The Effect of Refining on the Morphology and Properties of Bamboo Paper

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The morphological characteristics of commercial kraft bnmboo pulp and its four different fractions were studied in regard to their influence on the strength of bamboo paper and the changes that occur on refining. In the unrefined condition, paper made from the whole pulp was superior to that from the fractions in strength properties. On refining, however, the longest fiber fraction developed the highest strength. Relatively thick-walled fibers predominated in the pulp. Collapse of the fibers during dewatering was not common with the thick-walled fibers. Also, refining imparted very little flexibility to these fibres. Bamboo paper developed strength on refining the pulp because of good fibrillation. In the beginning stages, the primary wall of the fibres was removed and formed thin sheaths connecting separate fibres. With increased refining, the inner and outer secondary wall layers were exposed forming fibril bundles and fibrilis which enhanced the extent of hydrogen bonding between fibres. The longer pulp fractions were superior in this regard. The major effect of refining was therefore to increase the strength of the web by fibrillation rather than by increasing fiber flexibility. The amount of parenchyma cells increased from the longest to the shortest fiber fraction, but added very little strength to the paper sheet.

Introduction

Bamboos, which belong to the grass family, grow in many parts of the world. They are considered native to all continents except Europe[1]. There are more than 700 species of which 300 grow in Asia.

Bamboo is unique from the point of view of its growth. The roots are perennial and full growth is achieved in 7 to 10 years. However, shorter rotations of 2 to 4 years and even 1 year are practiced[2]. In the mode of growth, bamboo differs from softwoods and hardwoods in that its tissues originate mainly from primary growth. As a result, the different tissues are arranged only vertically, and basic uniformity is maintained throughout the bamboo culm. Fibres and parenchyma cells form the main part of the culm, parenchyma cells forming the ground mass in which fibro-vascular bundles are arranged in a characteristic manner. Fibers contribute 60-70% of the total weight[3]. The fibers are usually long and straight with tapering ends. parenchyma cells are more abundant in bamboo than in hardwoods and softwoods, and are expected to play a

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more important role in the final property of the paper.

While bamboo is an important raw material for paper making, the paper forming properties of bamboo pulp, in terms of fiber morphology, are not as intensely studied as for western woods. The present study examined the morphological characteristics that influence the strength of bamboo paper, and the changes on refining that affect paper forming properties of the pulp.

Materials and Methods

Cooking conditions of the unbleached bamboo kraft pulp used in the investigation are given in Appendix A. The pulp was classified in a Bauer-McNett classifier using 25 grams of pulp with 15 minutes classification time. The screen mesh sizes were 14, 28, 48 and 100 wires per inch. The process was repeated to collect sufficient pulp in each fraction so that runs in a Mead laboratory refiner could be performed. The percentage of each fraction in the whole pulp was determined. The pulp passing through 100 mesh and overflowing was collected with a 200 mesh screen.

The whole pulp and the four classified fractions were refined in a Mead laboratory refiner. Handsheets were made from unrefined pulp and from pulp refined to various freeness levels. Preparation of handsheets was according to Tappi Standard procedure T402 os-49. The handsheets were tested for their physical strength according to Tappi Standard T205.

The pulp fraction which passed through the 100 mesh screen and was collected on 200 mesh screen was refined for 20 and 30 seconds. Handsheets were made as for the other fractions.

Fibre Measurements

Fibre dimensions of the whole pulp and the four different screen size fractions were measured according to Isenberg[4] except that the fibers were not stained. A minimum of 200 measurements were made of fibre length, and a minimum of 100 measurements were made of fiber diameter and lumen width on each of the five pulp samples. The lumen width and fiber diameter measurements were made at the middle of the fibers.

Scanning Electron Microscopy

Dried handsheet samples were used to study surface characteristics by scanning electron microscopy. The handsheet samples of whole pulp and the four fractions of various degrees of refining were mounted on the specimen stub holders, and a thin conductive coating of gold was applied in the usual manner for scanning electron microscopy work. Representative micro graphs were taken.

Results and Discussion

Fiber Dimension

In the whole pulp the length varied from as low as 0.75 mm. to higher than 5 mm. The numerical average was 2.3 mm. Clark[5] suggested the use of weighted average fiber length, as a meaningful way of expressing fibre length, especially in the case of fibers which vary widely in length and contain fiber debris. The weighted average of the whole pulp was calculated from the percentage of each fraction in the whole Pulp. The weighted average length of the fraction passing through 100 mesh screen was assumed to be 0.2 mm.

