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Use of Modern Microscopes in Analysing Fiber and Paper Properties (II)

- New Aspect in Fibrillation of Pulp Fibers during Refining -

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ABSTRACT

The CLSM and the image analysis technique enhanced observation of fiber wall fibrillation occurred in both the outer and the inner fiber wall surfaces during refining by non-destructive techniques. In the early stages of refining, it was well observed that a partial separation between the S1 and S2 layer in the secondary wall was made generating a space in the wet fiber walls. With further refining, it was clearly shown that the shear forces imparted by the refiner bar surfaces caused the S1 layer to become totally separated from the S2 layer as well as creating microfibrils. Furthermore, the fibrillation in the inner fiber wall surfaces could be due to the normal force (F_n) by refiner bars, friction force between a fiber and refiner bars (F_s) and inner friction force between fiber walls (f_s). It was confirmed that the concept of fibrillation should be extended to fibrillation in the inner fiber wall surfaces as well as internal and external fibrillation.

1. Introduction

One of the most important operations in the papermaking process is stock preparation, beating or refining, since ultimate production and paper characteristics are essentially determined at this stage. It is, therefore, vitally important to be able to characterize beating and refining effects especially in relation to pulp fiber properties. The classical and most frequently used way of characterizing the result of beating is to measure the wetness (°SR) or freeness (CSF) of the refined pulp fibers and to produce laboratory sheets, from which certain physical properties are measured. Such an approach in beating research is generally not sufficient because the resultant data from measurement of wetness or physical properties are closely associated with various primary effects, such as fiber shortening, external and internal fibrillation, induced in the refined fibers. Thus it is hard to say that such indirect methods actually characterize individual beating effects.

The direct way of studying the refining effects on pulp fibers is to make photomicrographs of the beaten fibers. Since conventional microscopy like light microscopy (LM) and scanning electron microscopy

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(SEM) use dried fibers instead of wet fibers, fiber collapse or distortion during dehydration possibly occurs to varying degrees. Consequently, observing the dried fibers does not give information about the interaction of fibers with water nor about the actual changes that have been induced in the fibers as a result of the beating or refining operation. Thus it is vital to preserve the changed wet structure of pulp fibers morphologically during mechanical actions, like refining, since the papermaking process is carried out almost entirely in water. In particular, the opportunity given by microscopes to look at the hydrated fibrous material considerably enhances our understanding both of what has happened to the pulp fibers during refining and of the meaning of other test results.

However, as described in the previous papers,[1), 2), 3)] the CLSM made it possible to observe refining effects on individual pulp fibers in a natural state. It was also used to disclose refining sequence of pulp fibers including detachment of S1 layers, separation of S2 layers and removal of parts from the separated S2 layers during refining. Moreover, in combination with image analysis techniques, the most important thing done with the CLSM was to quantify internal fibrillation of beaten fibers. Finally it can be said that the CLSM opened a new way on refining study.

For observation of external fibrillation by the microscopes, most of works have been focusing on the fibrils on refined fiber surface due to limitation of microscopic capability. The micro- or macrofibrils might have great propensity to bond back to the parental fibers during drying. Thus this behavior of dry fibrils makes us difficult to understand what it happens during refining. Moss *et al.*[4)] showed that LTSEM (Low Temperature SEM) was an excellent tool in investigating external fibrils of hydrated fibers, but they recognized artifacts arising from extracted carbohydrates around microfibrils. On the other hand, the optical sectioning technique of the CLSM might be able to make our understanding on external fibrillation of wet fibers deeper than ever.

On this work, when wet fibers are observed with the CLSM, it is studied whether or not there is a new aspect on external fibrillation other than the information obtained from the conventional microscope.

2. Materials and Methods

Canadian Howesound softwood bleached kraft pulp was beaten by the Valley beater following TAPPI T 248 standard method. The pulp fibers were beaten to 20 ° SR.

The confocal microscope used in observing morphological changes by refining was a Bio-Rad MRC-1024 confocal laser scanning microscope attached to a NIKON Axiophot microscope in the National Instrumentation Center for Environmental Management at Seoul National University, KOREA. All observations were made with ×20, 40 and 100 oil immersion Plan-Apochromatic objectives with a numerical aperture of 0.5, 0.75 and 1.3 respectively.

