Effect of beating on the cell mechanics of the individual bamboo fibre

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SUMMARY

The microfibrillar orientation in the different regions of the cell wall and its influence in the swelling and fibrillation (both internal and external) characteristics of the individual fibre upon beating has been discussed. Based on the fairly established theories on the cell wall mechanics of wood fibres and the elasticity constants proposed for the amorphous matrix and the embedded crystalline frame work by Mark(5) it has been theorized that the individual fibre has both flexible and rigid regions along its axis depending upon the fibrillar orientation in the cell wall and is one of the criterion for the strength development of the final network. The cell wall machanics of the polylamellated bamboo fibre has been discussed and the applicability of the theories of wood fibre mechanics to bamboo fibres has been shown. The anomoly in the the strength development in bamboo fibres upon beating has been explained as due to the variability in fibrillar orientation of the secondary wall proper.

Beating is essentially a process which in principle does not differ from other mechanical action put upon the fibre. Considering this, better understanding of the material in question is of course of the greatest importance. Added to the fibre length and other fibre dimensions, the basic morphological features of the individual entity of the fibre are also important in explaining the beating phenomenon of the fibres. The assumptions (rather proven ones) proposed to explain the cell wall mechanics of the wood fibres has been taken into confidence, as can be applied to bamboo fibres with the required modifications.

ELEMENTARY FIBRIL

To explain the mechanisms of beating, apart from the fibre itself, the individual entity that constitute the fibre is also an important aspect to be considered. Though Hanna[1] could support his concept on the existence of sub-elementary fibrils in certain regions of the cell wall of the fibre, the ultimate individual entity of the fibre should be the elementary fibril because it is the final micro structure that should possibly be obtained morphologically through mechanical means[2,3]. Fengels[4] definition of fibre as a fibrillar bundle formed by the aggregation of microfibrils, which in itself is the clustur of elementary fibrils, hold good for all the fibres whether it is a wood fibre or a bamboo fibre. These fibrillar bundles lie as layers along the fibre axis in different angles, to form thedifferent *"Cell Walls"* of the fibres. So the orientation of the elementary fibril along the fibre axis, and those influencing factors which bring forth changes on it are the essential features to be considered upon to explain the beating theory.

CELL WALL MECHANICS OF WOOD FIBRES

Elementary fibrils are assumed upon to be perfectly crystalline with the crystal axis in line with the fibril axis and that these crystallites are embedded on an amorphous matrix of hemicelluloses[5]. Since the definition of fibre[4] makes the elementary fibril as the individual entity and that the macrofibril is nothing but a randomnly oriented and distributed microfibril network, it is appropriate to say that the

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elementary fibril lie in different angles in the microfibril and a three dimensional structure of fibre ensues. The distribution pattern of the force along the fibre axis is therefore different. This prompts to say that the fibre with its different cell wall construction and fibrillar orientation in them is like a beam having a differential structural features along its axis. The theory of solid mechanics, that govern the force distribution pattern in the beams and rods should apply unreservedly in predicting the behaviour of fibres under stress. The behaviour of microfibrils should then fall within the limits of Hook's law of elasticity and the force distribution along the different cell wall regions can be calculated using appropriate tensorial equations meant for soild mechanics of the anisotropic elasticity.

Mark[5] evaluated the elastic constants for the microfibril frame work and the amporphous regions. The elastic constants for the crystalline arrangement in the frame work has been given both for their normal and parallel orientation to the fibrillar axis *(APPENDIX 1)*. These proposed values of Mark[5] which are for the soft wood tracheids should apply fairly good to bamboo fibres too, because these are the absolute values calculated for a defined geometrical network based on proven assumptions. Deviations if any can be evaluated mathematically.

