

## **OBSERVATION OF THE SWELLING BEHAVIOR OF KRAFT FIBERS AND SHEETS IN THE ENVIRONMENTAL SCANNING ELECTRON MICROSCOPE**

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### **KEYWORDS**

Swelling, Kraft fibers, collapsed fiber, freeze-drying, fiber rising, roughening

**SUMMARY:** The swollen state of kraft fibers and sheets was observed in an Environmental Scanning Electron Microscope (ESEM). Chemical fibers swelled to a great extent during wetting. ESEM observations clarified the morphological features of freeze-dried kraft fibers in a handsheet soaked, from a handsheet soaked and defibrated, from a dry lap pulp beaten and of never-dried pulp. The fibers of a handsheet soaked in water for 3 days did not recover to complete tube-like shape. Image analysis made on three fibers in a handsheet showed that only fully collapsed fibers on the top surface, which faced a metal plate, recovered fairly to tube-like shape and the estimated vertical thickness increase of the fiber was about 50 %, corresponding to 11.5 % width shrinkage. But, less collapsed underlying fibers showed only 20 % thickness increase (4.0 % width shrinkage). Mechanical action of defibering did not affect shape recovery of fibers from a dried handsheet. Beating returned few dry lap pulp fibers back to tube-like shape though most of them remained collapsed. Fibers from never-dried pulp appeared tube-shaped, yet, with some parts collapsed presumably due to compression of the fibers to 20 % consistency for storage.

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Wood-containing papers roughen when exposed to water or high humidity, as in coating or offset-printing operations. One aspect of this roughening, commonly called "fiber-rising", is caused by the tendency of the fibers to recover their native tube-like shape (Skowronski, Lepoutre 1985 ; Hoc 1989; Aspler, Béland 1994). During drying, chemical pulp fibers collapse into ribbons; after calendering, mechanical pulp fibers are flattened. Only in rare occasions, fiber segments appear to recover a tubular shape in the work with Environmental Scanning Electron microscope (Forsberg, Lepoutre 1994). Chemical fibers do not recover their tubular form. In a similar, qualitative investigation, fiber rising was not due to a return of flat fibers to a precollapsed state and the rising fibers were in fact stiff fibers that withstood the calendering (Roever, Cosper 1996). On the other hand, in an excellent, detailed investigation which includes examination of cross-sections, lumen expansion in mechanical fibers after wetting or

exposure to 97% RH was observed, and the degree of expansion was proportional to the fiber wall thickness (Forseth, Helle 1997).

Generally, stiff wood-containing fibers are considered to be subject to lumen collapse when calendered but not when wet-pressed or dried. However, a possible lumen collapse during wet-pressing and drying is implied (Maloney et al. 1997). The authors report a large change of pore distribution of TMP by an NMR technique, though this pore closure includes both intra- and inter-fiber changes. They also report that, for chemical fibers, wet-pressing provides irreversible pore closure for beaten fibers and much less for unbeaten fibers.

There remains the question of the role of the chemical pulp in the roughening process. Irreversible changes in the surface structure caused by water was suggested (Lepoutre et al. 1986) for the first time. It was shown (Skowronski et al. 1988) that in papers made from kraft fibers, roughening was large but almost totally reversible. Swelling forces caused by water are the result of osmotic pressure developing within the cell wall (Grignon, Scallan 1980; Scallan, Tigerström 1992) and can be quite large. Kraft fibers have a large swelling potential - the swelling ratio of discs cut from kraft handsheets was assessed (Forsberg, Lepoutre 1994) to be 130 % - and although this may not be enough to cause the collapsed fibers to fully revert to the tubular shape they had in their never-dried state, it is nevertheless more than enough to cause disruptions in the sheet structure that may remain after the swollen kraft fibers return to their ribbon shape.

The objective of this work was to determine to what extent kraft fibers, isolated, or in paper, recover a tubular shape once wetted with water. The approach was to examine individual pulp fibers as well as paper sheets with different processing and drying history, after freeze-drying so as to preserve the swollen state (micropores in the cell wall should collapse, but the macro structure is assumed not to change). Kraft fibers underwent radiation damage in the ESEM when examined wet (Forsberg, Lepoutre 1994). Freeze-dried fibers can be examined at low humidity, thus reducing the risk of damage.