The fractions were designated as follows :

Fraction	Passed Through	Retained In
R-14	—	14 mesh
R-28	14 mesh	28 mesh
R-48	28 mesh	48 mesh
R-100	48 mesh	100 mesh
P-100	100 mesh	—

The amount of parenchyma in the pulp increased from R-14 to R-100 fractions, the P-100 being mostly parenchyma, broken fibers and debris.

Table I summarizes all the fiber dimension[5], fiber length (L), fiber diameter (d), lumen width (l), cell wall thickness (w) and the derived values: felting coefficient, coefficient of flexibility, Runkel ratio and the ratio of length to wall thickness.

Strength of Unrefined Sheets

Table II shows the strength properties of handsheets made from unrefined whole pulp and fractions. The pulp fractions showed a general tendency of increase of tensile breaking length and bursting strengths from R-14 to R-100. The fine (R-100) fraction showed higher tensile and bursting strength compared to the longest (R-14) fraction.

TABLE 1

Fiber Dimensions and Derived Values of Whole Pulp and Fractions, Unrefined

Pulp	% Retained	L (mm)	d (μm)	l (μm)	w (μm)	L/d	l/d x 100	2w/l	L/w
Whole	—	1.70	23.6	9.5	7.0	72	40.5	1.47	243
R-14	34.5	3.07	25.5	10.1	7.7	120	38.0	1.52	399
R-28	15.8	2.23	23.4	10.3	6.6	96	43.6	1.28	340
R-48	9.4	1.55	19.4	7.9	5.7	80	40.8	1.45	271
R-100	7.9	1.05	19.5	10.2	4.6	54	52.5	0.91	226

L.................fiber length
d..................fiber diameter
l............ ......lumen width
w... . ...cell wall thickness
L/d............ . felting coefficient
l/d × 100......coefficient of flexibility
2w/l...............Runkel ratio

TABLE II

Strength Properties of Unerfined Whole Pulp and Fractions

Pulp	Freeness CSf (Ml)	Bulk (cm³/gm.)	Burst Factor	M. I. T. Fold	Breaking Length (m × 100)	Tear Factor	Zero-span Breaking Length (m × 100)
Whole	718	2.47	8.6	3	27.5	130	64.1
R-14	727	2.73	5.6	2	14.8	116	87.8
R-28	723	2.67	6.6	3	19.9	94	74 3
R-48	705	2.55	5.6	3	18.3	65	77.5
R-100	683	2.35	6.0	2	20.0	42	56.6
P-100	—†	2.01	1.7	—*	7.9	11	16.2

†The pulp was too fine to be retained by freeness tester screen.
*Negligible.

This is different from the observed behaviour of the chemical pulp fractions from various wood species[6,7]. The longest fraction from the chemical pulp gives the strongest sheet while the finest fraction from mechanical pulp gives the strongest sheet. This behaviour in the case of ground wood was explained by Marton and Alexander[7] as due to the stiff non-elastic nature of the longer fraction. These are unfibrillated and stick like in nature. They produce a bulky sheet with low strength. The fines from ground wood pulp show increased fiber bonding because of their increased surface area. They also fill in the void spaces between fibers thus given more densely packed sheets. Together, these two properties give higher sheet strength. Cruz-Gonzalez and Escolano[8] gave a similar explanation in the case of pulp fractions from giant bamboo. The increased amounts of thin-walled vessels and parenchyma of the fine fraction fill voids between the stiff fibers and thus form a denser sheet and one of higher strength.

The fraction P-100 handsheets which contained mostly parenchyma, some broken fibers and fiber debris had very low strength properties including the zerospan tensile breaking length. This differs from the finding of Runkel[9] who separated parenchyma cells from African balsa wood pulp and made sheets which had high breaking length and fold.

Parenchyma in the whole pulp might contribute to the bonding because of their thin walls and collapsed nature, although by themselves they have very poor strength[10]. This may be a contributing cause of the greater strength in the unrefined condition of the whole pulp compared to its fractions which have a lower percentage of parenchyma.