CELL WALL STRUCTURE

Four Concentric layers have been recognized in wood fibre viz. the primary wall (P), the outer(S_1), middle(S_2) and inner secondary(S_3) or tertiary wall(T).

a) Primary Wall (P) : The cellulosic fibrils in the primary wall is highly individualized and somewhat randomly dispersed and to an extent interwoven. It is very thin ($\sim 0.5 \mu m$) having only a very small amount of crystalline cellulosic microfibrils. Mark[5] has assumed a transverse orientation for this wall. Very little is known about its chemical composition and specially about the distribution of hemicelluloses. For sulfite pulps, acid hydrolysis of non cellulosic carbohydrates results in the removal of most of the hemicelluloses and a weaker primary wall is obtained which is easily removed in the very early stages of beating. Furthermore, even if some hemicelluloses are present, their structural modification due to depolymerization would reseult in their loss of adhesiveness and bonding capacity. For Kraft Pulp the xylan stabilization and its deposition on the cellulosic chain during cooking makes the primary wall intact during mechanical action and if it comes out, it comes as a thin sheet[6,7] (Figure A)[7].

b. Secondary Wall : The bulk of the fibre is formed as the secondary wall which grows by apposition and hence appears to be trilamellated.

i) Outer Secondary Wall (S_1). Outer secondary wall is overlapped by the primary wall and is characterized by its close lateral packing and consequent parallel alignment of fibrils[7]. The cross layered fibrillar structure proposed by Hedge and Wardrop[8] has been confirmed by others[9,10]. Emerton and Goldsmith[11] has shown that S_1 seems to contain two counter rotating sets of stiarations, that were symmeterically disposed. This has later been confirmed by Frei et al[12]. The angles between the tangent to the spiralling fibrils and the axial direction

APPENDIX 1

ELASTIC CONSTANTS USED BY MARK (5)

Modulus of Elasticity of cystal in chain direction (EFL) (F Frame work)	1.34×10^{12} dyne/m^2
Modulus of Elasticity, normal to chains, average for (101) and $(10\bar{1})$ planes. (EFT)	0.272×10^{12} dyne/m^2
Shear modulus of rigidity, average for (101) and $(10\bar{1})$ planes (GFLT)	0.044×10^{12} dyne/m^2
Poisson's ratio of Contraction in the normal direction due to extension in chain direction. (FLT)	0.10
Poisson's ratio of Contraction in chain direction, due to extension in normal direction. (FTL)	0.011
Modulus for amorphous matrix (EM)	0.02×10^{12} dyne/m^2
Poisson's ratio for matrix (M)	0.3
$G_M = E_M/2\,(1+M)$	0.0078×10^{12} dyne/m^2

L = Longtitudinal
T = Transverse

Fig. A. Swelling of a fibre (*Pinus radiata D. Don*) The primary wall bursts. It forms a sheet and is still attached to the fibre at certain points. The outer layer of the secondary wall constricts the swelling wall and produces balloon swelling of the middle secondary wall[7].

(*helix angle*) for the wood fibres vary from $\pm 35°$ to $\pm 55°$ to $\pm 80°$[5,13]. The outer secondary wall is fairly thinner than the primary wall (0.1-0.3μm).

ii) Middle Secodary Wall (S_2). The wall which is laid down as a coaxial lamellae (cf : outer secondary wall) contains the bulk of the microfibrils and hence crystalline cellulose. The fibrils of S_2, like those of S_1 are highly parallelised and the stiarations are oriented steeper to the cell wall axis and varies from 0°-50°. Mark[5] has reported a variation in the helical angle of the S_2 layer in the tangential and radial wall filament in softwood tracheid and the helical angle is steeper in the former (20° and 36° respectively). The S_2 layer is the thickest of all the layers in the cell wall (10-30μm).

iii) Inner Socondary Wall (S_3). The structure of the S_3 wall is extensively studied[14], which like primary and outer secondary wall is very thin and highly crystalline. The spiral angle of the S_3 wall is a wide varying one depending on the wood species. The rotation of the spiral is of the 'S' form.

FORCE DISTRIBUTION ACROSS THE CELL WALL

Mark[5] pointed out that the fibril bundles wound around the axis in a variety of angles and the real difficulty lie in estimating their specific orientation. Also he has assumed that the crystalline orientation on the axis is same as that of the microfibrils. He has shown from the calculated elasticity constants that in a fibre where the crystalline frame work is embedded in the amorphous matrix as parallel strings (Figure 1, 2), the frame work will take the maximum load (*APPENDIX I*).

