of the paper by yourself and Perez on fibre properties; the other evidence is based on sonic moduli experiments of the nature described by Taylor & Craver. How conclusive are these experiments?

Fig. 7 is claimed to show the constancy of the modulus ratio (vertical axis) at a value of 0.34 for all values of RBA and the curve drawn is supposed to be a better fit, but I suggest that this graph indicates a high dependence of the ratio on RBA and that a line through zero at roughly 45° would not be a bad fit.

I want to make the plea that it is essential when comparing theoretically derived results with experimental data that some indication is given of what magnitude of the discrepancy between experiment and theory would cause one to re-examine and reformulate the theory.

Dr Kallmes—We fully realise that the stresses and strains within a sheet are not uniform. We have developed a technique for measuring the variations in strain along fibres within a sheet and found them to be quite large. For example, in one sheet strained 3 per cent, the strain along a fibre oriented in the direction of elongation varied 1.26–4.06 per cent. It would be possible to write equations for these strain variations and calculate stresses on the basis of them. The resulting equations for the modulus of a sheet at any given elongation would be extremely complex and would be of no value to the development of an understanding of the mechanisms that occur when a sheet is strained.

You must realise that our goal in this work is *not* the development of exact equations with perfect predictive powers. Rather, we are trying to develop a relatively simple picture of the stress/strain behaviour of paper and incorporate in it all the important phenomena in an approximate way. Our variables all represent easily pictured fibre properties and structural elements. These are assembled in a model representing a point on the stress/strain curve. The manner in which you go from one point on the curve to the next is specified somewhat arbitrarily. This is the only way we know of to describe a discontinuous mechanism such as the straining of paper to failure.

Measuring the modulus of fibres in its two principal directions (parallel and at rightangles to the fibre axis) by the sonic technique applied to highly oriented handsheets gives a relative rather than an absolute measure. We found a 10:1 ratio, which, incidentally, is about the same as the ratio of the amount of swelling a fibre undergoes in these two directions upon wetting. I believe you found that a fibre swells 20–25 per cent at rightangles to the fibre axis,* although it is well known that it swells only 1–2 per cent axially. These swelling values are an indication of the strength of a fibre in these two directions.

I have already commented on the precision of the data. We are quite pleased to have a model based on the known mechanisms of straining and the measurable fibre and structural properties giving predicted values within 100 per cent of the measured ones.

Mr P. E. Wrist—Kallmes' theory is based on the assumption of uniform strain in the sheet. Van den Akker used this assumption in his 1961 Oxford symposium paper and concluded that fibres lying approximately at rightangles to the strain direction were under compressive strain. In a well-formed sheet, the frequency of fibre bonding along a fibre is such that this compressible stress cannot be readily relieved by fibre buckling. Bond failure under these compressive strains is therefore probable. It is not clear whether Kallmes has taken such failure into account in his paper or only considered failure from tensional stresses.

My second comment is addressed to Page, who has cited his model experiment with bonded plastic strips to show that bond breakage between elastic fibres is not able to explain permanent set in a network. In a dry paper web, the fibres themselves are made of elastic fibrils, bonded together. Each fibre has dried-in stresses and, on straining a web, these internal bonds can break in addition to intrafibre bond failure. Relaxing such a structure will lead to a permanent change in dimension, since removing the external stress will not recreate the internal fibre stresses of the original sheet. I do not agree, therefore, that we must assume visco-elastic behaviour on the part of the fibres to

* *Nature*, 1963, **199** (4 892), 471–472

explain permanent set in a strain/relaxation cycle; internal bond breakage within the fibre is a sufficient mechanism.

Dr Kallmes—We neglected fibres in compression, because we believe they buckle and so contribute negligibly to the load developed by the sheet. Therefore, we integrated equation (*12*) from 30° to 90° instead of from 0° to 90°. I agree that buckling can cause bond failure, but we have not included the mechanism in the theory as yet—of course, it can be done.

Failures within fibres would be incorporated in the theory by changes in the fibre properties as a function of time or elongation. For example, instead of the modulus E, one could use the modulus $(E_0+E_1\,e_x+E_2\,e_x{}^2+\ldots)$ in equation (*5*). We did not do this in the first version of the theory, because such complications add little to the basic concepts we tried to emphasise in the paper.