## EXPERIMENTAL

### Samples

Five different kraft fiber samples were examined. **Figure 1** illustrates the processes of sample preparation. Sample 1 was a handsheet made from kraft dry lap pulp. The pulp, moisture content ca 8 %, was soaked in water for 12 hours and beaten 5000 revolutions in a PFI mill. The freeness of the pulp was ca. 540 ml CSF. The pulp was made into sheets without any additives, by couching with two sheets of blotting paper inserted, pressing at 345 kPa for 5 min then 2 min with dry blotters each time and drying in the rings, all according to Tappi test method. The sheets were then conditioned at 23 °C and 50 % relative humidity. The moisture content was 8.3 %. A small piece was cut out from Sample 1 and adhered with conductive tape on a ESEM specimen stub. After ESEM observation, the sample was soaked together with the stub in water for three days and then freeze-dried. This was Sample 2. Sample 3 was freeze-dried pulp fibers prepared from Sample 1 by defibering sheets for 5 minutes after soaking. Sample 4 was freeze-dried fibers from the dry lap pulp soaked and beaten. Sample 5 was freeze-dried fibers prepared from never-dried kraft pulp, which had been stored at a

consistency of 20 %. The pulp was soaked for three days before freeze-drying. All the pulp fibers and sheets were made from bleached softwood kraft pulp. The species was *Pinus ponderosa* Dougl. (ponderosa pine).

### ESEM observation

The samples above were examined in the ESEM at 5 to 6 Torr, that is, ca. 30 % relative humidity. In order to make cross sections, samples were cut at an angle of 45°, being fixed between plastic cutting blocks with a 45 ° plane. ESEM (E-3 model, ElectroScan, Wilmington, MA, USA) was used for observation and micrographs were taken. So, dimensions in the “vertical” direction were divided by  $\sqrt{2}$  for the true length.

To measure fiber width, image analysis was applied. Edge extraction filtering and width measurement were carried out using an image analysis program, ImagePC-β1 (Scion Corporation, USA).

## RESULTS AND DISCUSSION

### Handsheet surface

*Photo. 1* shows a surface of the dried handsheet (Sample 1). All the fibers were collapsed. The surface of the two fibers lying horizontally from the upper left to the lower right had extremely flat surfaces. Probably, it is because those fibers faced a metal plate during restraint drying. Those fibers were observed to have wide portions at overlaps on the underlying fiber, where the fibers seemed to have been compressed together to the plate. Small wrinkles were observed on surfaces of some other fibers.

*Photo. 2* shows the surface of the same part of the same sheet after soaking and freeze-drying (Sample 2). All the fibers, in appearance, remained collapsed and the sheet structure was unchanged even after soaking for 3 days. However, note that the two fibers that had flat surfaces in *Photo 1* shrank laterally very slightly. *Figures 2* and *3* are images having emphasized fiber edges by edge extraction filtering. Drawn lines there show the measured sites. *Table 1* lists shrinkage in width of the three chosen fibers resulting from wetting and freeze-drying. Freeze-drying prevents interfiber bonding or drying-induced shrinkage of fibers so that fibers can be observed just like they were in water. Fiber A, on the top of the plane, indicated the largest amount of mean shrinkage of 11.5 %. Fiber B, second from the top, shrank in width 9.0 %. Fiber C underneath those two fibers shrank only 4.0 %. The difference could depend on the compression in the sheetforming and the drying processes. Those shrinkage data provided an indication of fiber shape recovery, as seen in *Table 2*.

*Table 2* schematically illustrates shape changes of a cross section of a pulp fiber as a function of shrinkage in width. The behavior of this fiber was modeled as follows: the fiber was completely collapsed before swelling; the fiber had a fiber wall thickness( $r$ ) of 3 μm and an outside diameter in the horizontal axis ( $2a+2r$ ) of 28 μm in the fully collapsed state (see the sketch for meaning of symbols  $a$ ,  $b$  and  $r$  for the dimensions); the outside and the inside peripheries were both ellipse-shaped; increase in the cross sectional area of the fiber wall by swelling was not considered in the calculation, although it was of the order of 100 % (Water

