A STUDY OF LOBLOLLY PINE GROWTH INCREMENTS—PART III REFINING CHARACTERISTICS OF TRACHEIDS FROM KRAFT PULPS

Geza Ifju
Professor, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061

Peter Labosky, Jr.
Assistant Professor, Clemson University, Clemson, SC 29631

and

Frank D. Mitsianis
Electron Microscopist, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061

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ABSTRACT

Growth increments of loblolly pine were divided into five growth zones and kraft-pulped according to four different digestion schedules of 1½, 1¾, 2, and 3 hours. Each of the 20 pulps was subjected to four periods of beating and Canadian Standard Freeness (CSF) was determined for each. It was found that all pulps independent of the extent of digestion or origin of wood from the growth ring had about the same CSF after short periods of beating. After 35 minutes in the beater, latewood pulp freeness was drastically reduced whereas earlywood pulps changed very little. In general, significantly less energy, as indicated by shorter beating times, was needed for latewood pulps to achieve a specific CSF level than for earlywood pulp. Pulps of low yield were easier to refine than those of high yield. Scanning electron microscopic observations revealed that all pulps underwent a tracheid cell-wall peeling sequence. The layer removed from latewood tracheids by beating separated into long fibrillar bundles while earlywood lamellae freed from tracheids remained intact. The influence of this external fibrillation was attributed to differences between the pulps in refining.

Additional keywords: *Pinus taeda*, beating, cell wall layers, earlywood, fibrillation, freeness, growth rings, kraft pulps, lamellae, latewood, SEM, fiber structure.

INTRODUCTION

Loblolly pine (*Pinus taeda* L.) is the most important pulpwood species in the United States. It is therefore not surprising that pulping and papermaking characteristics of this species have attracted much attention during the last few decades. Tracheid and wood properties of loblolly pine have been described by several authors in terms of genetic and environmental conditions. In addition, these properties have been related to the location of wood with respect to intraincremental growth zones and to the associated morphological characteristics of the tracheids (Biblis 1969; Ifju 1969; Ifju and Labosky 1972). It has also been pointed out that pulping characteristics of wood from different parts of loblolly pine growth rings vary within wide ranges (Gladstone et al. 1970; Labosky and Ifju 1972; Gladstone and Ifju 1975). McIntosh (1967) has reported that responses of earlywood tracheids of this species to refining differ significantly from those of latewood. He found that latewood tracheids underwent more external fibrillation than earlywood ones. In addition, diameter of fibrillar bundles released by refining was larger for earlywood than for latewood tracheids.

It has been known for some time that hemicellulose content of pulp fibers affects rate of refining (Emerton 1957; Nordman 1968). Pulps low in hemicellulose are resistant to beating and develop poor sheet properties. Gladstone et al. (1970), Labosky and Ifju (1972), and Gladstone and Ifju (1975) have shown that retention of carbohydrates associated with hemicelluloses after kraft digestion is closely related to the original position of the tracheid within the growth rings of loblolly pine. It can therefore be postulated that tracheids originating from different parts of the growth rings would also have different refining characteristics.

Lignin content is another factor that has been shown to influence refining of pulps. Rate of change in sulfate pulp properties with respect to beating time is inversely proportional to lignin content (Emerton 1957). Thus, sulfate pulps of high lignin contents are likely to have high freeness, and slow beating characteristics and to develop sheets of low tensile strength. Lignin in the cell walls restricts swelling of cellulose and hemicelluloses, thereby preventing development of surfaces available for interfiber bonding (Jayme 1958).

Fiber or tracheid morphology has also been shown to influence refining characteristics of pulps. It was pointed out in Part I of this series (Ifju and Labosky 1972) that highly significant differences exist in the morphological characteristics of tracheids obtained from various parts of loblolly pine growth rings. Gladstone and Ifju (1975) discussed the effects of intraincremental wood morphology on kraft pulping. McIntosh and Uhring (1968) showed that as a result of such morphological differences, loblolly pine earlywood tracheids were harder to refine than latewood tracheids.

In summary, chemical and morphological variations of tracheids obtained by pulping chips from different zones of loblolly pine growth rings are large enough to influence refining properties of pulps. Thus, the objectives of this study were to determine intraincrement refining characteristics of loblolly pine kraft pulps and relate them to inherent tracheid properties.