The zero-span tensile strength showed a highest value for R-14 indicating the highest individual fiber strength. R-28 and R-48 showed almost equal values, and R-100 the least strength. The whole pulp value was slightly better than the R-100 fraction.

Neither the fiber measurements and their derived values nor the individual fiber strengths showed any correlation with the sheet strength in the unrefined condition.

Freeness

Figure 1 shows the freeness response of whole pulp and the various fractions to the mechanical refining. The longest fiber fraction showed the highest freeness in the unrefined condition. This initial freeness decreased from the longest to the finest fraction. The freeness of the unrefined whole pulp was intermediate between that of R-28 and R-48 fractions.

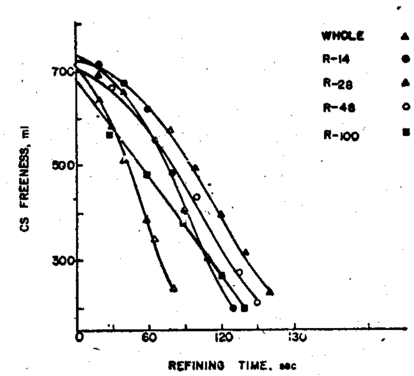

Fig. 1. Relationship of Canadian standard freeness and refining time.

The whole pulp showed the highest refining rate. Among the fractions, the longest fraction showed a slightly greater rate of freeness decrease than the other fractions. All others followed curves almost parallel to each other.

Refining and Strength Properties

Figures 2 and 3 show the development of burst and tensile strengths of the sheets with degree of refining, expressed as

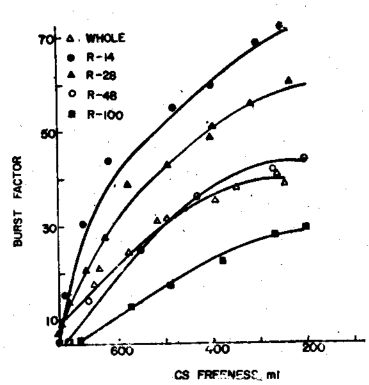

Fig. 2. Relationship of burst factor and Canadian standard freeness for whole pulp and four different fractions.

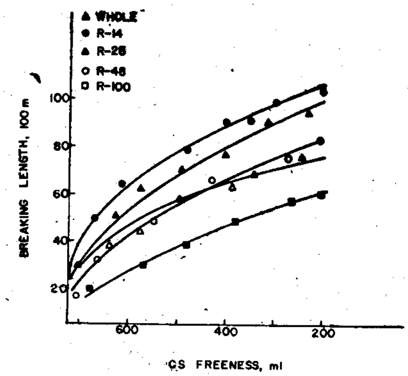

Fig. 3. Relationsphip of tensile breaking length and Canadian standard freeness for whole pulp and four different fractions.

burst factor and breaking length, respectively. The pattern of development was very similar for both. The longest fraction developed the highest strength and the capacity to develop strength decreased with screen mesh size of the fractions:

Stretch and tensile energy absorption (Figures 4, 5) followed the pattern of burst and tensile strength on refining. After a

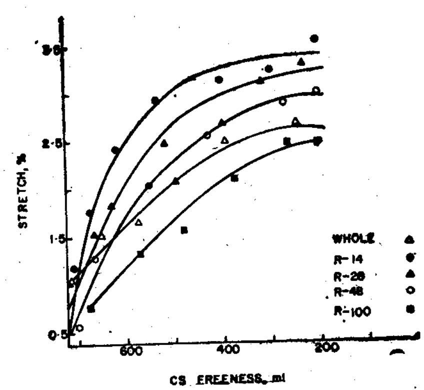

Fig. 4. Relationship of stretch and Canadian standard freeness for whole pulp and four different fractions.

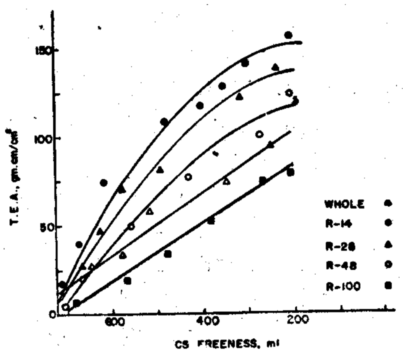

Fig. 5. Relationship of tensile energy absorption and Canadian standard freeness for whole pulp and four different fractions.

certain freeness level was reached no further increase in stretch was noticed. Figures 6 and 7 show the effect of refining on fold and tear values. Here again can be seen the highest tear & fold values for R-14 and decreasing values with decreasingscreen mesh size of the fractions. In all the above cases, it could be seen that the strength values for the whole pulp were near and often exceeded. by the R-48 fraction values.

