THE EFFECT OF PULPING UPON THE DIMENSIONS
OF WOOD TRACHEIDS

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ABSTRACT

The dimensions of the fibres in oven-dry blocks of wood from three species (white spruce, Douglas-fir, and larch) were deduced from measurements of bulk density and the number of fibres per unit cross-sectional area. The blocks were then cooked by the soda process to various yields, and the fibre dimensions were redetermined after the blocks had been washed and oven-dried.

The weight of the fibres per unit length decreased almost in proportion to the yield loss, being reduced at 40% yield to 42% of its value in wood. This finding indicates that the fibres were shortened to only a small extent by pulping (ca. 4% at 40% yield) and that the major changes were in the transverse dimensions of the fibres. At 40% yield, cell-wall thickness and fibre width were respectively reduced to 52.5% and 84% of their original values. The results were independent of wood species.

The changes in the dimensions of the fibres are in keeping with current concepts of the structure of the cell wall.

Additional keywords: Picea glauca, Pseudotsuga menziesii, Larix decidua, fiber dimensions, cell-wall thickness, fiber diameter, fiber length, soda pulping, yield, bulk density, wood, cross section.

INTRODUCTION

The physical properties of paper depend to a large extent upon the dimensions of the constituent fibres. However, the dimensions of fibres, particularly cell-wall thicknesses, are ill-defined in the literature largely because of the application of methods of measurement that have become suspect. In a recent paper [Scallan and Green 1974], we proposed a method for determining the transverse dimensions of the fibres in small blocks of wood that avoided sources of error associated with many previous determinations. In the present paper we report the use of the same technique to investigate the changes in the dimensions of wood fibres that occur upon pulping. Little attention has been paid to this subject in the past, although the information is necessary in order to reach a complete understanding of the relationship between wood and paper properties. In the present work, the dimensions studied were those of fibres in the oven-dry state, this state being of fundamental interest. First, the dimensions of fibres conditioned to any relative humidity may be calculated from those in the dry state using knowledge of moisture uptake and the mode of swelling. Second, it is possible that, as will be shown, the changes in the dry dimensions that result from pulping are related to the structure of the cell wall, and by studying the changes we may learn a little more of the structure.

The cell walls of both wood and pulp fibres are nonporous in the dry state [Stone et al. 1966] and therefore, with the removal of up to 60% of the material of the wall, as occurs in pulping, it is to be expected that there will be a contraction in one or more of the primary dimensions of the fibres. Since the cellulose component is in the form of extremely long microfibrils, the bulk of which run the length of the wood fibre, it can be conjectured that the dissolution of the lignin and hemicelluloses, which are distributed transversely about the microfibrils, will leave the fibre essentially unaltered in length. The weight of the fibre per unit length, the fibre coarseness, would, however, be reduced in proportion to the weight of material dissolved. If indeed the fibre length does not change, then the changes brought about by pulping must

be in the transverse dimensions of the fibre, i.e., wall thickness and/or fibre width. Of these two, wall thickness appears likely to show the greater change since it is known that the lignin-hemicellulose-rich middle lamella surrounds the fibre as a tangential sheath, and it has been suggested that within the cell wall, the lignin-hemicellulose is distributed in aggregates that are more extensive in the tangential direction of the wall than in the radial direction [Scallan 1974; Kerr and Goring 1975]. Removal of the lignin and hemicellulose during pulping and subsequent closing up of the wall on drying should therefore be reflected in a considerable reduction in cell-wall thickness with little change in fibre width.

The literature contains only fragmentary evidence that may be used to confirm or deny the dimensional changes hypothesized above. While some authors have used suspect techniques, others have confined their measurements to the dimensions of fibres in the wet state. Little use can be made of these data in the present study since it is now known that the water content of water-saturated cell walls is greatly increased by pulping [Stone and Scallan 1967]. This increase is so great that any decrease in the dimensions of fibres as a result of pulping that might be observed if the fibres were examined in the dry state is much reduced or even negated when the fibres are observed in the water-swollen state [Stone and Scallan 1967; Stockmann 1971].