retention value of 0.6 g-water/g-dry fiber for a rewetted kraft fiber = 0.6 cc water / 0.67 cc of cellulose  $\approx$  100 % swelling, Forsberg, Lepoutre 1994). Much of the expansion was assumed to occur inwards. Therefore, the outer dimensions might not be too affected. On those rough assumptions, the shape of the cross section would be as shown in the last column. Considering the calculated shapes during wetting of fiber A (11.5 % width shrinkage), --- kraft fibers were considered to recover fairly to a tube-like shape and the vertical thickness increase ( $100b/r$ ) came to about 50 % in the fully wet state after soaking for 3 days. On the other hand, the underlying fiber C barely changed shape (20 % vertical thickness increase). Note that, in prior to the calculation, the values of  $r$  and  $a$  at 0 % shrinkage was roughly defined based on the mean size of the fibers observed with a cross section of a sheet in an electron micrograph. Those values needed no accuracy (10 % error in  $a$  at 0 % shrinkage makes just 1, 3 and 4 % errors in thickness increase for 4.0, 9.0 and 11.5 % in width shrinkage, respectively). These findings suggest that only fully flattened fibers like fibers A and B had a potential to recover some degree of tube-like shape.

### **Fibers obtained from soaked and defibrated handsheet**

*Photo. 3* shows freeze-dried fibers prepared from soaked and defibrated handsheets (Sample 3). Fibers look very similar to those which were only soaked (Sample 2) in that they appear more or less collapsed. Defibering should give a modest but certain effect on fiber swelling. However, the effect was not great enough to give rise to recovery to tube-like shape. Besides, the small wrinkles in the fiber surfaces remained as they were in the sheet.

### **Fibers obtained from beaten dry lap pulp**

*Photo. 4* shows freeze-dried fibers from the dry lap pulp defibrated and beaten (Sample 4). Only the fiber from the upper left to the lower right appears round, most of the fibers are collapsed. It is possible that only those fibers that received intensive mechanical treatment during beating could recover the tube-like shape due to a higher swelling degree. Besides, surfaces of the fibers are smoother and show fewer wrinkles than the fibers of Sample 3. The difference is that Sample 4 was air-dried before beating, while Sample 3 was air-dried, soaked and defibrated after beating. It is likely that, the surfaces of the fibers of sample 4 did not shrink very much when the dry lap pulp was produced, as the cell wall was not delaminated and the fibers were rather stiff. The uncollapsed state can be completely recovered by beating a commercial spruce kraft pulp (Page 1967). But, "the uncollapsed" state in the reference does not mean tube-like shape, also incompletely collapsed fibers with ribbon shape are included.

### **Never-dried fibers**

*Photo. 5* shows freeze-dried fibers from never-dried wet softwood kraft pulp. The fibers were much rounder than those air-dried. However, some parts of the fibers were collapsed. For storage, the pulp had been squeezed to ca 20 % consistency. The small pressure applied may have deformed the fiber shape.

### **Soaking of cross section**

*Photo. 6* shows a cross section of a dry handsheet (Sample 1). The fibers were packed tightly with only slight interfiber spaces. Small lumens were seen in some fibers.

*Photo. 7* shows a cross section of another handsheet (Sample 1) soaked and freeze-dried. The cutting was carried out in prior to soaking. Swelling made the fibers structure fairly loose. Every lumen became clearly visible and interfiber spaces were observed. The thickness of the sheet increased from 80  $\mu\text{m}$  before soaking to 150  $\mu\text{m}$ , which was much more than that expected from single fiber measurements. It should be kept in mind, however, that the fibers were not bonded with other fibers on one side and this accentuates the effect, compared to that of the fibers inside the paper structure.