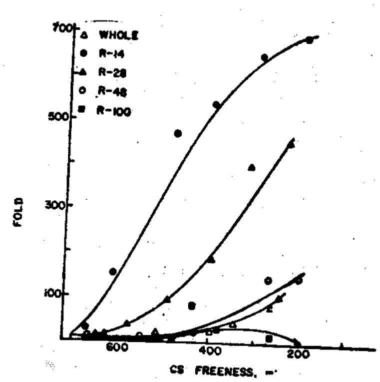

Fig. 6. Relationship of M. I. T. fold and Canadian standard freeness for whole pulp and four different fractions.

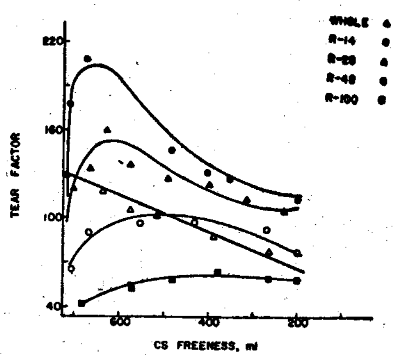

Fig. 7. Relationship of tear factor and Canadian standard freeness for whole pulp and four different fractions.

Fiber Length and Strength Properties

In the case of wood pulp fibers, correlation of fiber length to burst and tensile strength has been established by many investigators([5,11,12]). Figures 8 and 9 show the change in burst and tensile breaking length, respectively, with fiber length. These strength properties for different freeness levels were

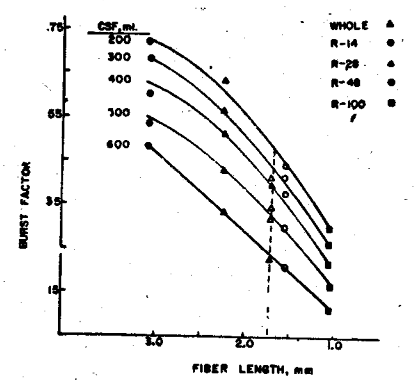

Fig. 8. Relationship of interpolated values of burst factor with weighted average fiber length at various freeness levels.

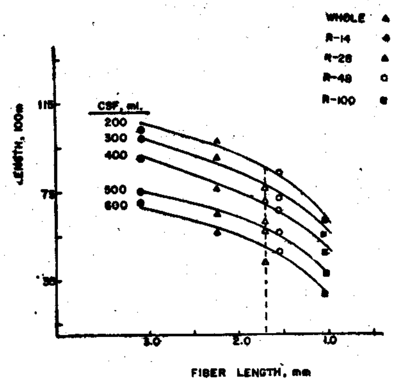

Fig 9. Relationship of interpolated values of tensile breaking length with weighted average fiber length at various freeness levels.

obtained by interpolation from the original values. As can be seen from the figures, the burst and breaking length follow a nearly linear relationship with the fiber length; and all the curves representing different freeness levels are roughly parallel. The dotted line follows the data of the whole pulp at its weighted average fiber length. These values lie below the value expected for that fibre length from the fraction curves except at the higher freeness levels. This could be explained as due to the effect of a high percentage of parenchyma in the whole pulp which did not contribute strength to the paper[10]. This is also supported by the drooping of the line at the point corresponding to the R-100 fraction which contained the highest percentage of parenchyma among the fractions.

Figures 10 and 11 show tensile energy absorption and stretch percentage versus fiber lengths. These curves are similar to the pattern shown by burst and tensile strength.

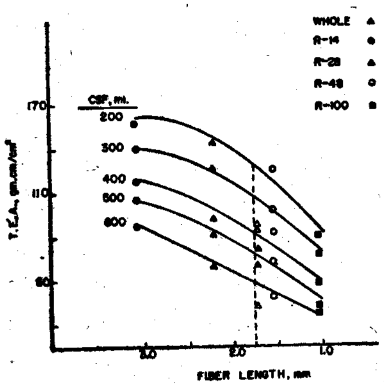

Fig. 10. Relationship of interpolated values of tensile energy absorption with weighted average fiber length at various freeness levels.