Acridine Orange was chosen for dying a bleached softwood kraft pulp due to its bright fluorescent signal at the 488 nm of the excitation Argon laser. Most results presented here were for fibers dyed at room temperature in a very dilute solution (0.01 g dye diluted in 1 L distilled water = 10 μm/mL). The dyed fibers were thoroughly washed to remove excess dye. After the wet fibers were mounted on the microscope slide, they were covered with #1 cover slips (0.169 mm thick) which had a similar refractive index ($n \approx 1.5$) to immersion oil and pulp fibers.

The KS 400 (version 3.0, Carl Zeiss) as an

(a) Disintegrated fibers

(b) Refined fibers

Figure 1. CLSM images of wet fibers: left image - ×400, right image - ×1,000 (scale bar unit: μm).

image analysis software was used to analyze optical sections acquired by the CLSM.

3. Results and Discussion

3.1 External fibrillation observed by CLSM

As mentioned earlier, external fibrillation was regarded as one of the most important effects of refining. When observing dried fibers on the microscope stage, many of the fibrils and microfibrils present in a wet state were possibly not observed due to their propensity to dry back onto the parental fiber. It is extremely difficult, once the fibers have dried, to see any of the external fibrillation that might have been present. Even rewetting did not allow these fibrils to become visible again as entities.

However, CLSM made it possible to observe, in a wet state, fibrils attached to refined fibers, as shown in Fig.1. First of all, it is worthwhile comparing the images from swollen fibers (Fig. 1) with the images from dried ones (Fig. 2). As expected, there is no striking appearance of fibrillar material on the surface of disintegrated fibers (see Fig. 1). They are intact and well defined in shape and appearance, being imaged in a highly hydrated state. However, external fibrils of the refined fibers (see Fig. 1(b)) become more evident and more clearly visible. It can be seen that the fibrils are wound around the fiber, indicating that the external fibrils mostly exist as long bundles of cellulose microfibrils, namely, macrofibrils. At the same time, it can be seen that the S1 layer has separated from the S2 layer. With further beating, the S1 layer would be removed and become a component of the secondary fines present in the pulp. When looking at images of the dry state compared with the wet state, it is far more difficult to distinguish the separated S1 and S2 layers (see Fig. 2). This is because the gaps between the S1 and S2 layers disappeared after drying. Thus it can be said that the CLSM makes it easier for us to observe any separations that might exist between the S1 and the S2 layers of beaten fibers in a hydrated state.

Considering the magnified image in Fig. 2(b), the cellulose microfibrils in the swollen cell wall appeared like a fuzz of hair around the beaten fibers. Higgins *et al.*[5] coined the fuzzy fibril theory merely indicating that the image was indistinct (where the word fuzzy was usually applied) but that the fibrils appeared like a fuzz of hair around the fiber in question. The microfibrils exhibit rather long and thin microfibrils (marked by ⇨) that might be peeled off from the secondary wall during refining. At the same time, with external fibrillation during refining, the secondary wall was exposed at the fiber surface and the fibril alignment in the S2 layer is seen in the image. It is interesting to note

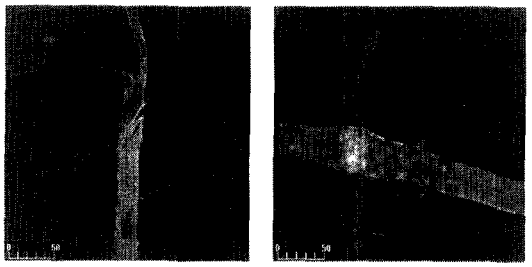

Figure 2. CLSM images to show (a) external fibrils of a dry fiber and (b) inter-fiber bonding created at the crossing fibers (scale bar unit: μm).