Though there are many ways of load distridution on a structure consisting different helical windings the important aspect is how each one of these helical windings will radially move under a axial stress. Because of the very high modulus of elasticity of the crystalline fibrillar (*Helical*) orientation parallel to the chain and low elasticity and poissions contraction ratio for the amorphous matrix(*APPENDIX-I*) the following behaviour could be postulated.

i) The modulus of elasticity will result in a poor dynamic respcnse by the amorphous matrix compared to the crystalline cellulose and the matrix, low shear modulus of rigidity will make it to collapase during an applied force.

ii) The helical angle of the fibril in the individual walls of the fibre determines the rigidity or the flexibility of the respective region. The orientation of the fibrils parallel to the fibre axis give them the maximum possible flexibility and the orientation normal to the fibre axis give the maximum possible rigidity. For the intermediate orientation the force distribution is the vector resultant, resolved for the normal and parallel distributed load (Figure 1).

iii) The steeper the helical angle between the axis and the coil direction, more will be the radial shrinkage, when stretched, than do the flatter helices of greater coil angle From the figure 1,

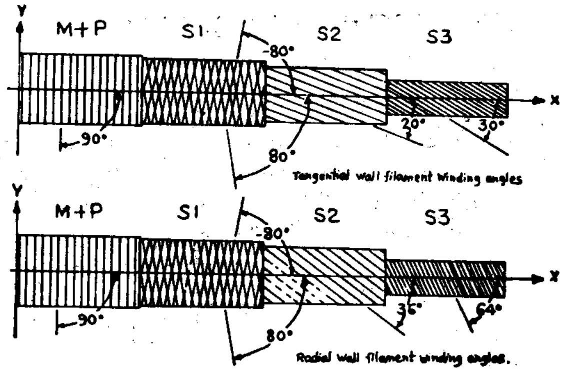

Fig. 1. Winding angles of tracheid microfibrils used by Mark.

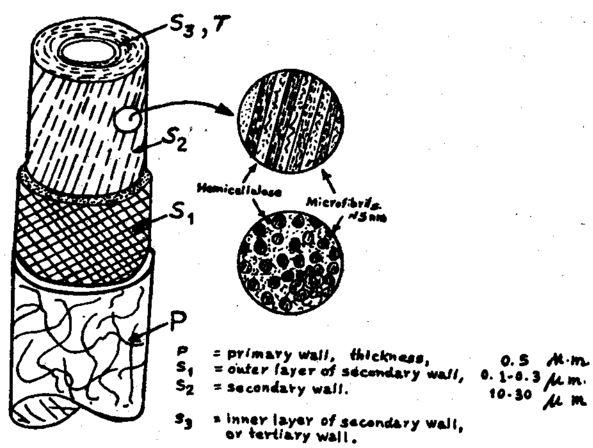

Fig. 2. The structure of a softwood tracheid fiber.

it is evident that because of the steeper helical angle of S_2 than the S_1 layer, and also the cross woven structure of S_1 layer, the radial shrinkage is more in S_2 than in S_1 and there will be a tendency for separation between the two. Thus failure is likely to occur between the interface of S_1 and S_2 layers.

The primary wall with its fibrillar orientation normal to the fibre axis is isotropic from a mechanical point of view. The high rigidity (modulus of elasticity is 0.272×10^{12} dynes/m^2) and lower thickness (below 0.5μm) makes this particular section rigid in character. Added to this, any applied force to the fibre axis will be an uniform distributive load, like placing the load at the centre of the beam. The rigidity of the primary wall will make it to take the load less frequently: The lower co-efficient of contraction will make this part of the fibrillar network to resist any mechanical action on it. The region is rigid, brittle and will not swell and if a strong mechanical action is but upon, it will come out of the fibre as a thin sheath (figure A)[7]. Any fibrilation of this primary wall produces fines which are very difficult to activate. Though it appears that this portion of the fibre wall is difficult to remove, in practice this is the region which is stripped off at the very early stage of beating.

The S and Z type helical windings of S_1 layers give it an interwoven structure. The cross lying orientation of microfibril makes that any applied force in one of the helices will be opposed by its counterpart with an equal but oppsite force. This acts as an opposing vector force of equal magnitude and hence the resultant work done on this region is zero. (It is not so simple as predicted due to the three dimensional aspect of the underlying fibre and the fibrillar orientation). The S layer because of this interwoven structure will contract or expand very little and also this cross woven structure gives less water accesibility to the underlying lamella and is a constraint to swelling. The S_1 layer remains with the fibre till such time a very severe mechanical action is applied on the fibre.