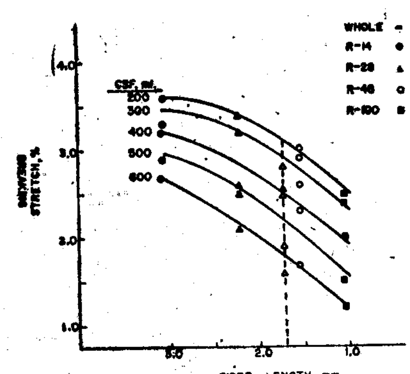

Fig. 11. Relationship of interpolated values of stretch with weighted average fiber length at various freeness levels.

Figure 12 shows the interpolated values of tear with fiber length. The dependency of tear on fiber length is so great that the R–100 fraction containing the shortest

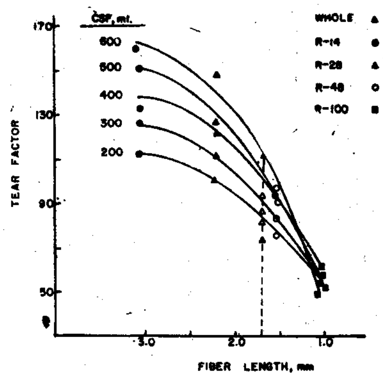

Fig. 12. Relationship of interpolated values of tear factor with weighted average fiber length at various freeness levels.

fibers show very low tear values. In contrast the longest fraction showed very high tear values.

Derived Relations :

Runkel ratio and coefficicient of flexibility are significantly correlated with tensile and breaking length in the case of wood fibers. The present study with bamboo fibers showed no particular correlation between strength properties and these dimensional variables.

Density:

Density has been considered as a most important and dominant factor contributing to paper strength. It is also considered to have an effect on other strength properties. Density is largely governed by the degree of plasticity developed in the wet fibers in the process of refining. The longer fibers tend to give a higher density because of a high axis ratio or flexibility ratio[13].

Density is also related to interfiber bonding, because the denser the sheet, the closer the fiber surfaces are during the drying process. This enhances the probability of hydrogen bonding ([14,15,16]) with accompanying strength. This again depends on the type of fiber and keeping the process of sheet formation, wet pressing and drying conditions identical. Doughty[17] considered density as the dominant factor whether developed by beating or wet pressing.

The bulk values (inversely proportional to density) of different fractions in Figure 13 differ

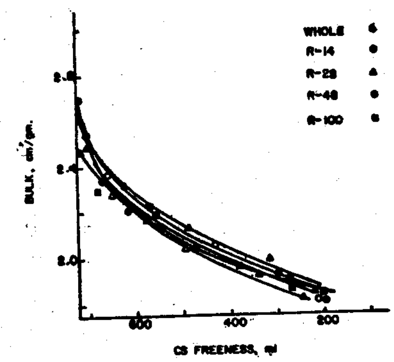

Fig. 13. Relationship of bulk and Canadian standard freeness for whole pulp and four different fractions.

very little from each other after the initial stage of refining. If density is the dominant factor, then the strength values of different fractions at the same density or freeness values should be identical. This is not actually the case. Thus density is not the only factor contributing to strength with regard to bamboo paper.

The Structure of Bamboo Fibers:

Preston and Singh ([18,19]) studied individual fibers from a variety of bamboo species and described their essentially lamellated structure, Figure 14 ([20])

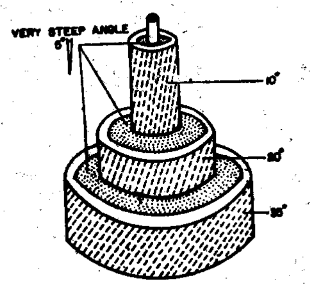

Fig. 14. Orientation of cellulose microfibrils in the bamboo fiber, according to Preston[20].

represents a model of a bamboo fiber as described by these workers. Outermost is a thin layer of cellulose fibrils making a 35° angle to the cell axis. In subsequent inner layers the angle steadily decreases first to 20° and then to 10°. These layers are separated by thick layers in which the angle is seldom more than 5–6° on the average. Figure 15 is a