## CONCLUSIONS

Freeze-drying was used when observing in ESEM how much bleached kraft fibers swell in water. The observation provided the following results. The fibers of a handsheet soaked in water for 3 days did not recover to complete tube-like shape of intact never-dried fibers. Only fully collapsed fibers on the top surface recovered fairly to tube-like shape and the estimated vertical thickness increase was about 50 %, corresponding to 11.5 % width shrinkage. But, a collapsed underlying fiber showed as low as 20 % thickness increase (4.0 % width shrinkage). However, it must be stressed that observations, which require a lot of time for preparation and actual measurements were made on 3 fibers only and that the results should be confirmed in an extensive study. The mechanical action of defibering did not affect shape recovery of fibers from a dried handsheet. Even beating returned few dry lap pulp fibers back to tube-like shape while most of them remained collapsed. Fibers from never-dried pulp appeared tube-shaped, yet, with some parts collapsed presumably due to compression of the fibers to 20 % consistency for storage. While all measurements reported here were made on a limited number of fibers thought representative of the sample examined, the results should be supported in a comprehensive study.

## ACKNOWLEDGMENT

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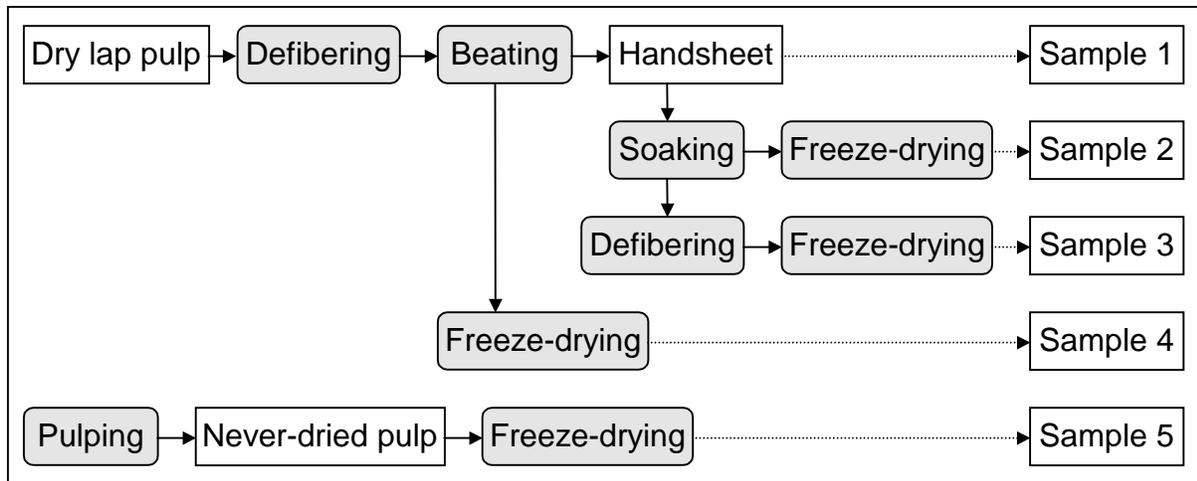
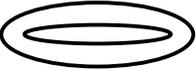
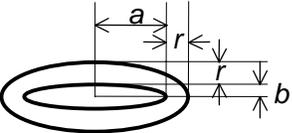


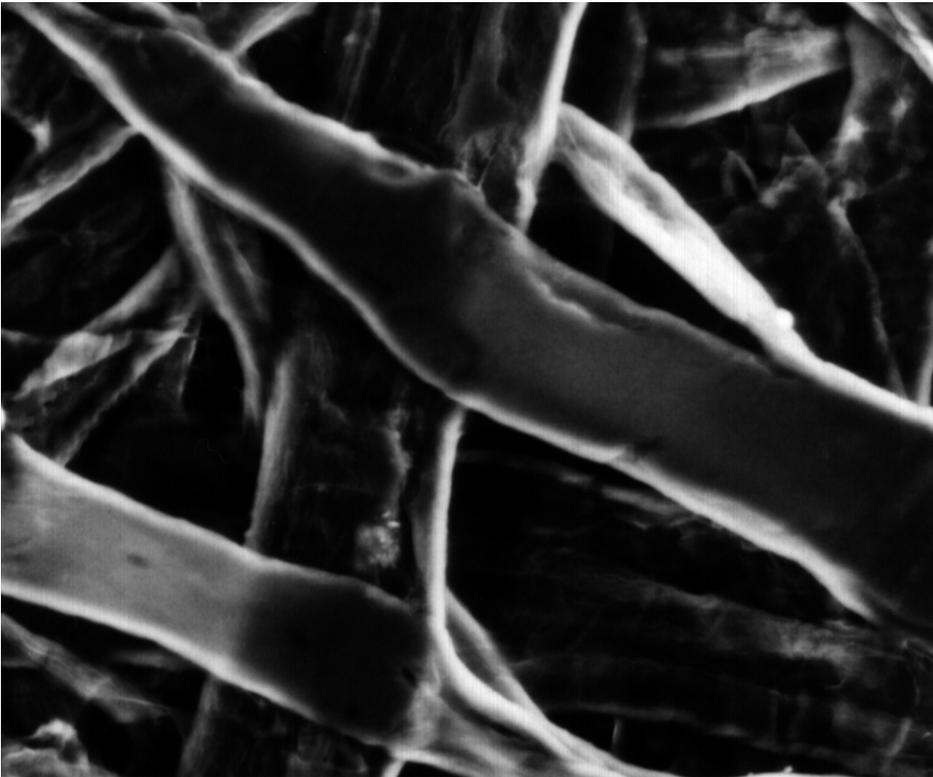
Fig.1 Flowsheet of fibers sample preparation for ESEM observation.