EXPERIMENTAL PROCEDURES

Loblolly pine growth rings were divided into five growth zones: early earlywood (fraction 1), late earlywood (fraction 2), transition wood (fraction 3), early latewood (fraction 4), and late latewood (fraction 5). Four kraft pulps were prepared from each growth zone; the 20 pulps ranged in yield from 48.8 to 60.7% based on unextracted oven-dry weight of wood. The four cooking periods were 1½, 1¾, 2, and 3 hours. In Part II (Labosky and Ifju 1972), the pulping procedure and the results of chemical analyses of the 20 pulps were reported in detail. In Part I (Ifju and Labosky 1972), the preparation of the material and the morphological characteristics of tracheids and wood were given.

TABLE 1. *Summary of beating times in standard Valley beater applied to experimental pulps obtained from four different digestion schedules*

Digestion Time (h)	Beating Time (min)						
	5	15	25	35	45	50	55
1.50		✓		✓	✓		✓
1.75		✓		✓	✓	✓	
2.00		✓	✓	✓	✓		
3.00	✓	✓	✓	✓			

Refining of the pulps was done in a laboratory Valley beater according to Tappi Standard Method T200-ts-66 with minor deviations from that standard. A total of 250 g disintegrated pulp of each cook (oven-dry basis) was charged to the beater instead of the recommended 360 g. The pulp was then diluted to 16.0 l total volume giving the 1.565% consistency required by Tappi standards. The slurry was circulated in the beater for a period of 5 minutes. Deionized water was used for all pulp dilutions.

Sample withdrawal from the beater was in accordance with the standard except that two 800-ml aliquots were withdrawn rather than one after each interval. These samples were used for the evaluation of handsheet properties. Tappi Standard Method T227m-58 was used for the determination of Canadian Standard Freeness (CSF) after each beating interval. Each of the 20 pulps were treated in the beater for four beating periods as shown in Table 1. This experimental arrangement produced 80 duplicates of treated pulps for analysis. It was decided not to apply the same beating period to all pulps uniformly because of the great differences in handsheet-making characteristics of the tracheids after the different cooking schedules. Although measurements of drainage by the CSF method would have been possible for all pulps, the value of such data seemed doubtful if some of the pulps were unsuitable for papermaking because of over- or under-treatment in refining. However, two common beating periods (15 and 35 min) were applied to all pulps to allow direct comparisons on the basis of common beating times. The usual corrections were made to account for the variations in freeness due to the small experimental deviations from standard temperature and consistency.

After selected beating periods, random tracheids were observed in a scanning electron microscope (SEM) to evaluate the morphological effects of beating. A drop of pulp slurry was placed on the SEM aluminum specimen holder. Water was allowed to evaporate; tracheids thus dried adhered to the aluminum surface. A 10-nm-thick gold-palladium film was evaporated onto the tracheids to produce the necessary conductive surface for SEM observations.

RESULTS AND DISCUSSION

In earlier publications, tracheid morphology and pulping properties of growth zones of loblolly pine increments were related to relative position of wood within the growth rings (Ifju and Labosky 1972; Labosky and Ifju 1972; Gladstone and Ifju 1975). Relative position was defined as radial location within the growth ring expressed as a percent based on total radial width of the growth ring. Similarly, refining characteristics in this paper are also related to the origin of chips with respect to the total width of growth rings. Mechanical treatment of pulps in refiners is known to affect

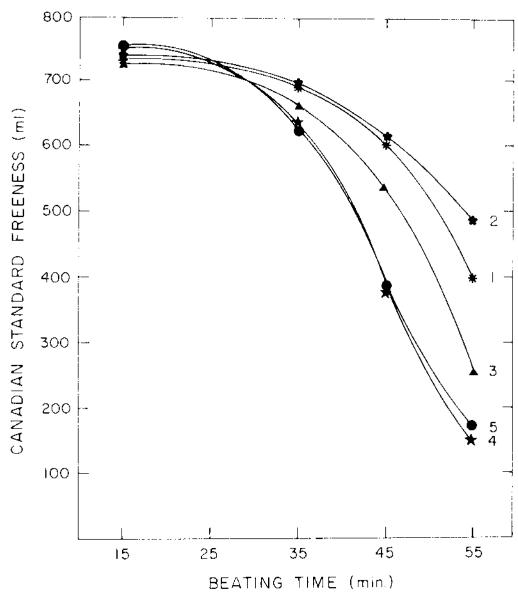

FIG. 1. Influence of beating time on CSF of pulps made from wood of five incremental growth zones using a 90-min kraft digestion.

fiber morphology, drainage characteristics, and physical properties of paper made from them (Emerton 1957; Page and DeGrace 1967). Accordingly, results of this experiment are discussed in terms of CSF as a measure of drainage and changes in cell morphology influencing CSF. Physical properties of paper sheets will be discussed in a subsequent paper.