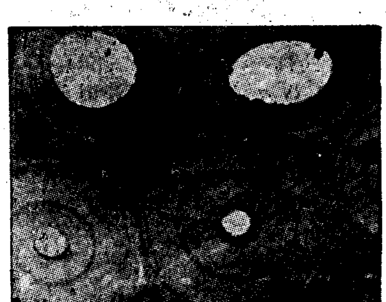

Fig. 15. Ultra-thin cross section of bamboo showing variation in cell wall layering of fibers. 4350x.

cross section of fibers as seen in the transmission electron microscope[20] showing the concentric layering. Because of the concentric layering, bamboo fibers retain their inherent stiffness until advanced stages of processing. Though the majority of the fibers are thick-walled, there also exist a certain amount of thin-walled and medium thick-walled fibers.

The structure of different cell wall layers from softwood fibers on drying was investigated by Jayme and Hunger[21]. They showed that if the primary wall, after removal, is suspended without support, such as when spanning between fibers, it will form a ring net or perforated sheet-type structure. However, if the primary wall is bound to another fiber surface, it does not show any alteration of its normal structure. The secondary wall material when removed and dried, shows micro-fibril bundles that appear as rope-like structures or sheets of parallel oriented micro-fibrils. These structures, originating from the primary wall and secondary wall, were also observed with bamboo fibers, as seen in Figures 16 through 19.

Imporved paper structure and fiber bonding are the resultant effects of refining. There are two main factors which contribute to better bonding between fibers: first, the breaking of intra-fiber bonds and fibrillation, and secondly, which results from the first, increased wet flexibility and subsequent fiber collapse which increases contact area and total bond strength at fiber crossings.

The achievement of bonding due to refining in the case of bamboo fibers was observed, Figure 20

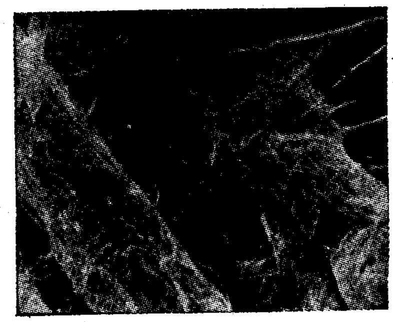

Fig. 16. Handsheet surface of refined bamboo pulp showing perforated sheet type structure probably due to primary wall material (lower center of micrograph). All fibers possess drying ridges. 3000x.

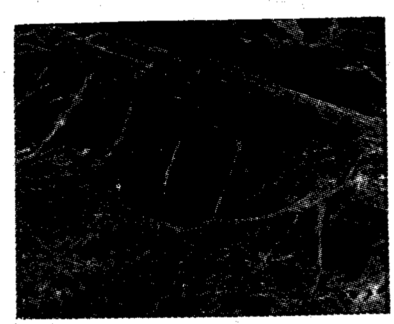

Fig. 17. Handsheet surface of refined bamboo pulp showing formation of rope-like fibrillar bundles from secondary wall. 1530x.

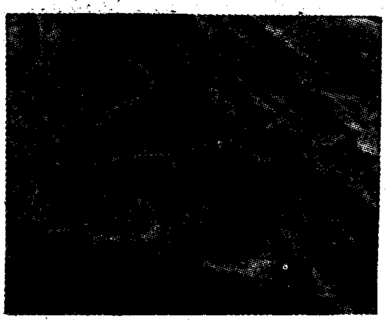

Fig. 18. Handsheet surface of refined bamboo pulp showing removal of secondary wall layer and exposure of inner secondary wall material (upper right of micrograph). Large rope-like bundles of microfibrils from secondary wall present (center of micrograph). 3900x.

Fig. 19. Handsheet surface of wall refined bamboo pulp showing possible secondary wall-type bonding typified by sheets of parallel oriented micro-fibrils (upper left of micrograph.) 1520x.

shows the unrefined pulp with inflexible, stiff fibers, with slight refining the flexibility of the fibers was not affected (Figure 21), but bonding due to removed primary wall material was evident at practically the same freeness level (Figure 22). At 495 ml CS, fibers have retained their stiffness but bonding has improved (Figure 23), With increased refining, a high degree of fibrillation results and the secondary

Fig. 20. Handsheet surface of urefined bamboo pulp showing stiff fibers with very little fibrillation and bonding between fibers 425x.