On the subject of the effect of pulping on fibre length, most authors appear to have taken it for granted that the length is little altered. Few measurements are made on wood fibres as they exist in wood; rather the length is taken to be that as measured on single fibres after maceration. However, maceration, a chemical treatment of the wood causing fiberization, is itself an extensive pulping treatment. Nevertheless, Ladell [1959], using a new method to measure fibre length on tangential sections of wood, reported that the lengths determined on wood were "comparable" with those obtained after maceration. Britt [1965] compared the weights per unit length of the fibres in several samples of wood with those measured on samples of commercial pulps and observed that there appeared to be a reduction in proportion to yield. This finding also indicates a negligible change in length as a result of pulping.

Concerning the effect of component removal upon the transverse dimensions of wood fibres, Stone et al. [1971] studied black spruce tracheids at various stages of delignification. Here it was found that the cell wall decreased in thickness proportionally to the amount of lignin removed, whereas the lumen width stayed constant. Using identical techniques, Kerr observed the same changes when hemicelluloses were removed from birchwood [1974]. This mode of behaviour was attributed to the components in the cell wall being concentrated in tangential layers concentric with the fibre axis. No similar study has been carried out where both hemicellulose and lignin are removed simultaneously as in pulping. Possibly the only consistent trend that may be observed in the literature is that, where care is given in the method of measurement to avoid the sources of error associated with direct measurement, the cell-wall thicknesses of dry pulp fibres appear to be of the order of half those measured on wood fibres. For example, the cell-wall thicknesses of spruce, Southern pine, and jack pine pulp fibres as measured by Kallmes and Bernier [1963] may be compared with those measured on samples of wood of the same species by Scallan and Green [1974].

<center>EXPERIMENTAL</center>

Wood samples

Three species of wood were used in this study—white spruce (*Picea glauca*), Douglas-fir (*Pseudotsuga menziesii*), and European larch (*Larix decidua*). From cross-sectional discs of each, the earlywood was cut from a number of consecutive rings within the mature wood. The bands of earlywood were then subdivided in the radial direction of the wood into small blocks that measured about 25 mm² in cross-sectional area by 10 mm in the longitudinal

direction of the wood. The ends of the blocks were surfaced by razor blade.

Pulping

For each cook of a particular wood species to a given yield, two or three blocks upon which measurements had been made were used along with a number of similar blocks for the determination of yield. To ensure uniform penetration of the cooking liquor, the blocks were first water-logged, drained, and then soaked overnight in the cooking liquor (molar sodium hydroxide solution).

Soda cooks were carried out with a large excess of cooking liquor in small stainless steel bombs immersed in a heated oil bath. Each cook was commenced at 80 C and the temperature was raised at the rate of 1 C/min to a maximum of 170 C. Different pulp yields were achieved by varying cooking time.

Cooked blocks were washed free of spent liquor by soaking in distilled water with repeated changes over a period of 2 to 3 days. To prevent cracking of the blocks upon drying, they were allowed to air-dry overnight before oven-drying at 105 C. Pulp yield was determined without fiberizing the blocks.

Determination of fibre dimensions

The technique used for the deduction of the transverse dimensions of the fibres in small blocks of wood has already been described in detail [Scallan and Green 1974]. The actual measurements made on the oven-dry blocks before and after cooking were:

(1) The number of cells per unit area in the cross-sectional face of the block (N).

(2) The fraction of the area of the cross-sectional face of the block that is occupied by nonfibrous elements i.e., ray cells, resin ducts etc., (F)

(3) The bulk density of the block as found by mercury displacement (D_B).