Prof. B. Steenberg—The purpose of any of the many models we have is a practical one: to condense our knowledge into a simple picture comprehensible to anyone and of use both qualitatively (for example, atomic models in wood) and quantitatively for calculations. Consequently, a new model should explain more details than do earlier models, so that an acid test is not only that it explains what is already known, but predicts something new.

The mechanical model with dashpots and springs is a useful way of depicting a complicated series of equations for the behaviour of many materials under stress and strain, taking time-dependent properties into consideration. A system made up of electrical elements can be used to depict the same equations, but we are not used to electrical analog components today in such a way that an electrical model is useful.

The important point is that a series of empirical equations depicted to the layman by, for instance, a model of springs and dashpots describes remarkably accurately the stress/strain properties, including loops, creep and relaxation curves for paper. It was four parameters, two moduli of elasticity and two parameters that describe the viscous properties of an Eyring fluid.

If the Kallmes *et al.* theory is converted to a mechanical model, it consists of a series of identical springs coupled in parallel. These springs break one after another during straining: to describe this, he has to use the complete stress/strain curve. I cannot see that it has fewer parameters than the old model, nor that it can depict more than the 'plain' stress/strain curve—obviously, not the permanent set, creep or relaxation. Is there an unknown feature of paper that can be predicted by it? If so, the model is useful with all its shortcomings.

Mr Tydeman—There is a contradiction in the theory. As the bond is broken, there are fewer fibre elements under strain, therefore the modulus is lower: it does not remain the same.

Dr Kallmes—I agree that the modulus of a sheet does not necessarily remain constant while a sheet is being strained; Taylor & Craver (at this symposium) have shown that it rises. The modulus depends on the number of fibres developing load $(1-f_c)$ and on their modulus E. This matter is discussed more fully in my first reply (page 805).

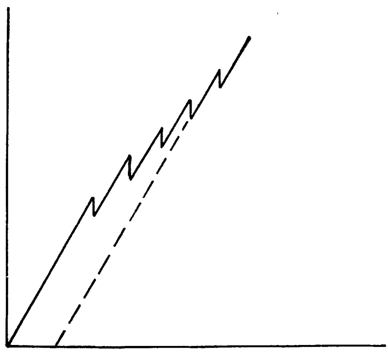

Fig. O—Mechanism by which theory accounts for permanent set

The elastic modulus and the entire slope of the stress/strain curve of a sheet at a given elongation are different items. Referring to Fig. 5, the former is represented by the solid slope ν and the latter by the dotted curve drawn through the sawtooth curve. I have already referred to the factors on which the modulus depends (page 805). The slope of the stress/strain curve as a whole at an elongation e is a function of the elastic modulus of the sheet at e and the size of the stress drops caused by bond failure (elongation 9). The smooth stress/strain curve with which we are all so familiar is, in fact, the dotted curve in Fig. 5. It comes about because stress/strain recorders do not have the sensitivity to construct the true sawtooth stress/strain curve of paper.

For a sheet whose elastic modulus remains constant during straining, the load/relaxation curves do not go through the origin (see Fig. O).

Dr J. A. Van den Akker—It seems to me that Kubát's remark is very cogent: the visco-elastic properties of the fibres must be brought into the model. This (it seems to me) is extremely important for the theoretical explanation of the known tensile stress/strain characteristics of paper.

Dr Kallmes—As is shown on page 810, the visco-elastic properties of fibres can be taken into account quite readily.

Dr Van den Akker—This was indeed brought out in the discussions at the 1961 symposium, which were the basis for the work that we and others are doing today on the visco-elastic properties of individual fibres.

Dr A. H. Nissan—I would like to point out one thing. There are explicit and implicit ways of introducing arbitrary parameters. When one makes something known to vary strongly into a constant, a whole battery of arbitrary parameters have been introduced, because of the contra-acting parameters brought in to eliminate what is known to be variable and to make it constant. Therefore, the statement that your theory is nothing but observables is not strictly correct: you are introducing unobservables by omission. This, in itself, is not uncommon in theoretical work nor is it bad—indeed, it is essential for progress—but we ought to be aware of it.