Drainage characteristics of pulp

Although CSF is an empirical measure of the extent of refining, it was chosen in this study for the evaluation of the influence of beating on the drainage characteristics of the pulps. It is realized that drainage is only one effect of mechanical treatments on pulps, but it is well accepted by the industry as a quick way to assess influences of beating on fibers.

Refining characteristics of tracheids from the five growth zones obtained by the four cooks are shown in Figs. 1 through 4, respectively. Each figure represents CSF vs. beating time for one of the four digestion schedules. The five lines in the figures de-

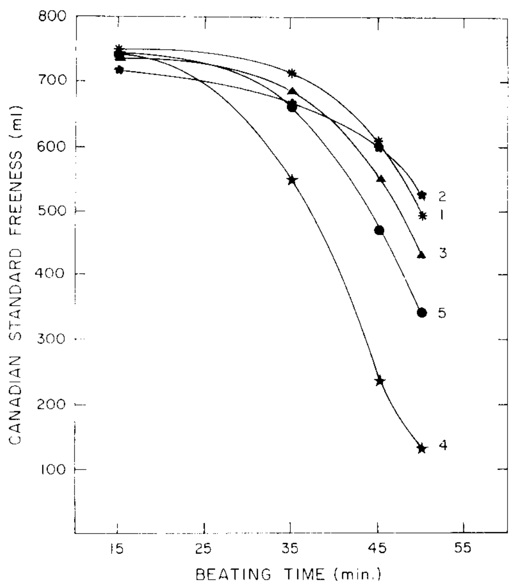

FIG. 2. Influence of beating time on CSF of pulps made from wood of five incremental growth zones using a 105-min kraft digestion.

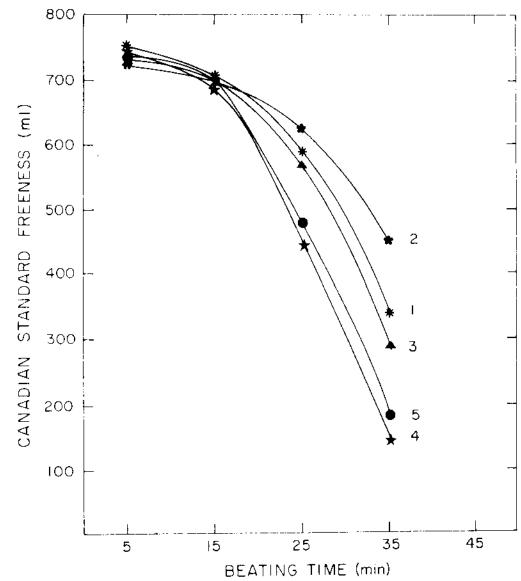

FIG. 3. Influence of beating time on CSF of pulps made from wood of five incremental growth zones using a 120-min kraft digestion.

scribe the responses of the five fractions obtained from the five growth zones to beating in terms of changes in CSF.

Although the general appearance of the lines in the four figures is quite uniform, a closer examination reveals significant differences among them. First, it should be noted that each diagram shows a general change of freeness with beating time from a maximum of about 700 ml for all pulps to a minimum of 150 ml CSF for some pulps. However, the high-yield pulps obtained by shorter cooking times (Figs. 1 and 2) required longer beating periods than did the low-yield pulps (Figs. 3 and 4) to achieve the same CSF levels. Second, pulps from earlywood fractions changed relatively little in CSF upon beating as compared to those made from latewood zones. Third, early earlywood pulps (fraction 1 in the figures) appear to show an anomalous behavior in changing CSF with respect to beating time.

The reduction of CSF with increasing beating time is entirely according to expectations. Also the generally faster change of CSF of low-yield pulps in the beater is a well-known phenomenon to the papermaker. In this study the pulps from the 3-h cooks attained essentially the same freeness values in 35 min as those from the 2-h cooks in 45 min. Corresponding beating time to achieve the same CSF values for the 1¾-h cook was 50 min and for the 1½-h one was 55 min. These results are explainable on the basis of residual lignin contents. It was shown earlier (Labosky and Ifju 1972) that lignin content of pulps varied with respect to digestion time. Emerton (1957) and Jayme (1958) considered lignin content as one of the major factors influencing rate of change of pulp properties upon refining. Results of this study appear to support the lignin content-refining rate interrelationship.