From these measurements, it has been shown that, assuming a square cross-section for the average fibre, the fibre width, "b,"

the lumen width, "a," and the fibre coarseness, "C," may be derived from the equations:

$$b^2 = \frac{1}{N} \qquad (1)$$

$$a = b\left[1 - \frac{D_B}{D_C(1-F)}\right]^{1/2} \qquad (2)$$

$$C = \frac{D_B}{N(1-F)} \qquad (3)$$

where D_c is the density of the cell-wall material. In the present work, D_c was assumed to have a fixed value (1.5 g/cm^3) independent of wood species and pulp yield. This is an approximation but one which must be made in the absence of exact data. Since lignin and hemicellulose have lower densities than cellulose, it is probable that the density of the cell wall will rise from somewhat below 1.50 to over 1.55 g/cm^3 with pulping. The assumption of a constant value of 1.50 will thus lead to an error in the estimation of cell-wall thickness, but it can be calculated that this should be an underestimation of less than 5% at the lowest yields studied. Fibre width and fibre coarseness will not be affected by the assumption.

The pulping procedure had little effect on the suitability of the block surfaces for the microscopic measurements. The fraction of nonfibrous elements (F) in the cross-sectional face of the blocks had changed slightly in some cases. Occasionally F had decreased because of the closing of ray-cells and in other cases had increased because of the development of fissures between cells. Such fissures were, however, remarkably few and when they did occur were quite small. As an example of the results obtained, the data for the Douglas-fir samples are given in Table 1. However, for analysis of the data, the percentages to which the various measurements on each block were reduced by cooking were considered.

TABLE 1. *Data obtained on the pulping of Douglas-fir showing the properties of each block before and after cooking*

| Block No.	Yield, %	Bulk density, g/cm^3	F factor	Fibres per mm^2	Fibre coarseness, $mg/100m$	Fibre width μm	Cell Wall thickness. μm
4	100	0.4208	0.0667	849	53.11	34.32	2.81
4	91	0.4234	0.0758	914	50.12	33.08	2.75
5	100	0.3181	0.0560	840	40.12	34.50	2.06
5	91	0.3025	0.0533	804	39.74	35.27	1.99
6	100	0.2731	0.0526	773	37.29	35.97	1.82
6	91	0.2599	0.0580	823	33.52	34.86	1.68
7	100	0.3000	0.0591	812	39.27	35.09	1.97
7	79	0.2725	0.0667	960	30.41	32.27	1.65
8	100	0.3097	0.0540	861	38.02	34.08	1.97
8	79	0.2759	0.0602	879	33.40	33.73	1.74
9	100	0.2686	0.0580	739	38.58	36.79	1.84
9	79	0.2303	0.0573	876	27.89	38.79	1.43
10	100	0.3237	0.0600	833	41.34	34.65	2.12
10	75	0.2826	0.0779	1033	29.67	31.11	1.68
11	100	0.3285	0.0555	980	35.49	31.94	1.97
11	75	0.2832	0.0683	1057	28.76	30.76	1.64
12	100	0.3165	0.0544	837	39.99	34.57	2.05
12	75	0.2675	0.0606	986	28.88	31.85	1.59
13	100	0.2808	0.0649	783	38.35	35.74	1.89
13	73	0.2454	0.0700	864	30.54	34.02	1.57
14	100	0.3119	0.0595	799	41.51	35.38	2.08
14	73	0.2749	0.0630	876	33.49	33.79	1.74
15	100	0.2741	0.0511	675	42.79	38.49	1.95
15	73	0.2334	0.0546	794	31.09	35.49	1.52
16	100	0.3011	0.0706	863	37.54	34.04	1.95
16	66	0.2452	0.0753	975	27.19	32.03	1.48
17	100	0.2881	0.0696	761	40.69	36.25	1.98
17	66	0.2422	0.0618	929	27.79	32.81	1.48
18	100	0.3018	0.0640	806	40.00	35.22	2.01
18	66	0.2591	0.0625	1017	27.18	31.27	1.32
19	100	0.2827	0.0493	738	40.29	36.81	1.92
19	61	0.2271	0.0644	1023	23.73	31.36	1.52
20	100	0.2697	0.0603	730	39.32	37.01	1.86
20	61	0.2142	0.0581	964	23.59	32.21	1.27
21	100	0.2908	0.0483	778	39.27	35.85	1.93
21	61	0.2364	0.0548	1025	24.40	31.23	1.36
22	100	0.2654	0.0615	813	34.78	35.07	1.74
22	54	0.2005	0.1176	1165	19.50	29.30	1.15
23	100	0.3716	0.0540	922	42.60	32.93	2.32
23	54	0.3092	0.0993	1441	23.82	26.34	1.60
24	100	0.2696	0.0539	861	33.10	34.08	1.70
24	54	0.1959	0.1069	1177	18.64	29.15	1.11
25	100	0.2830	0.0613	763	39.51	36.20	1.92
25	46	0.2025	0.1155	1194	19.17	28.94	1.15
26	100	0.2926	0.0600	792	39.30	35.53	1.95
26	46	0.1872	0.1213	1229	17.33	28.52	1.05
27	100	0.3075	0.0634	826	39.75	34.79	2.02
27	46	0.2013	0.1263	1181	19.51	29.10	1.16
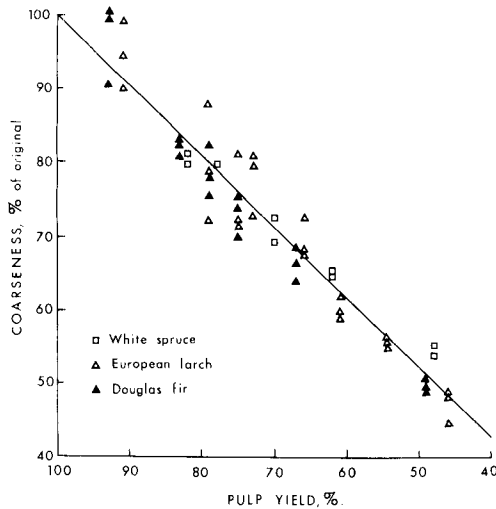
FIG. 1. The effect of pulping upon the coarseness (i.e. weight per unit length) of wood fibres as measured oven-dry before and after pulping.