Fig. 21. Handsheet surface of slightly refined (640 ml CSf) bamboo pulp showing little increase in flexibility from unrefined condition. Uncollapsed thick-walled fiber (upper part of micrograph) and collapsed thin-walled fiber (lower part of micrograph) visible. Central large diameter fiber demonstrates prominent drying ridges. 770x.

wall takes an integral part in inter-fiber bonding (Figure 18).

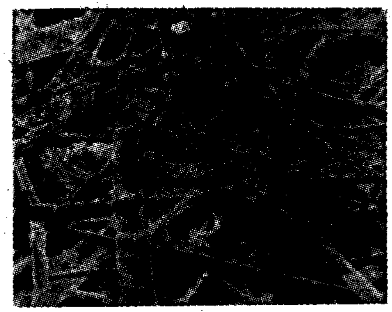

Fig 22. Handsheet surface of slightly refined (620 ml CSf) bamboo pulp showing thick and thin-walled fibers. Primary wall-type, perforated sheet bonding is evident (right center of micrograph). Collapsed thin-walled fiber forming ridges along its edges and bonding to underlying fiber is seen (left center of micrograph.) 160x.

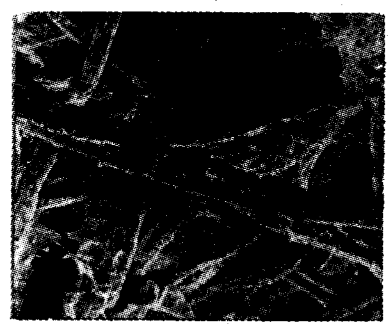

Fig. 23. Handsheet surface of moderately refined (495 ml CSf) bamboo pulp showing increased bonding and fibrillation. Perforated sheet-type bonding evident (upper right of micrograph). 390x.

The removal of the primary wall enhances the fiber bonding[22,23] by exposure of the secondary wall and also connects the inter-spaces between the fibers[21]. The resulting increase in bonding is evident in a corresponding rapid increase in strength properties (Figures 2 through 6).

Collapsing plays the major role in bonding thin walled fibers and is assisted by fibrillation. This is found to be the case with early wood tracheids of softwoods, and was also observed in thin-walled bamboo fibers (Figures 21, 22). When the fibers are thick-walled and stiff, however, fibrillation plays the dominant role. Robertson and Mason[24] showed the development of appreciable strengths by fibrillating rayon filaments.

In the case of bamboo fibers, because of the concentric layering, the layers are probably susceptible to loosening which eases intra-fiber bond breaking by the mechanical action of refiners. Fibrillation is thus assisted and is probably the dominant factor contributing to the bonding of thick-walled, stiff bamboo fibers. The removal of the primary wall, in the early stages of refining is followed by removal of layers from the secondary wall and finally by the removal of fibril bundles from the secondary wall.

Jayme and Hunger[21] contend that the formation of drying ridges on the surface of fibers reduces the chance of hydrogen bonding. This could seriously obstruct inter-fiber bonding in bamboo, where longitudinal ridge formation is prominent (Figures 16, 21). These ridges were also observed by Koran[25] and Thorpe[26] in the case of wood fibers. Buchanan and Lindsay[27] showed that at the fiber crossings, lateral shrinkage is prevented resulting in a lack of ridges in the whole contact area, and there is fibrillar attachment. Although the fibers are not callapsed, the surfaces are brought in close contact by the fibrils at the fiber crossings.

Study with the scanning electron microscope shows that the capacity for bonding in bamboo fibers decreases with decreasing fiber length. The longer fiber fraction fibrillates much better than the shorter fractions, even in the early stages of refining, and thereby develops high strength. Figures 24 and 25 represent the unrefined condition of the longest fraction (R-14) and the shortest (R-100), respectively, where the longer fraction fibers are stiff and rod-like, compared to R-100. At 400 ml CSf, well bonded structures in R-14 are evident (Figure 26) while in R-100, even at 225 ml CSf (Figure 27) there is less bonding and very little fibrillation.

Fig 24. Handsheet surface of unrefined R-14 fraction of bamboo pulp showing stiff fibers with no vessel elements or parenchyma cells. 160x.