It is evident in Figs. 1 through 4 that latewood pulps required appreciably less energy of refining to attain a particular freeness level than earlywood pulps. All pulps had about the same drainage characteristics after short refining times, but they clearly separated into pulps of distinctly different freenesses. The two latewood fractions, designated as 4 and 5 in the figures, began at about 750 ml but reached

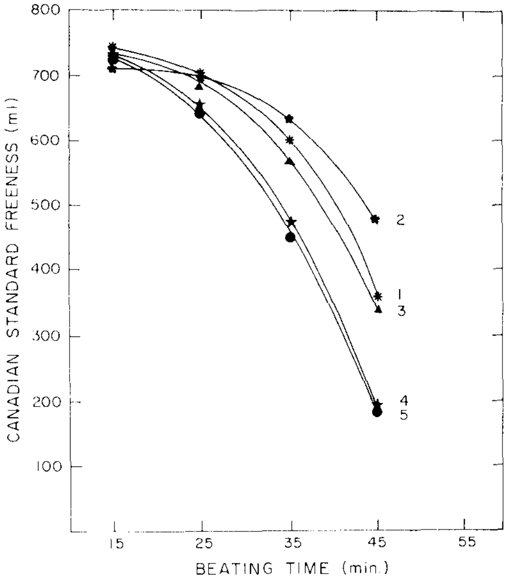

FIG. 4. Influence of beating time on CSF of pulps made from wood of five incremental growth zones using a 180-min kraft digestion.

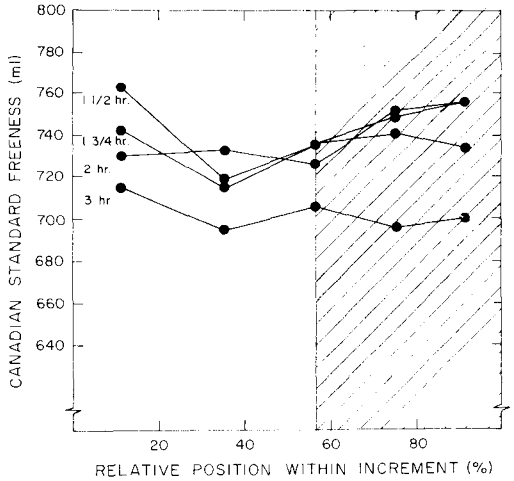

FIG. 5. Relationship between CSF of pulps from four different kraft digestions beaten for 15 min and relative position of wood chips with respect to the growth rings.

CSF values below 200 ml for periods in the beater of 30–40 min depending on cooking time. For the same beating time one fraction of the earlywood pulps, designated as 2, was reduced from CSF of 725 ml to about 500 ml. Fraction 3, the transition zone pulp, behaved in an intermediate manner in terms of its response to refining between earlywood and latewood pulps. These results are in general agreement with those of McIntosh and Uhring (1968), indicating that thin-walled earlywood tracheids require appreciably more energy to refine than do latewood ones.

Refining curves of pulps produced from early earlywood, designated in this study as fraction 1, lie between late earlywood and transition wood in Figs. 1–4. The anomalous properties of wood laid down at the beginning of the growing season have been pointed out earlier regarding tracheid morphology and pulping (Ifju and Labosky 1972; Labosky and Ifju 1972). This study shows yet another property of this growth zone indicating that in some respects it behaves like earlywood, in others more like latewood. Drainage characteristics of pulps are influenced by many factors. Morphology of early earlywood tracheids would suggest pulp drainage similar to earlywood, while residual lignin content appears to make it behave like latewood. The result is that its drainage rate and its response to beating take place between those of the two extreme growth zones.