FIG. 2. The effect of pulping upon the length of wood blocks as measured oven-dry before and after pulping.

Fibre length was not measured directly: rather, the effect of pulping on fibre length was deduced from the change in fibre coarseness and also from the change in block length as a result of cooking.

DISCUSSION

Change of fibre coarseness with pulp yield

Figure 1 show how the calculated weight of the wood fibres per unit length (i.e. coarseness) changes with yield. The trend is, to a good approximation, independent of softwood species and, as expected, indicates a fractional reduction almost equal to the fraction of material removed.

Closer examination does, however, reveal that a least-squares fitted line lies somewhat above the 45° line and passes through a fractional coarseness of 0.52 at 50% yield. The simplest explanation for this slight deviation is that there has been a reduction in fibre length of the order of 4%. Having suspected this in preliminary cooks, the reduction in block length as a result of cooking was followed in subsequent cooks; it being supposed that any reduction in fibre length would be reflected in block length.
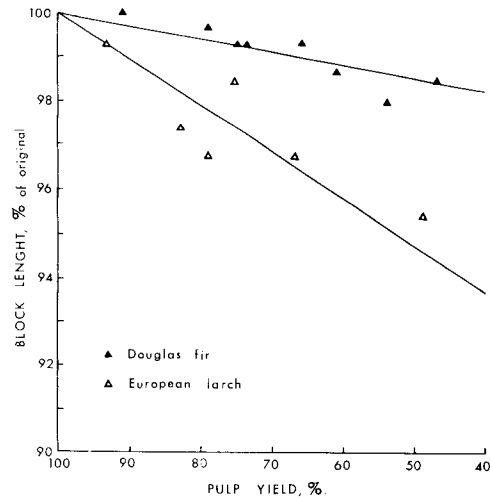
Although the precision of these measurements was not high, reductions of 2% and 6% were observed at 40% yield in the Douglas-fir and European larch samples respectively (Fig. 2).

Change of the transverse dimensions of fibres with yield

If, as has been shown, the length of fibres is hardly affected by pulping, then we may expect that the removal of wood components will be most manifest in a change in the transverse dimensions of the fibres. Indeed, it may be calculated that a plot of the fraction to which the cross-sectional area of the fibre wall ($b^2 - a^2$) is reduced with yield is identical to that of coarseness versus yield (Fig. 1).