The middle secondary wall (S_2) is anisotropic because of its helical orientation. The fibrills can shrink longitudinally but rather week laterally. Swelling therefore takes place laterally and any mechanical action causes fibrillation. The S_2 layer is thicker (10–30μm) and with a high modulus of elasticity, is flexible, behaves like a sponge whereby the applied load is taken up at frequent intervals in small but highly effective pulses. During the course of the beating this region becomes softer. The rigid primary wall and the water inaccessible (hence swelling restricting S_1 makes the middle secondary wall S_2, the region of fibre break down. After the removal of S_1, middle secondary wall S_2 collapses, coming out as highly fibrillated ribbons.

INTERNAL FIBRILLATION

During beating, the fibre is not only subjected to mechanical action but also to the stresses of water. So the constraints at P and S_1 layer initiates the breakage of interfibrillar bonds between the celluloses and the hemicelluloses (internal fibrillation). None of the cell walls of the fibre is a solid block but layered regions. The building material between them may be water (through hydrogen bonds) or hemicelluloses. When fibres are immersed in water, they imbibe and swell when beaten The swelling is is a *"limited swellirg"* because of the constraining crystaline cellulose and the partly crystallined hemicelluloses. A weak force is enough to break the bonds in the already swollen gel which is under stress.

EXTERNAL FIBRILLATION

The swollen fibre has subjected to all the mechanics as described already. When beating proceeds or when the fibre is exposed to extra strong mechanical action, the fibrillar layer of the middle secondary wall splits in the longitudinal direction to form fibrils external fibrilation).

The swelling pressure of the interfibrillar substance is an imp rtant aspect to consider upon in discussing the mechanism of the fibrillation. The lateral expansion and the consequent longitudinal split up indicates that the fibre explodes only after a thorough internal fibrillation. So for fibrillation, a certain advanced swelling is necessary. Moreover, the maximum the swelling and more softer the material the maximum fibrillation is achieved. This sort of advanced swelling can take place only in the regions of steeper helical winding and higher hemicelluloses concentration. The concentration of hemicelluloses decrease from primary wall (P) to the inner secondary wall S_3). This may prompt to say that the maximum swelling would take place at the S_1 layer. But it is not so, because, it is primarily dependent on the accessibility of water which in turn is affected by the fibrillar orientation (poor swelling of S_1 because of interwoven fibrillar structure).

BAMBOO FIBRES

Having explained the cell wall mechanics of the wood fibres to an applied load, the necessary logic to counter upon is, can these theories proposed for the wood fibres be extended to explain the morphological changes of the bamboo fibres during beating ?

Purkayastha *et al*[15] has taken the polylamellate structure of the thick walled bamboo fibre proposed by Parameswaran and Liese[16] for studying the changes in the morphological characters of bamboo upon beating. Krishnagopalan[17], has shown that bamboo has both thin and thick walled fibres in its vascular bundles Referring the micrographs (Figures B,C)[17] the polylamellate fibre wall structure of both thin and thick walled fibre is evident.

Unlike the wood fibres where there is defined primary, trisecondary walls, the bamboo has a polylamellated structure[16] consisting of alternative broad and narrow lamellae having different fibrillar orientation in its secondary wall (Figure 3). The

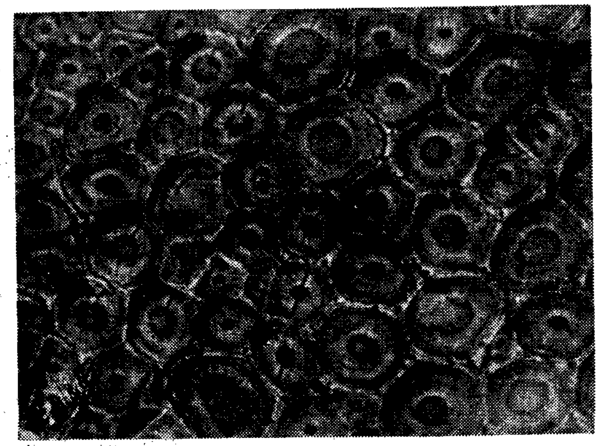

Fig. B. Microtome section of portion of Fibro-vascular bundle in Bamboo, showing variation of cell wall layering in fibres[17]. (Magnification 870 ×)