In this study the differences between refining characteristics of pulps from the two extreme growth zones were especially large for the two high-yield pulps, indicating an interaction between tracheid morphology and pulp yield or residual lignin content. This interaction is best illustrated in Figs. 5 and 6 where CSF values of the pulps obtained from the four digestion schedules beaten to 15 and 35 min, respectively, are plotted against origin of the chips within the growth ring. After a short period of refining, there appears to be very little difference among pulps independent of inherent tracheid morphology or of the extent of delignification (Fig. 5). Only the low-yield pulps of 48.7 to 54.7% yield, obtained by three hours of digestion, appear to have drainage properties different from all the other pulps. In addition, no apparent effect

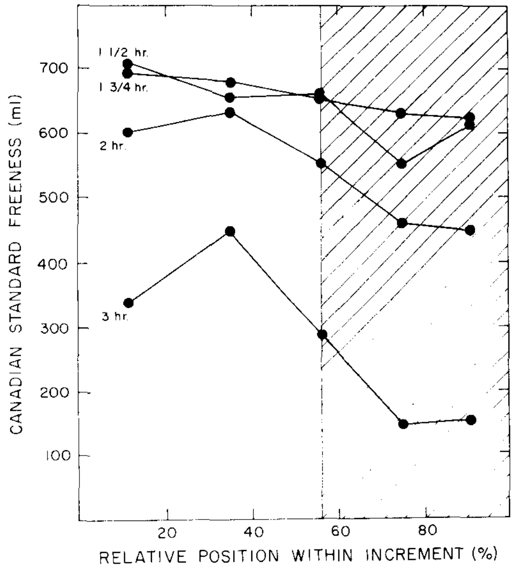

FIG. 6. Relationship between CSF of pulps from four different kraft digestions beaten for 35 min and relative position of wood chips with respect to the growth rings.

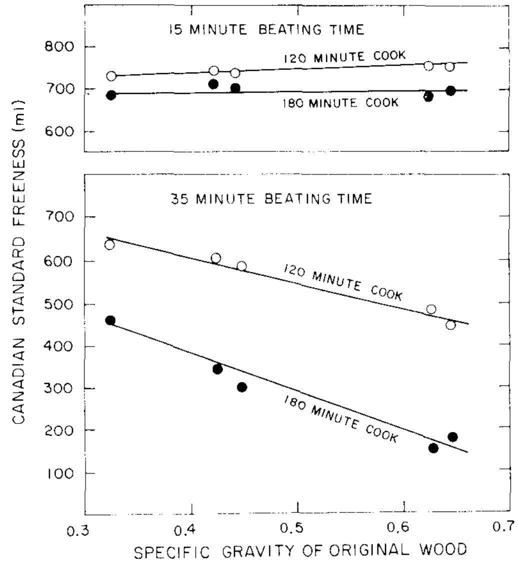

FIG. 7. Dependence of CSF of loblolly pine kraft pulp on specific gravity of wood, cooking time, and beating time.

of tracheid morphology can be seen in Fig. 5 as the lines representing CSF are more or less horizontal over the range of within-growth-ring tracheid characteristics. This might be surprising in light of the rather large differences in tracheid morphology reported by Ifju and Labosky (1972) for the same pulps. It might be expected that thin-walled, flexible tracheids obtained from the beginning of increments would exhibit appreciably slower drainage properties than latewood ones. Apparently, all unbeaten or lightly beaten tracheids remained quite stiff, independent of cell-wall thickness or other morphological characteristics. This observation is in agreement with general pulpmill and laboratory experience that most chemical pulps have an initial freeness around 700 ml. Apparently a minimum amount of mechanical work has to be spent on pulps of widely different morphology and even yield before tracheid characteristics become important in determining drainage properties.

Figure 6 bears out the above conclusion. After 35 min of refining, all pulps of the same digestion time decrease in CSF, from earlywood to latewood. Especially striking is the difference between fractions 2 and 4 of the 3-h cooks. While the earlywood pulp dropped from 695 CSF to 450 CSF between 15 and 35 min of beating, the corresponding loss of freeness for the latewood fraction was from 695 CSF to a level below 150 CSF. This large difference in the response of tracheids of different cell-wall morphology to refining suggests that in industrial operations some fibers must be over refined while others are not changed to the point where they can effectively contribute to sheet properties. These differences are probably magnified on the industrial scale when pulps obtained from different species of wood are refined together, a common practice in many pulpmills.