The effects of pulping upon the individual parameters, fibre width, and cell-wall thickness, are given in Figs. 3 and 4. As suspected, the main effect is a reduction in cell-wall thickness. The percentage reduction is not, however, exactly equal to the reduction in yield as indicated in earlier work where only either lignin or hemicellulose was removed from the wall [Stone et al. 1971; Kerr 1974] and there is a significant reduction in fibre width not indicated
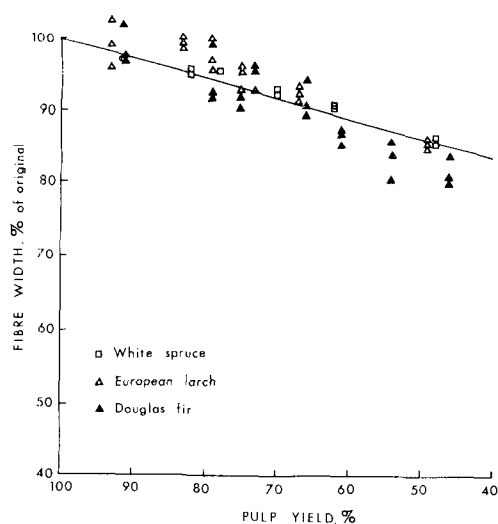
Fig. 3. The effect of pulping upon the width of wood fibres as measured oven-dry before and after pulping.

Fig. 4. The effect of pulping upon the cell-wall thickness of wood fibres as measured oven-dry before and after pulping.

in the earlier work. To check whether these differences occurred because previously only one component was removed, whereas both lignin and hemicellulose were removed in the present work, blocks were examined by the present techniques before and after a chlorite delignification to 78% yield. It was found that within experimental resolution the effect of an essentially pure delignification on fibre dimensions was identical to that of removing both hemicellulose and lignin by a pulping reagent. We believe that the techniques used in the previous study were only sufficient to detect the main trend in transverse dimensional changes, i.e., the wall thickness change, and were not sensitive enough to detect the more subtle change in fibre width.

Figure 5 is a sketch, based on the present work, of how the cross section of a typical earlywood fibre of a softwood changes with pulping. As is seen, there is an appreciable inward contraction of the fibre—the outer perimeter of the cooked fibre wall being less than the perimeter of the lumen before cooking. We may suppose that this is a result of considerable movement of structural elements within the wall during the pulping process.
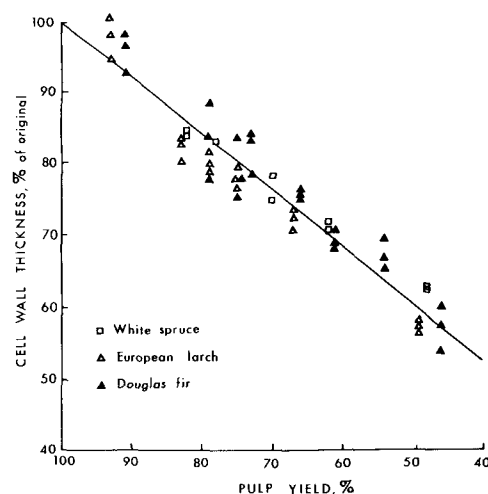
Arrangement of components in the cell wall

Much microscopic evidence indicates that the cellulose component of the cell wall of *pulp* fibres exists as a series of fine lamellations concentric with the fibre axis. It has therefore been reasonable to assume that the
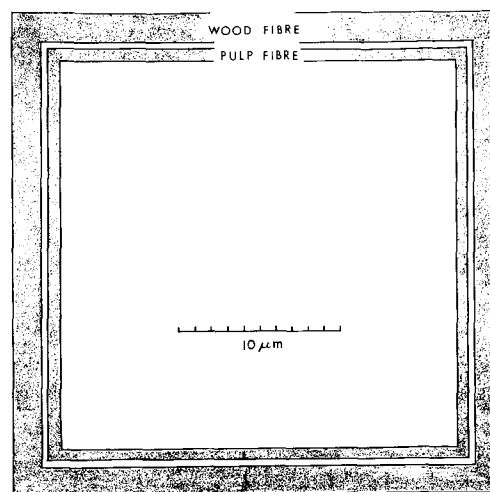