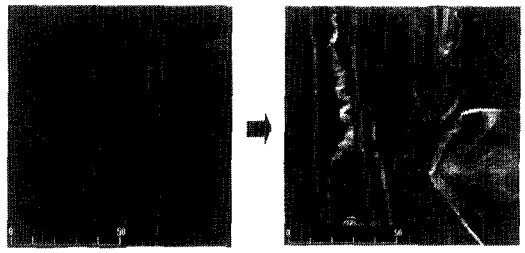

Figure 3. CLSM images demonstrating external fibrillation during refining (scale bar: μm).

that the external fibrils of the swollen fibers mostly exist in the shape and size of fluffy microfibrils rather than macrofibrils. It is well understood that such fibrils will contribute to drawing of the neighbouring fibers or the upper fibers in a fiber network together encouraging inter-fiber bonding during drying. However, it could be clearly seen by reference to Fig. 2(a) that drying processes forced microfibrils to flocculate and subsequently to form thin film-like webs. Such external fibrils generated during refining are found to form bridges (marked by ⇨), thus contributing to inter-fiber bonding during drying, as shown in Fig. 2(b). That is, as water is removed from the fiber network during drying, a strong adhesion system - which seems to be accentuated by the presence of the microfibrils - will be created by the increase in Campbell's force, leading to improvements in paper strength. Finally, it is almost impossible with conventional microscopes, to study bonding sequences in particular how fibrils between neighboring fibers bond to each other during drying - using dry specimens.

A procedure leading to the generating of external fibrils during refining is displayed in Fig. 3. In the early stages of refining, a partial separation between the S1 and S2 layer in the secondary wall occurred forming a space between two layers, as displayed within a rectangular box and in a circle in Fig. 3(a). This indicates that the early stages of beating condition the fiber surfaces to be ready for later ease of removal by further imposed shear stresses. With further refining, it is observed that the shear forces imparted by the refiner bar surfaces caused the S1 layer to become totally separated from the S2 layer as well as creating microfibrils, as displayed in the rectangular box of Fig. 3(b). It is also noted that the S1 layers are completely removed during the advanced stages of refining. The macrofibrils shown inside the circle in Fig. 3(b) indicated that even a part of S2 layer became detached from the secondary walls. It was proposed that such microfibrils or macrofibrils from the S2 layers could also be a source of the secondary fines as refining progresses.

Interestingly, from the images obtained from 33~36 μm inclusive from the base of the fiber, there was something else happening in the inner fiber walls (lumen walls). It could be clearly seen that refining caused the inner wall surfaces of the fiber to become fibrillated, much like external fibrillation but within the space of the central lumen. Even if the length of fibrils of the inner walls was shorter than those on the outer walls of fibers, the microfibrils

(marked by ⇨) were clearly visible in the CLSM sections.

3.2 Fibrillation characterized by the CLSM and the image analysis

Using the image on the right in Fig. 1(b), an optical sectioning technique was applied with a constant stepping distance, 1 μm, using the CLSM. The optical sections from the CLSM generated a total of 60 slices from one single fiber, and a newly created image showing external fibrillation of a swollen fiber was classified in Fig. 4.

The fibrils observed in the lumen wall were confirmed using the KS 400 3D software. A three-dimensional function of the KS 400 was used in order to clarify that the

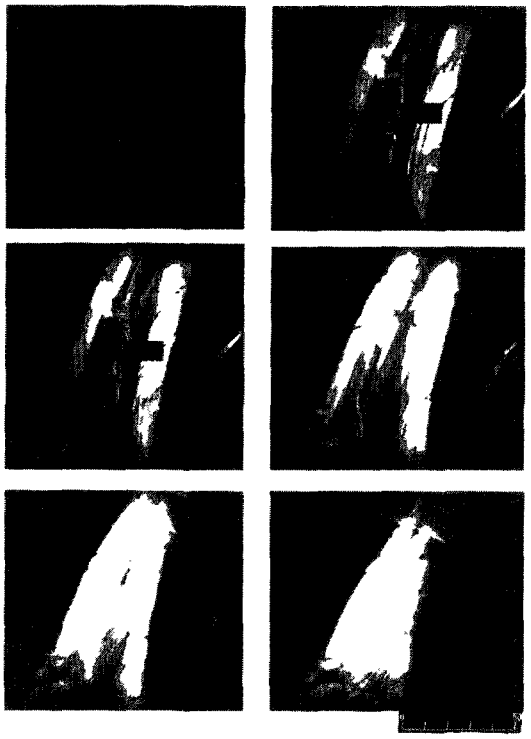

Figure 4. Optical sections of the swollen fiber with a stepping distance of 1 μm (scale bar unit: μm).