When assessing wood for any use, including pulping, it is customary to determine specific gravity as an indicator of quality. In the context of this paper, wood density should be considered an indirect but easy measure of tracheid morphology. In this regard, then, relating pulp freeness to specific gravity of wood from which the

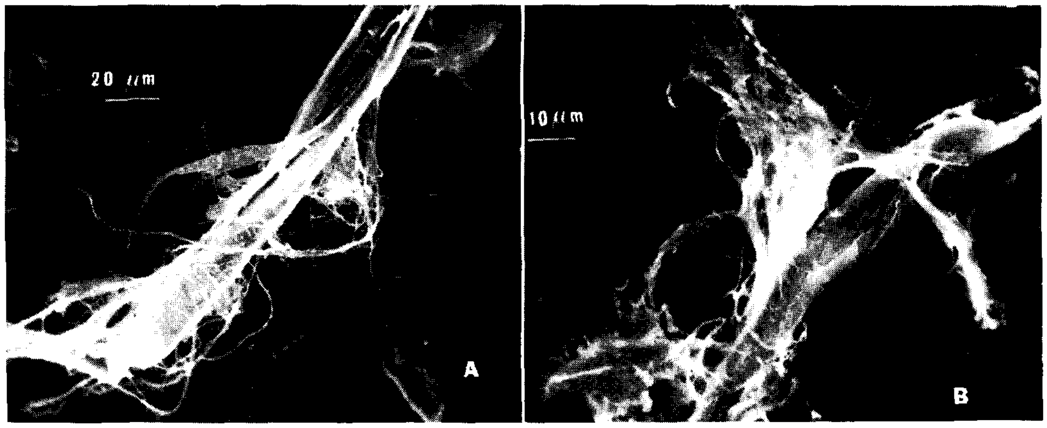

FIG. 8. SEM micrographs of two latewood tracheids from kraft pulps after 35 min beating; A) from pulp of 105-min digestion; B) from pulp of 180-min digestion.

pulp was produced is meaningful. Figure 7 shows the relationships between freeness and specific gravity after 15 and 35 min of beating for the 2- and 3-h pulps. Again the response of pulps of thick-walled tracheids to beating beyond 15 min is evident. Figure 7 shows perhaps more clearly that wood properties as estimated by specific gravity should be taken into account in deciding on optimum refining treatment of pulps for a specific use. In addition, optimum utilization of mechanical energy in refining requires some type of separation of pulps of widely different fiber morphology before the treatment.

Morphological effects of refining

In order to find the reasons for the differences in refining properties of pulps of different tracheid characteristics, individual tracheids were observed in the SEM. Here only a few typical tracheids are shown, but a great number were studied after various beating periods.

The primary external effect of beating on all tracheids observable in the SEM appeared to be delamination of the cell walls and gradual peeling of the laminae as beating continued. This peeling action was noticeable after 35 min of beating of all tracheids independent of their origin with

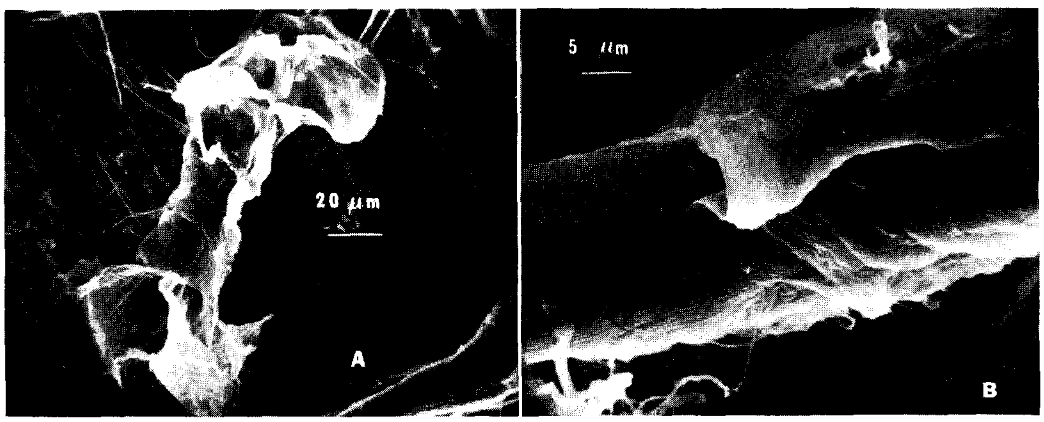

FIG. 9. SEM illustration of the effects of 35-min beating on earlywood tracheids from kraft pulps; A) intact wall layer separated like "snake skin" from tracheid; B) peeling effect of beating.