Dr Kallmes—We made numerous simplifications of the theory in the paper, because we did not want to complicate the basic concepts with many mathematical embellishments. The basic concepts would get lost in a swamp of equations. Since this paper was submitted, we have incorporated many refinements and the only way that we can integrate the final equation is numerically on a computer. Although these improvements in the theory have produced more accurate stress/strain curves, they have added relatively little to our storehouse of basic knowledge.

Dr H. K. Corte—I would like to reply to Nissan's last remark and support Kallmes. It is one thing to simplify what in reality is fibres bonded together and so avoid complicated statistical-geometrical calculations. It is quite another thing to analyse a completely artificial model, even though it might have fewer constants, which behaves as if it were similar to a piece of paper, but with no element that can be traced or identified in a sheet of paper, because of which it was eventually abandoned. Although simplifications were made in this work, Kallmes is speaking of fibres and, of course, fibres are elastic, they are kinked, they are microcompressed and plastic. To take all these features into account in a random or non-random network is extremely complicated arithmetically, but not conceptually.

Mr P. E. Wrist (*in a written contribution*)—Support for Giertz' proposition that the hemicellulose present in the fibre may be soluble and therefore sufficiently mobile to provide the bonding adhesive both within and between fibres is supported by some recent studies of McIntosh of our laboratories on high-yield holocellulose fibres prepared by the peracetic acid method from loblolly pine. Fig. P shows several fibres that have been beaten in a Waring blender for 10 min to a freeness of 310 CSF, then dried down to on a glass slide. The sample was metal shadowcast for improved contrast. The photograph shows

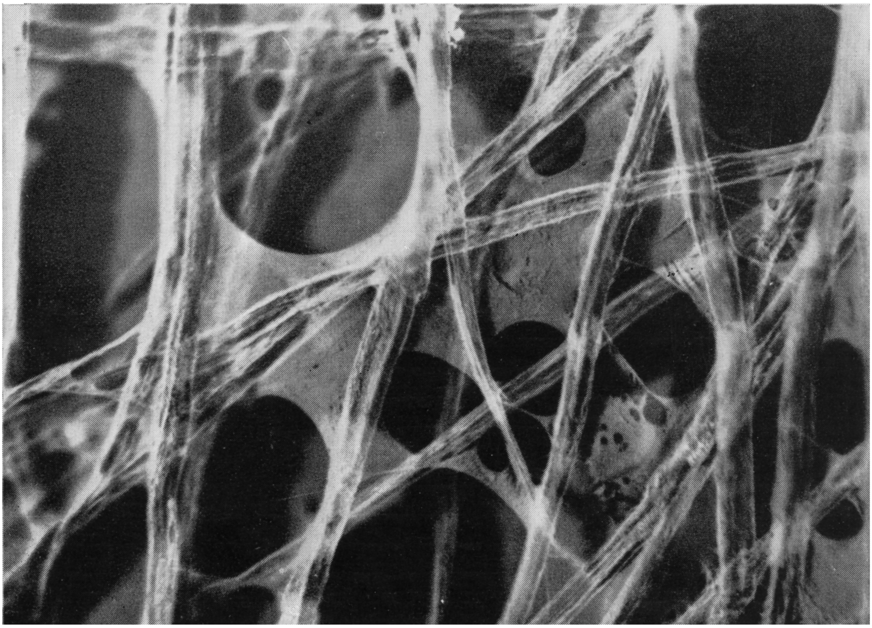

Fig. P—Loblolly pine summerwood holocellulose fibres (peracetic acid method), refined for 10 min in Waring blender with distilled water to 310 CSF [× 147]

the presence of an unoriented film of hemicellulose, which formed as the water was evaporated. The film is seen to span quite large voids between the fibres and to be drawn also around points of fibre interaction. Although much of this hemicellulose would be removed in a conventional cook, some hemicellulose remains and it is not unreasonable to expect it to behave as Giertz proposes.