*Table 1 Shrinkage in width of softwood kraft fibers in a handsheet, resulting from wetting and freeze-drying. Average of shrinkage was rounded to the nearest 0.5 %.*

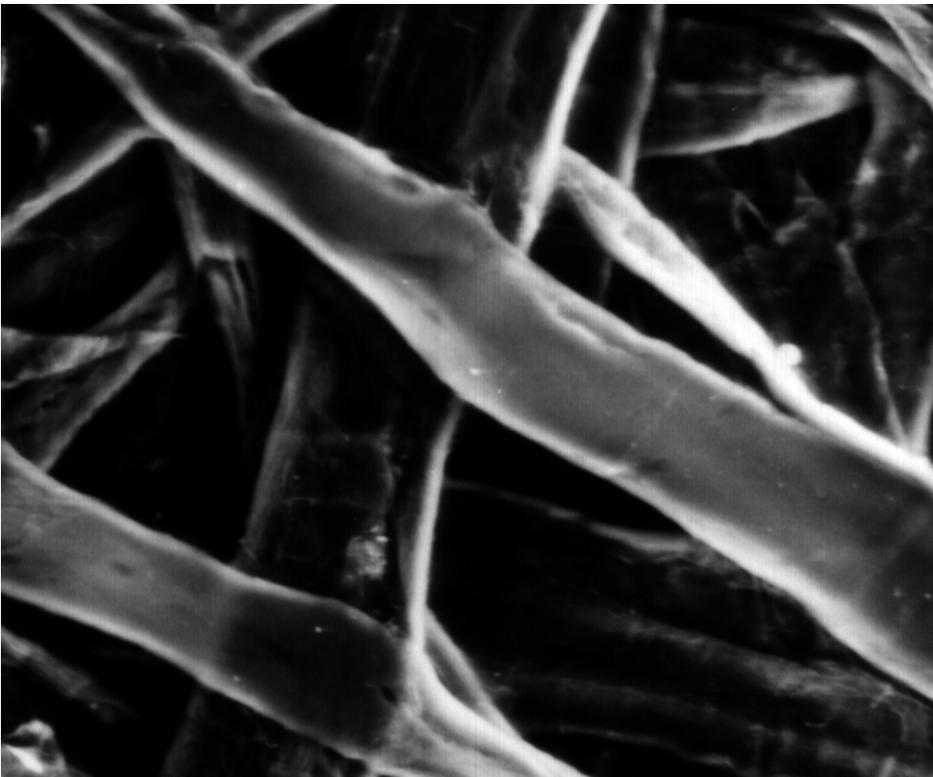
Site indicated in images	Width, $\mu\text{m}$		Shrinkage, %	
	Before	After	Average	
A 1	26.5	23.6	11.2	
A 2	32.9	29.3	10.9	
A 3	39.9	34.6	13.3	
A 4	38.2	34.4	9.9	
<hr/>				
B 1	29.1	26.3	9.7	
B 2	34.4	31.4	8.5	
<hr/>				
C 1	51.1	48.8	4.5	
C 2	44.8	43.2	3.5	

*Table 2 Estimated fiber thickness increase and shape change of kraft fibers. The calculations are based on the assumption that fiber wall thickness is unchanged. Thickness increase was rounded to the nearest 10 %.*