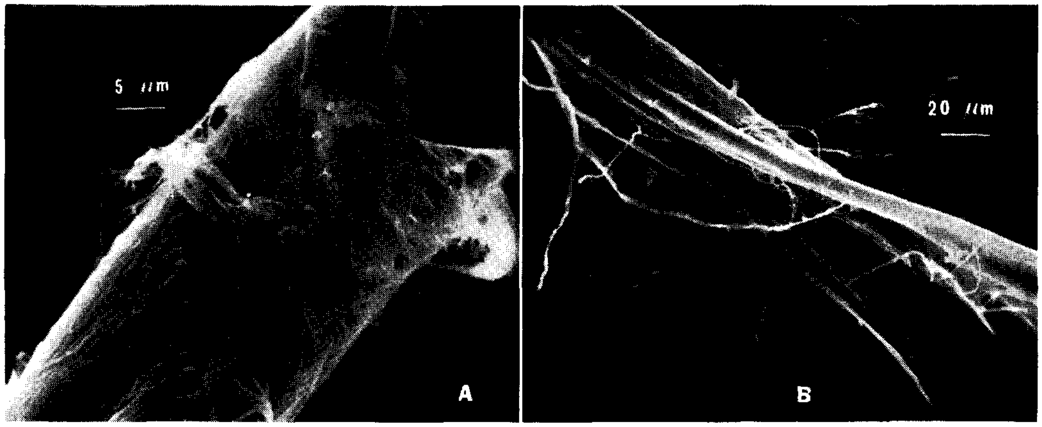

FIG. 10. SEM micrographs of transition wood tracheids from kraft pulps after 35-min beating; A) S_1 layer peeling of tracheid from 90-min digestion; B) latewood type breakdown of tracheid from 120-min digestion.

respect to the growth increment or extent of digestion. After short beating times of less than 35 min, the majority of the tracheids appeared unaffected by the treatment. Delamination as a primary effect of refining is in line with the laminated cell-wall structure shown for southern pine latewood tracheids by Dunning (1968), caused by the preferential entry of water into tangentially oriented spaces between microfibrils as suggested by Scallan (1974).

Figure 8 shows two latewood tracheids after beating for 35 min. The delamination of the cell walls may be seen in the figure. The cell-wall lamellae appear to originate from the S_2 layer as evidenced by the orientation of the microfibrillar bundles liberated by peeling. It should be noted in Fig. 8 that the cell-wall layers removed from the tracheids are well fibrillated. The layers peeled off latewood tracheids did not as a rule remain intact but disintegrated into long continuous fibrillar bundles.

In Fig. 9B a typical earlywood tracheid is shown after 35 min of beating. The peeling action of the refining process may be seen again in this figure. However, the cell-wall layer being removed from the

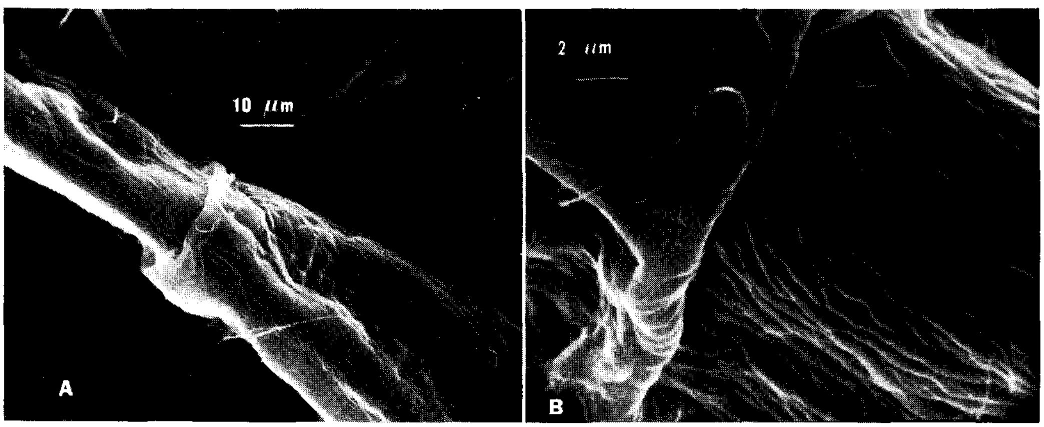

FIG. 11. Two SEM micrographs of an earlywood tracheid from a 120-min kraft digestion and beaten for 35 min; A) lamellation of S_2; B) undisturbed parallel arrangement of fibrillar bundles on exposed S_2 layer.