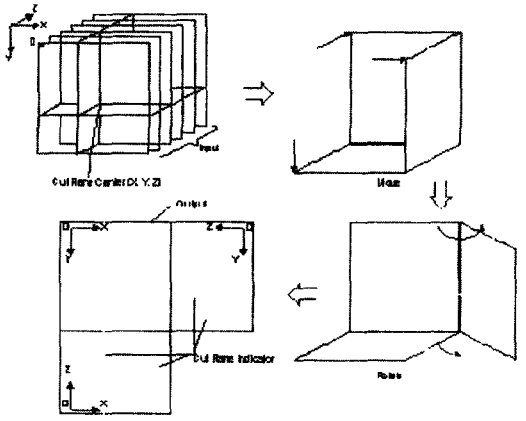

Figure 5. A schematic diagram showing a cut main plane function in KS 400.[6]

fuzzy shaped fibrils were created from the inner wall surfaces during refining. The cut main plane function of the 3D functions generates an image sequence that contains the slices of all three main planes, as shown in Fig. 5. Three slices were cut through the input image sequences (here 60 slices obtained in Fig. 4) along the main planes and displayed in composed form in a resulting image sequence. In Fig. 6, the dotted lines on the image show the position of the intersection as a set of cross-hairs and, in the position display of the lower right quadrant, the current coordinates (the 33rd of 60 slices in Fig. 4) of the intersection are shown. The horizontal section of the lower left portion shows that the lumen in the fibers was partly filled with the fibrils created from the inner wall of the fiber. The vertical section of the upper right portion more clearly disclosed the existence of the fibrillation occurring in the inner fiber wall surfaces. This did mean that external fibrillation occurred to both the outer and inner walls together during refining. The indications are, though, that the involved beating mechanism is not only relying on direct physical contact of the beating bars but also relies on the contact of cellulose surface with cellulose surface,

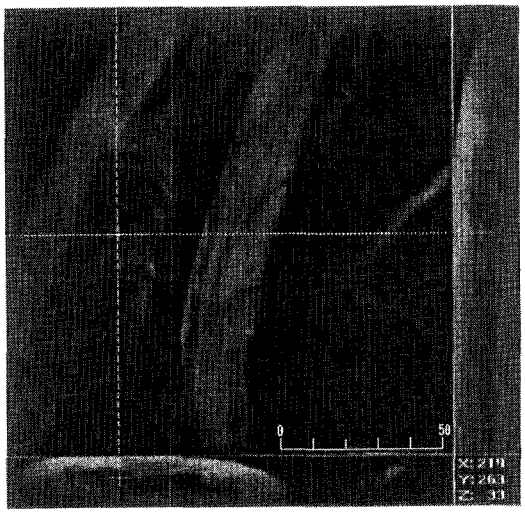

Figure 6. KS 400 image showing fibrillation generated from the inner wall (lumen wall) of fiber cross-sections (scale bar unit: μm).

shear, flexing and so on.

Fig. 7 demonstrates (a) a schematic diagram of a fiber under compression between refiner bars and (b) the forces acting on the fiber.This diagram explains how outer and inner walls of fibers are externally fibrillated during refining. The forces acting on fibers during refining can be classified into three: a normal force - by refiner bars, F_n, a surface friction force between the refiner bars and fibers, F_s, and an inner friction force between inner fiber walls, f_s. When the fibers pass through the gap between refiner bars, they are compressed by the normal force, F_n, which contributes to the work applied to the fibers in the direction of refiner bar motion. In the case of F_s, the force comes from the bar surface and therefore may act on a whole bar width. Since there are equal and opposite reaction forces from the opposing bars, the friction force, F_s, between refiner bars and fiber surfaces makes the fiber walls externally fibrillated. At the same time, the forces, F_n and F_s, can be a major factor in creating the inner friction force, f_s, generated by contact of the upper and the lower walls within the inner fiber walls. The force, f_s, may cause the inner fiber walls to become fibrillated, much like external fibrillation. It should be noted that both F_s and f_s depend upon the normal force, F_n. The diagram clearly shows how fibrillation in the inner fiber walls occurs.