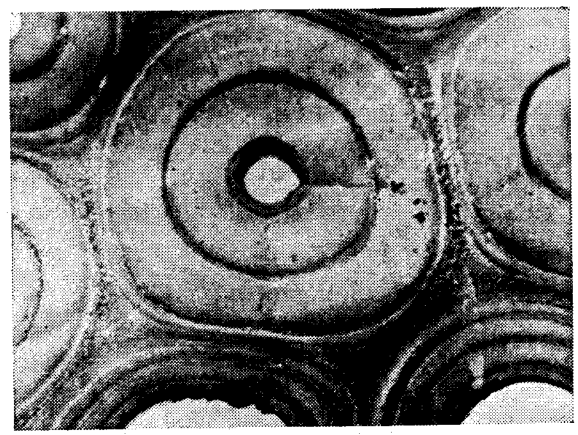

Fig. C. Ultra-thin cross section of Bamboo showing variation in cell wall layering of Fibres[17]. (Magnification 5800×)

number of such layers vary with the species. Parameswaran aud Liese[16] has reported 18 layer structure for *Oxystenanthera abyssinica* whereas Tono and Ono[18] has reported 7 to 9 layers for *Phyllostachys*. The primary wall has an interwoven structure (cf. outer secondary wall of the wood fibre) whereas the outer secondary wall has an inclined orientation to the fibre axis (50°) (figure 3).

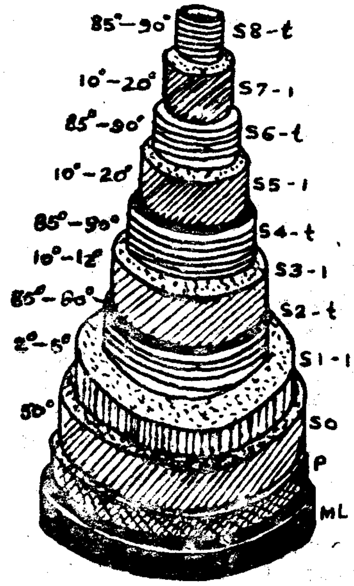

Fig. 3. Model of the polylamellate structure of a thick-walled bamboo fibre. (after Parameswaran and Liese 1976).

(×95)

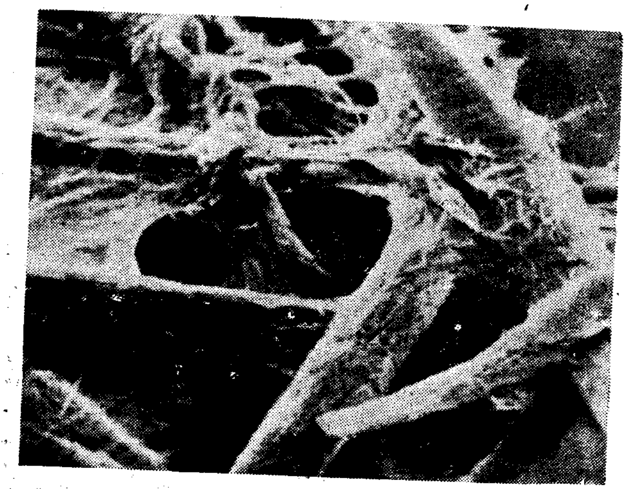

Fig. D Hand sheet surface of well refined (270 ml CSF) Bamboo pulp showing the intactness of the primary wall (Right hand Top and bottom and left middle) reduced fibrillation and inter-fibrc bonding[17]

(Magnification 1000×)

In bamboo fibres the secondary walls that immediately succeeding the outer secondary wall has a diversity unlike that of wood fibres. (The whole layers of the secondary wall succeeding the outer secondary wall is designated the nomenclature "Secondary Wall Proper" and used in the discussions).