Width shrinkage, %	$r, \mu\text{m}$	$a, \mu\text{m}$	$b, \mu\text{m}$	Thickness increase, %	Shape of cross section
0	3	11	0	0	
4.0	3	10.45	0.55	20	
9.0	3	9.75	1.25	40	
11.5	3	9.40	1.60	50	



*Photo. 1 Handsheet surface made from beaten softwood kraft pulp*



*Photo. 2 The same area of Photo. 1 after soaking and freeze-drying.*

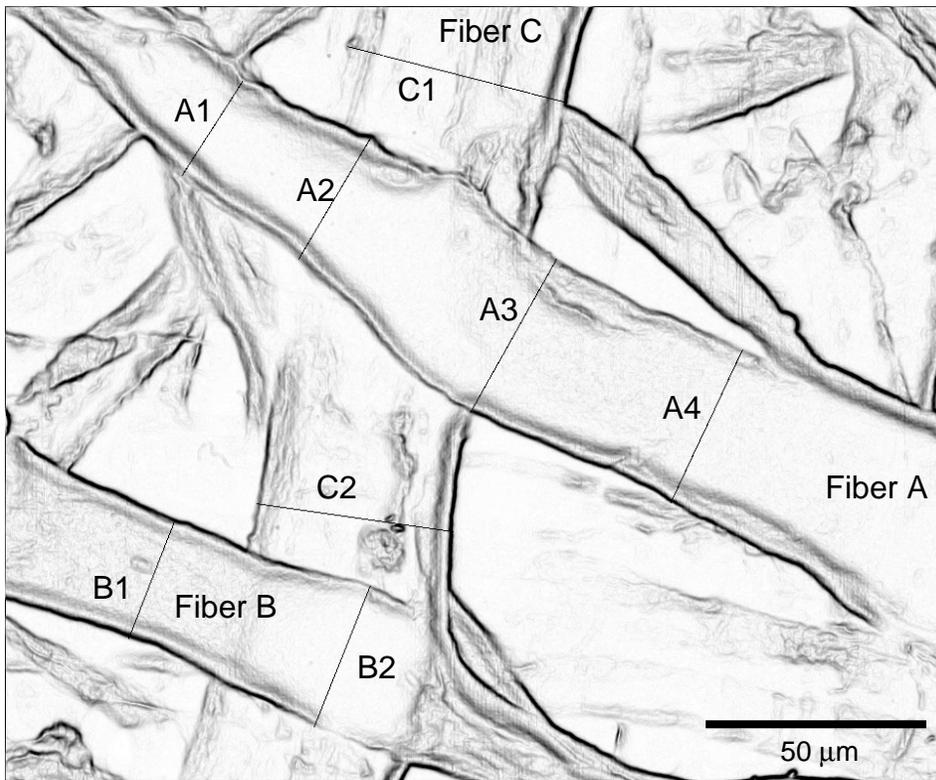


Fig. 2 Modified image of Photo. 1 by edge extraction filtering of image analysis

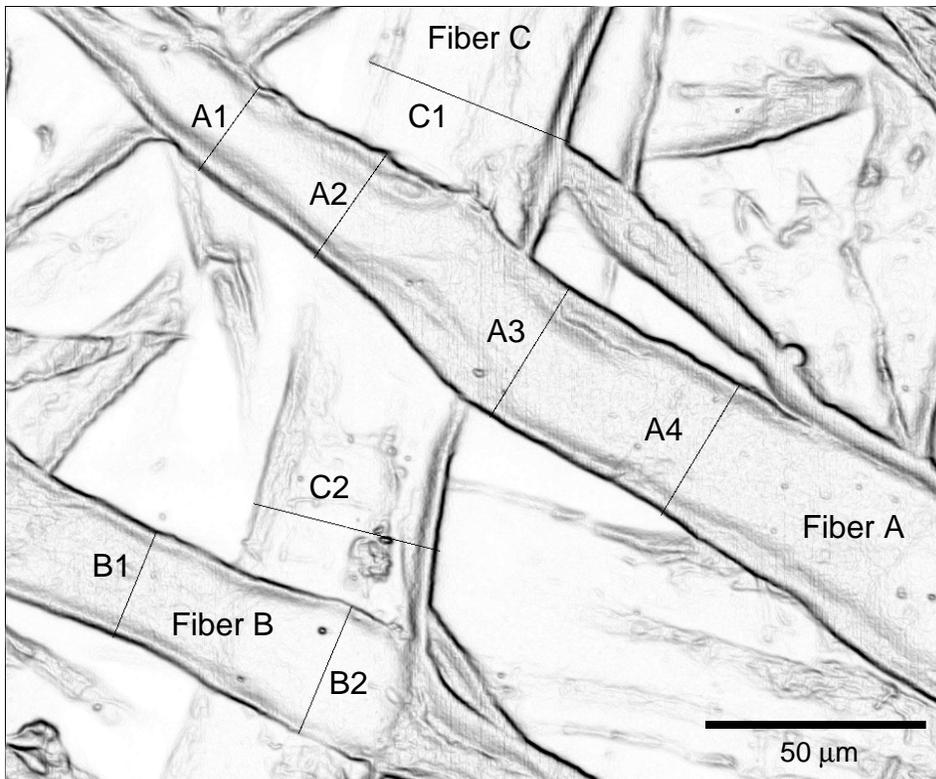
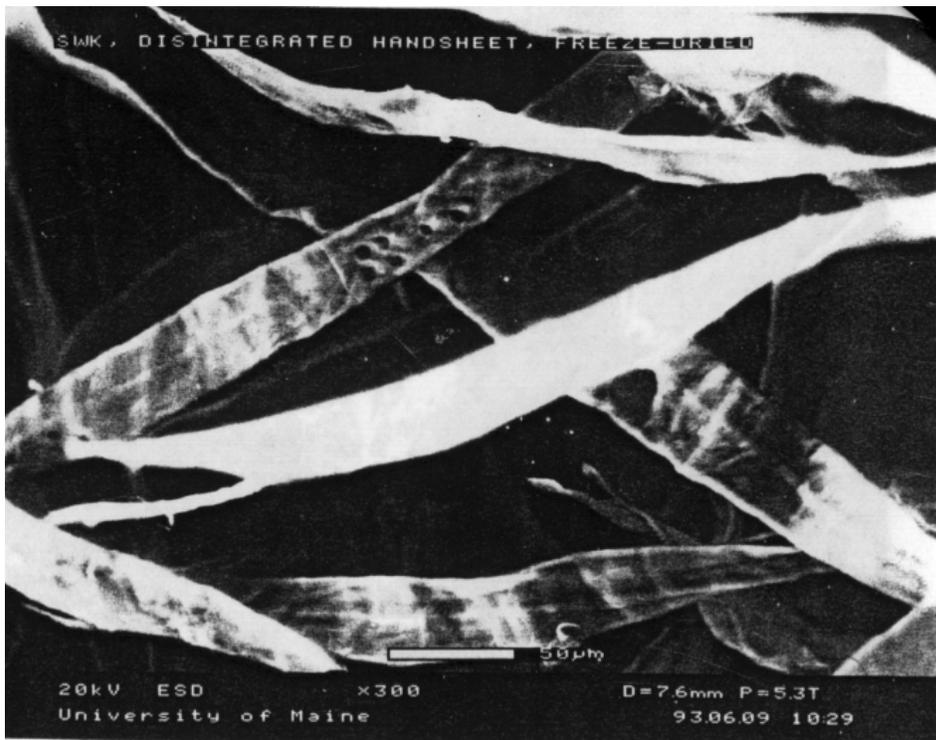