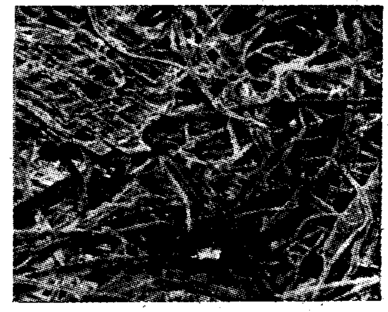

Fig. 25. Handsheet surface of unrefined R-100 fraction of bamboo pulp showing short fibers and high percentage of parenchyma cells and vessel elements. 155x.

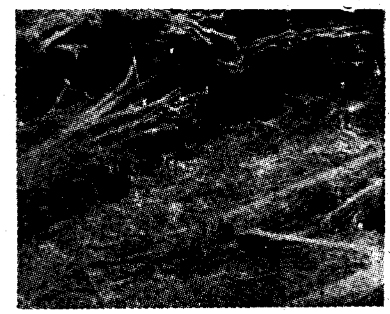

Fig. 26. Handsheet surface of well refined (400 ml CSf) R-14 fraction of bamboo pulp showing good inter-fiber bonding as a result of good fibrillation. 1550x.

Fig. 27. Handsheet surface of well refined (225 ml CSf) R-100 fraction of bamboo pulp showing considerable reduction in fibrillation and inter-fiber bonding than seen in the R-14 fraction. 800x.

Other Bamboo Elements

The presence of vessel elements was not prominent although the amount noticeably increased from R-14 to R-100 (Figures 24, 25). The presence of parenchyma was a dominant factor, being minor in the R-14 fraction (Figure 24), but increasing from R-28 to R-100 (Figure 25). The presence of parenchyma in R-48, R-100 and whole pulp gives a compact sheet, as indicated by low bulk values (Figure 13).

The sharp decrease in the freeness of the whole pulp compared to the other fractions may be attributed to the large percentage of parenchyma present, giving drainage resistance without noticeable strength contribution. This is also supported by the fact that the fraction passing through 100 mesh screen developed very little strength on refining (Table III).

Conclusion

1 In the *unrefined condition*, paper made from whole pulp was generally superior in strength properties to paper made from the sized fractions. Within the fractions, paper made from the shorter fiber fractions showed higher strength than from the longer fractions.

2 On *refining*, the paper from the longest fraction developed the highest strength, The whole pulp strength was similar to the fraction retained on 48 mesh screen (1.55 mm fiber length) throughout the freeness range investigated.

3. The fiber length or the ratio of the length to wall thickness (or fiber diameter) correlated satisfactorily with paper strength on refining. Burst, tensile, stretch and tensile energy absorption all indicated a linear relationship to these parameters. Tear and fold were highly dependent on fiber length. All the strength properties were also found to be dependent on individual fiber strength as measured by zero-span tensile strength,

4 The amount of parenchyma cells increased from the longest fiber fraction to the finest one. Although the parenchyma cells appeared beneficial in paper from unrefined and slightly refined pulp possibly by improving inter-fiber bonding, they did not add strength to the refined pulp sheets. The whole pulp which had the highest percentage of parenchyma cells showed overall lower strength for its weighted average fiber length.

5 The major effect of refining was to increase inter-fiber bonding (and thereby the strength) by fibrillating the secondary fiber walls. Since the majority of fibers were thick-walled, the increase in flexibility of the fibers on refining remained secondary in importance. The longer fiber fractions had better fibrillating charactristics and hence better bonding characteristics than the shorter fractions.

TABLE III

Strength Properties of Fraction P–100 Passing Through 100 MESH Screen on Refining*

Refining Time (Sec)	Bulk (cm^3/gm.)	Burst Factor	Tear Factor	Tensile Breaking Length (m)
0	2.01	1.7	11	765
20	2.01	2.0	10	733
30	1.96	1.9	15	906

*Freeness measurements could not be made because the pulp was too fine.

APPENDIX A
Cooking Conditions and Yield for the Commercially Cooked Bamboo Pulp

Active alkali	19%
Sulfidity	21%
Liquor to chips ratio	2·5
Temperature	165°C
Time to temperature	1 hr. 45 min.
Time at temperature	1 hr.
Permanganate number	25
Yield	49–50%

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