The crystalline orientation of the primary wall as given by Parameswaran and Liese[16] has a structure which is the very domain of the outer secondary wall in the wood fibres. The interwoven structure of this region like in wood, would have resultant work done on is zero (Opposing vecter forces). This layer is very difficult to remove by simple mechanical action. The primary wall tries to remain intact (Figure D)[17]. Also the interwoven structure gives it a higher water inaccessibillity. The primary wall is much more complex in bamboo than in the wood fibre because of the partly crystallined hemicelluloses, mostly stabilized and redeposited xylans. So the dimensional changes if any and as well the swelling is restricted for this region. Due to this the swelling of the bamboo fibres may be effected only after a long time of beating compared to wood fibres, in which the primarywall is removed during very early stage of beating. But this restrict swelling because of the structural features of the primary wall initiates a higher internal breakage between the interfibrillar bonds of the outer secondary and secondary wall proper. Once the primary wall is removed the swelling achieved is maximum at the outer secondary wall because of the inclined fibrillar orientation The beating therefore may proceed faster, achieving a maximum external fibrillation. The stabilized hemicelluloses present in the regions because of their structural changes, as already pointed out, lost their adhesive and bonding capacity. The primary wall even if it comes out, comes as a sheet (Figure E)[17]. The fines obtained by the fibrillation of the primary wall results in the formation of crills which could not be activated by surface tension forces and could not take part in any of the consolidation activities. So the strength property development in bamboo may be slow unlike the wood fibres where the strength development starts at the early stages of beating[19,20].

The outer secondary wall has a feature similiar to that of the inner secondary wall of the typical tracheid[5]. The difference being in the hemicelluloses content which is quite high in case of a bamboo fibre. The fibrillar angle is steeper (50) to the axis and have an intermediate flexible velues (resultant vector). Once the primary wall is removed the outer secondary wall swells better due to high hemicelluloses content and flexibility. Greater swelling and

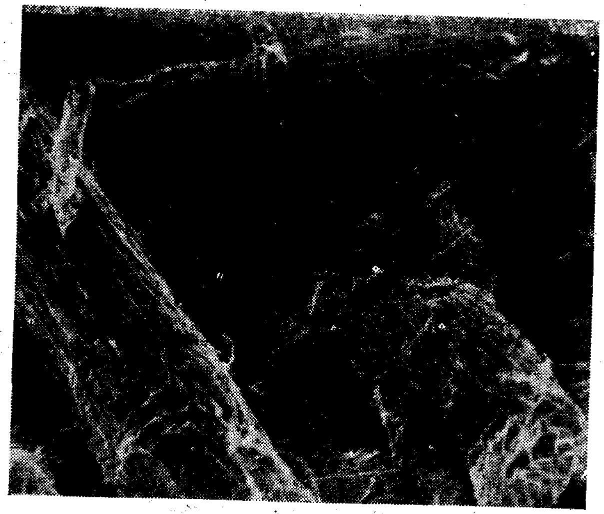

Fig. E. Hand sheet surface of refined Bamboo pulp showing perforated sheet type structure, possibilly due to primary wall material (lower centre of micrograph)[17] (Magnification 2000×)

flexibility results in a higher fibrillation of this wall. This evidently supports one of the findings of Krishnagopalan *et al*[21] that the fibrillation rather than the flexibility is the essential features of the beaten bamboo fibres It can be fairly said that the fines produced in the system, maximum of which comes out from the outer secondary wall fibrillation, is a contrary feature of bamboo fibres over wood fibres. So the fibrils or fines) produced from this outer secondary wall maybe actively taking part in the sheet consolidation.

The *"Secondary Wall Proper"* has alternating broad and narrow lamallae havig different fibrillar angle to the fibre axis (Figure 3). To distinguish these lamellae from those occuring in wood fibres, Parameswaran and Liese[16], have designated them as S1—l, S2—t, S3-l, S4—t, etc. The suffixes l and t indicates the orientation of the fibrils in the longitudinal and transversal directions respectively. The cellulose fibrils in the broad lamellae are oriented almost parallel to the long axis of the fibre (2°-20°) and if inclined, inclined steeply to the fibre axis, exhibiting only a slight increase in the angle from the outer region to the lumen The outermost layer is almost parallel (2°—5°) to the fibre axis. In all the narrow lamellae he fibrillar angle is normal to the fibre axis (85°—90°). Unlike the middle and inner secondary wall of the wood fibres where the fibrillar orientation is inclined to the fibre axis giving the fibres the necessary flexibility in these regions, in case of bamoo we have an alternating regions of very high flexibity and rigidity. The S2—t, S4—t, S6—t, S8—t,[S(2-8 t)]has the typical fibrillar arrangements as that of the primary wall of the wood fibres. This layers are highly rigid, resist all the mechanical action on them (due to their isotrophy and again will take an infrequent load, will distribute it uniformly along the region. The lateral movements of these S(2—8 t) regions are restricted due to the uniform load distribution. Added to this, the region is highly crystalline with a varying hemicelluloses content from S2—t to S8—t. So in addition to the rigidity, swelling constraints are also put upon This S2-t, S4—t & S6—t regions overlap the S3—l, S—5l and S7—l layers respectively. The S(3—7l) regions has a broader lamallae structure having parallel fibrillar orientation. The S(3—7l) has all the characteristics of the middle secondary wall of the wood fibres. This region is flexible, take frequent loads in small but effective pulses and during the course of the beating becomes softer. It has dimensional extension in the lateral direction because of the parallel fibrillar orientation.