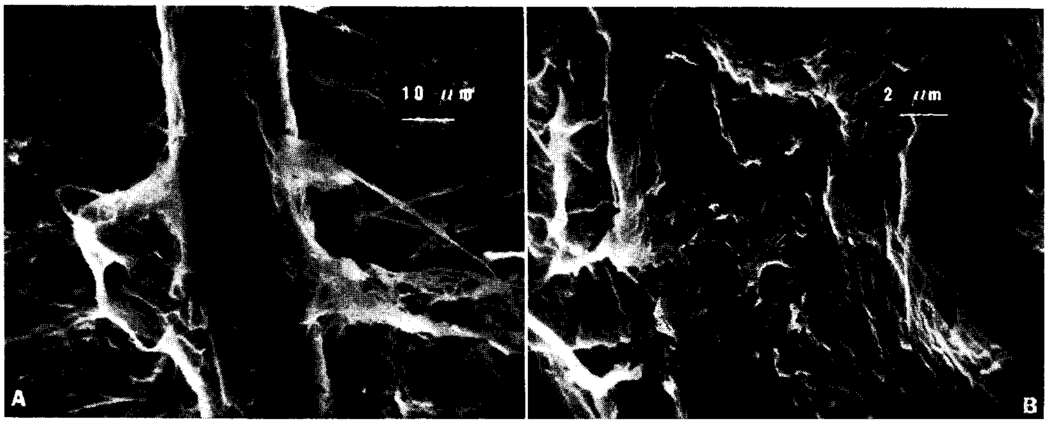

FIG. 12. Two SEM micrographs of a latewood tracheid from a 120-min kraft digestion and beaten for 35 min; A) peeling and fibrillation of outer cell-wall layer; B) disorganized fibrils creating a rough appearance of exposed S_2 layer.

earlywood tracheid does not appear to be fibrillated. This type of peeling was found to be quite common for earlywood pulps. In fact, the layers removed by beating could often be found intact but separated from parent tracheids. Such a "snake skin" is shown in Fig. 9A.

In Fig. 10 two tracheids originating from the transition zone are shown. The tracheid in Fig. 10B shows a latewood-type delamination with subsequent fibrillation. The tracheid in Fig. 10A is undergoing a removal of the S_1 layer, while its outermost S_2 gives an earlywood-type appearance. Both types of tracheids could be found in great numbers in the transition zone.

As was pointed out earlier, the basic difference between earlywood and latewood pulps after beating was in the fibrillation of the layers removed. While earlywood peelings remained intact, latewood ones disintegrated into fibrillar bundles. This difference is seen to be responsible for the greater rate of reduction in freeness of the latewood pulps with beating as compared to earlywood pulps. Freeness is a measure of the drainage characteristics of pulp. Small, flexible fibrillar bundles should be more effective in filling the gaps of the screen upon draining than do intact lamellae removed from earlywood tracheids by beating.

Another difference between earlywood and latewood pulps was the appearance of the tracheid surfaces after beating. Removal of external lamellae from earlywood tracheids left relatively smooth surfaces. Figure 11 shows such a tracheid in two different magnifications. Figure 12 illustrates the appearance of a latewood tracheid. Both of these tracheids were taken from pulps of 3-h digestion beaten for 35 min. Again the fibrillar appearance of the latewood cell-wall layer may be contrasted against the intact earlywood lamellae. Underneath the removed lamellae the latewood cell wall is quite rough, showing no evidence of a regular fibrillar structure. The microfibrillar bundles of about 0.5 micrometer diameter are quite regularly arranged in the outermost S_2 of the earlywood tracheid. This may come from the partial fibrillation of the latewood S_2 even before removal from the tracheid. Upon drying of the cell wall during specimen preparation for the SEM, the loose microfibrillar bundles could be reattached to the surface of the latewood tracheid but no longer in their original, regular, parallel arrangement. Earlywood tracheids, because of their resistance to fibrillation, retained their cell-wall organization even after extensive beating.

CONCLUSIONS

From the results of this study the following conclusions may be drawn:

1) Thick-walled loblolly pine latewood tracheids are refined faster than thin-walled earlywood tracheids.
2) Low-yield loblolly pine pulps are easier to refine than high-yield ones.
3) Initial freeness of both earlywood and latewood pulps is approximately the same after digestion and disintegration.
4) The primary visible effect of beating on loblolly pine tracheids is peeling of the outer cell-wall layers.
5) Fibrillation of cell-wall lamellae removed from loblolly pine latewood tracheids is one major reason for this type of pulp to refine faster than earlywood pulps.
6) Because of the great influence of fiber morphology on refining rate, fibers of greatly different characteristics, such as those originating from loblolly pine earlywood and latewood, should not be refined together if beating energy and pulp quality are to be optimized.

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