Fig. 5. Sketch showing the reduction in the transverse dimensions of an average earlywood fibre of white spruce as brought about by pulping to 40% yield.
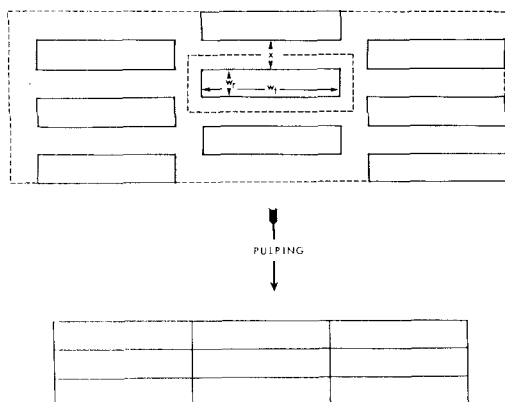
Fig. 6.   Diagram of the type of ultrastructural arrangement of cellulose (unshaded) and lignin-hemicellulose (shaded) within a transverse section of the cell wall, which could explain the changes in the transverse dimensions of fibres upon pulping.

lignin-hemicellulose material in *wood* fibres occupies the spaces between these lamellae and hence takes on a lamella-like distribution itself [Stone and Scallan 1968]. However, examinations of cross sections of wood by electron microscopy either after dissolution of the carbohydrate [Sachs et al. 1963] or after staining the lignin by permanganate [Heyn 1969] have led to the conclusion that the lignin is distributed randomly as fine particles. In order to explain this anomaly, it has been suggested that the cellulose component of the cell wall might appear as perfectly concentric lamellae only after pulping and extensive swelling and that, in wood, the lamellae are periodically broken by lignin intrusions and alternate sections laterally displaced [Scallan 1974]. A cross section of such a structure, as drawn diagrammatically in Fig. 6, is in keeping with the findings of the present study. Removal of the sheath of lignin-hemicellulose by pulping, followed by the drawing together of the asymmetrical bundles of microfibrils upon drying, would necessarily result in shrinkage that is greater in the radial than in the tangential direction of the wall.

Relative dimensions may be given to the average repeating unit of the structure by assuming that the sheath is completely dissolved at 40% yield and then by equating

the reduced tangential and radial dimensions of the repeating unit to the fractions to which the fibre width and cell-wall thickness are observed to be reduced at the same yield. That is:

$$\frac{W_t}{W_t + x} = 0.84$$

and:

$$\frac{W_r}{W_r + x} = 0.525,$$

where W_t and W_r are the average widths of the cellulosic core in the tangential and radial directions respectively, x is the separation of the cellulosic cores, and 0.84 and 0.525 are taken from Figs. 3 and 4 respectively. Solving these equations we find that the width of the cellulose core is 4.75 times greater in the tangential direction than the radial. Further, if we suppose that the cellulose unit is only one microfibril wide in the radial direction ($W_r = 35\text{Å}$), then it will be about five microfibrils wide in the tangential direction of the wall and the lignin-hemicellulose layer separating cellulosic cores will be somewhat thinner than a microfibril ($x = 32\text{Å}$).

The dimensions thus calculated are very close to those recently reported by Kerr and Goring [1975], who have also presented evidence for this type of model. Examining closely micrographs of transverse sections of wood after permanganate staining of the type previously obtained by Heyn [1969], Kerr and Goring concurred with Heyn that stained and unstained regions alternated in the radial direction of the wall and were each of the order of 35Å. However, whereas Heyn reported a similar distribution in the tangential direction, Kerr and Goring reported a repeat distance of 152Å, which they attributed to groups of 2 to 4 microfibrils bonded on their radial planes.

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