Unlike the wood fibrils, the fibrils of bamboo has the following constrains for swelling and lateral movements (i) the intact primary wall, (ii) alternating narrower rigid structures of the *"Secondary Wall Proper."* Except the S1—l which has not been covered by the transverse narrower lamallae all other longitudinal fibrillar oriented regions S(3—7l) experience this. The S(1—l) layer swells more and fibrillate much easily. Likewise, the S(8—t) lamella may be collapsed during beating. The swelling constraints put upon the S(2—6t) brings out an effect which is peculiar to bamboo fibres only. As the beating proceeds the swelling constraints of the S(2—6t) regions result in the internal fibrillation of the S(3—7l) regions. The S(2—6t) regions infrequent loads take up and S(3—7l) regions flexibility and frequent load take up has a *"mechanical shock effect"* on these regions. At a particular point, the swollen (due to internal fibrillation) S(3-7l) regions burst open the S(2-6t) regions and comes out, producing lot of debris, the debris resulting from the S(2—6t) regions. Moreover the constraining factors of S(2-6t) layers and the swelling and fibrillation of S(3-7l) layers will have a *"pull and push"* effect, that the fibrils coming out of the S(3—7l) layers come as string (Figure F, G[17]) and consolidation of the network has to be effected by these strings (Figures F, GH)[17]. The debris produced from the S(2-6t) lamellae like the crills of the primary and outer secondary walls of the wood fibres are very difficult to activate because of their higher surface tension than the water in which they are dispersed during the net work formation.

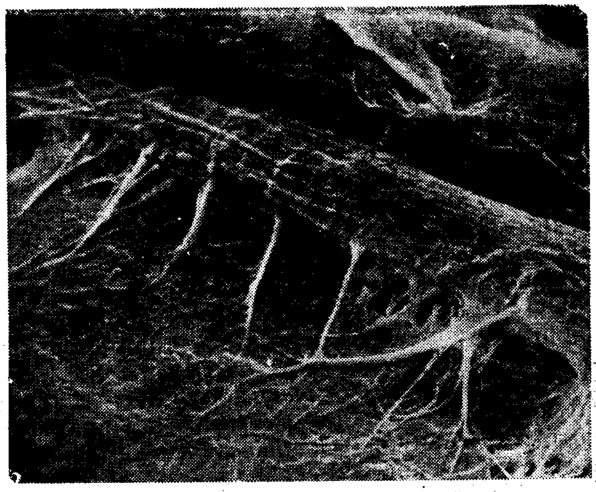

Fig. F. Hand sheet surface of refined Bamboo pulp showing formation of large string like Fibrillar bundles from secondary wall[17].

(Magnification 2000×)

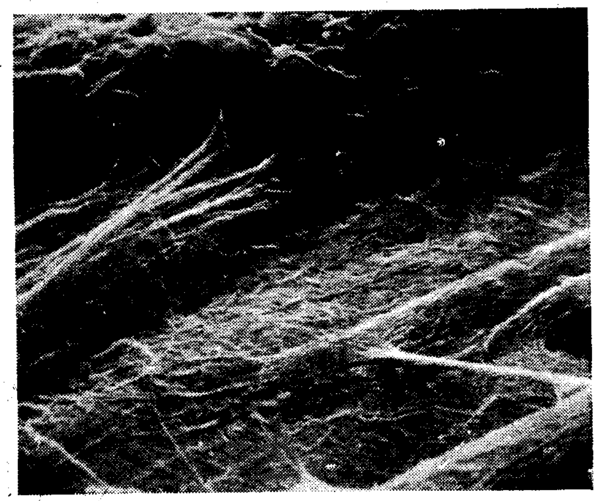

Fig. H Hand sheet surface of well refined (400 ml CSF) Bamboo pulp showing string like structure of secondary wall type bonding[17]

(Magnification 2000×)