The optical sectioning technique of the CLSM has generated the slices through the *xz* plane in order to indicate fibrillation within the fiber walls. Using the optical sections obtained by the CLSM, KS400 3D software was used to confirm fibrillation in the inner fiber walls. As water is removed from the cell wall, such inner fibrils might promote lumen collapse as well as transverse shrinkage by fiber wall-to- fibril contacts. If there are many crossing fibers bonded to the horizontal fibers in a web, the later shrinkage of upper fibers causes microcompressions in the lower fibers. For good bonding to occur between adjacent fibers, we need good areas of overlap. If the central lumen is pulled together, i.e., fibers collapse, then we maximize the surface area available for bonding. External fibrillation of the inner cell wall, the increase in surface tension forces and so on will all contribute to the collapse tendency. As an aside, this feature could also be one of the significant factors affecting the recycling potential of fibers. If the bonds are strong between the inner surfaces of the fibers no matter what we do in subsequent refining, it is impossible to uncollapse the fibers. It is supposed that a kind of hornification of the inner cell walls happens, too.

It was a new observation to see the fibrillation in the inner fiber wall surfaces. Up to now, fibrillation has been realized on two major effects including external and internal fibrillation but, from now on, we should extend our knowledge on fibrillation to the

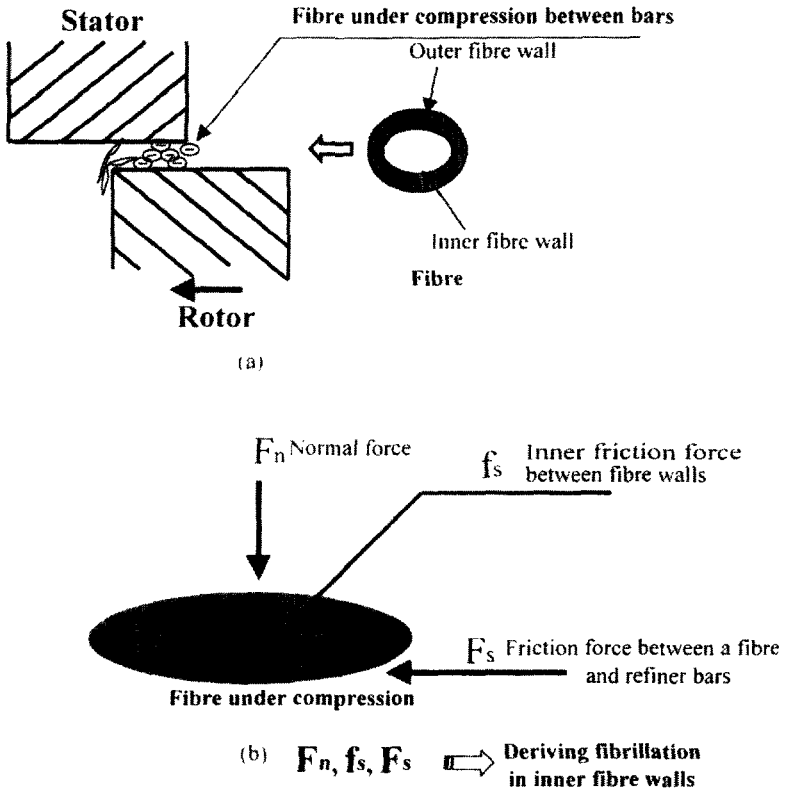

Figure 7. A schematic diagram of the force applied to fibers by passing bars.

inner fiber wall surfaces.

4. Conclusions

The optical sectioning technique of the CLSM and the image analysis technique showed that fibrillation occurred in both the outer and the inner fiber wall surfaces during refining by non-destructive techniques. In the early stages of refining, it was observed that a partial separation between the S1 and S2 layer in the secondary wall occurred forming a space in the wet fiber walls. further refining, it was clearly shown that the shear forces imparted by the refiner bar surfaces caused the S1 layer to become totally separated from the S2 layer as well as creating microfibrils.

Furthermore, the fibrillation in the inner fiber wall surfaces might be due to the normal force (F_n) by refiner bars, friction force between a fiber and refiner bars (F_s) and inner friction force between fiber walls (f_s). It was confirmed that the concept of fibrillation should be extended to fibrillation in the inner fiber wall surfaces as well as internal and external fibrillation.

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