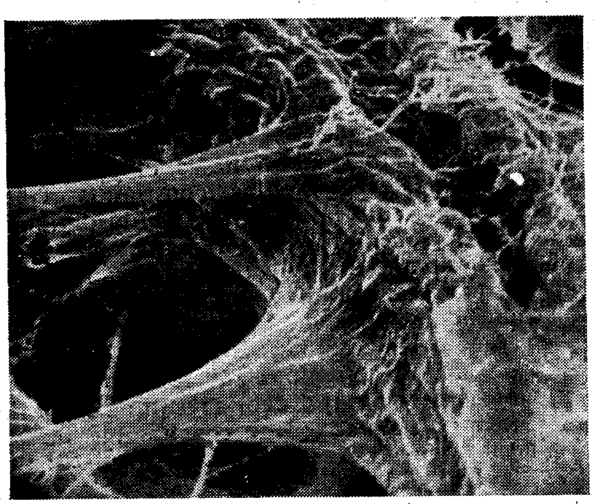

Fig. G. Hand sheet surface of refined Bamboo pulp showing removal of secondary wall layer and exposure of inner secondary wall material (Upper right of micrograph), large string-like bundles of Microfibrils present (left side of Micrograph)[17]

(Magnification 5000×)

Fig. I Hand sheet surface of refined Bamboo pulp showing perforated sheet formation (Right of centre of Micrograph) possibly due to primary wall material. Remaining smooth textured surface due to unaltered primary wall material lying on the fibre surface[17].

(Magnification 1000×)

So the situation for the bamboo fibre during beating is like this.

i) The primary wall tries to remain intact with the fibre unless otherwise a strong mechanical action is put upon and even if it comes out comes as a sheet. Fibrillation results in the formation of crills (Figures E I)[17].

ii) Outer secondary wall produces fines which are typical to that of the fines from the middle secondary wall of the wood fibre.

iii) The fibrillation of the broader lamellae and the rigidity and swelling constraints of the narrower lamellae of secondary wall proper has two effects

(a) debris from the narrower layer,

(b) string like fibrills from the broader layer (Figures F, G, H)[17].

Annergran *et al*[22] commented that bamboo which resembles hardwoods in chemical composition, has an average fibre length approaching that of the softwoods. It has a tensile strength lower than softwoods but its tear strength is higher compared to the hardwoods. The reason for bamboo pulp not reaching a higher tensile strength needs to be explained.

Based on the above discussion, the authors feel that this anomoly of bamboo fibres can adequately be explained. A lot of debris from the secondary wall proper (S_{2-t},S_{4-t} and S_{3-t}) during the burst opening of the fibrils from $S_{(3-7l)}$ and the crills formed by the primary wall, gives the system a larger amount of fines than other wood species[23,24]. But unlike the fines from beaten wood fibres this fines and crills are stiffer and difficult to activate. Also the intactness of the primary wall (Figure I)[17] again makes the consolidation a difficult process, compared to wood fibres specially to the tracheids. Moreover the network bulid up has to be done by the string like fibrills of the $S_{(3-7l)}$ layers (Figure F,G,H)[17]. The only fines that could be activated are result from the outer secondary wall and again from the outer region of the secondary wall proper $S_{(1-l)}$.

So the fibre network (sheet) formed from the beaten bamboo fibres when loaded from mechanical point of view) have the following typical characteristics.

i) The interfibre bond slippage is more pronounced resulting in the higher Tear Index than hardwoods (because of the poor consolidation of the fibres resulting in less RBA).

ii) The fines and crills of the system are stiffer, rsulting poor activation and initiating an earlier bond slippage. The consolidation between the fibres are reduced because of the primary wall intactness. So the active segments that take part in the loading is less (RBA is less), resulting a lower Tensile Index.

iii) Unlike the wood fibres, the strength property development will be achieved only in the later stages of beating.

One important evidence derived from this study is that Mark's[5] cell wall mechanics, as it holds good for the wood fibres, could be applied to bamboo fibres too, as the theory is dependent on the fibrillar orientation of the micro/elementary fibrils on the cell wall, whether it is a four walled wood fibre or poly lamelleted bamboo fibre. For two fibres of the same fibre length and to an extent of same chemical composition, the development of strength during beating and in final network may be different if they have different fibrillar orientation in these cell walls.

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