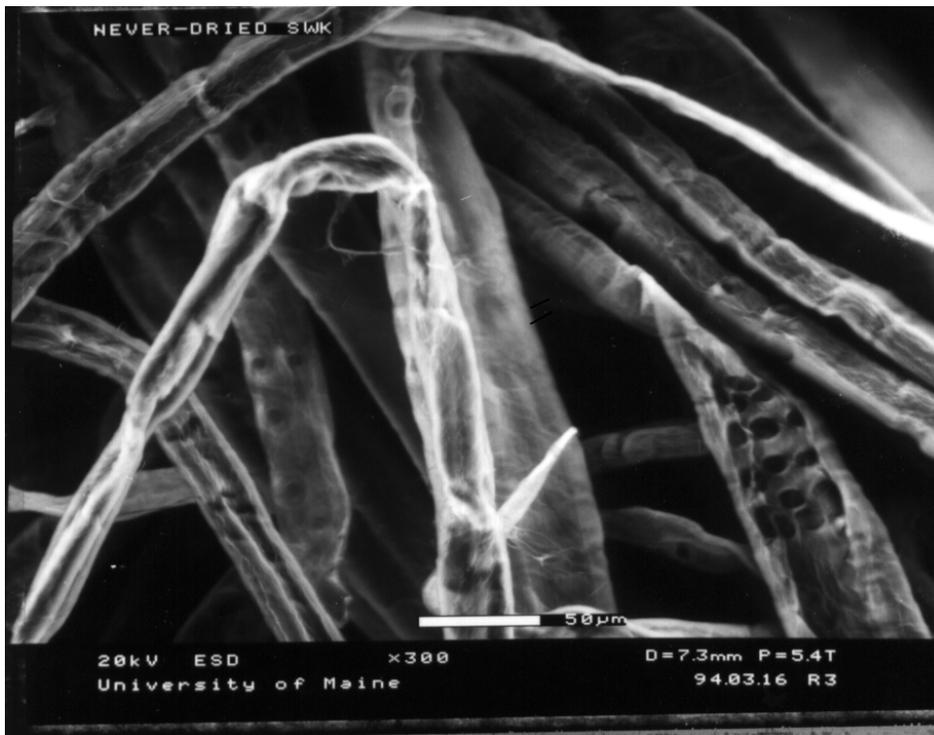
Fig. 3 Modified image of Photo. 2 by edge extraction filtering of image analysis



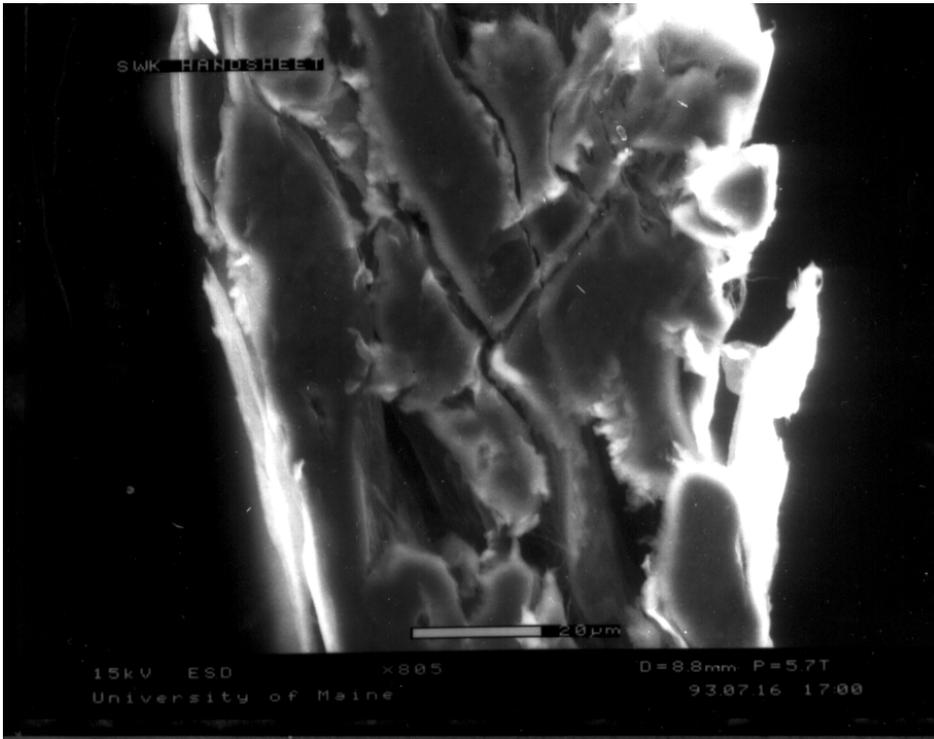
*Photo. 3 Freeze-dried fibers from handsheet soaked and defibrated.*



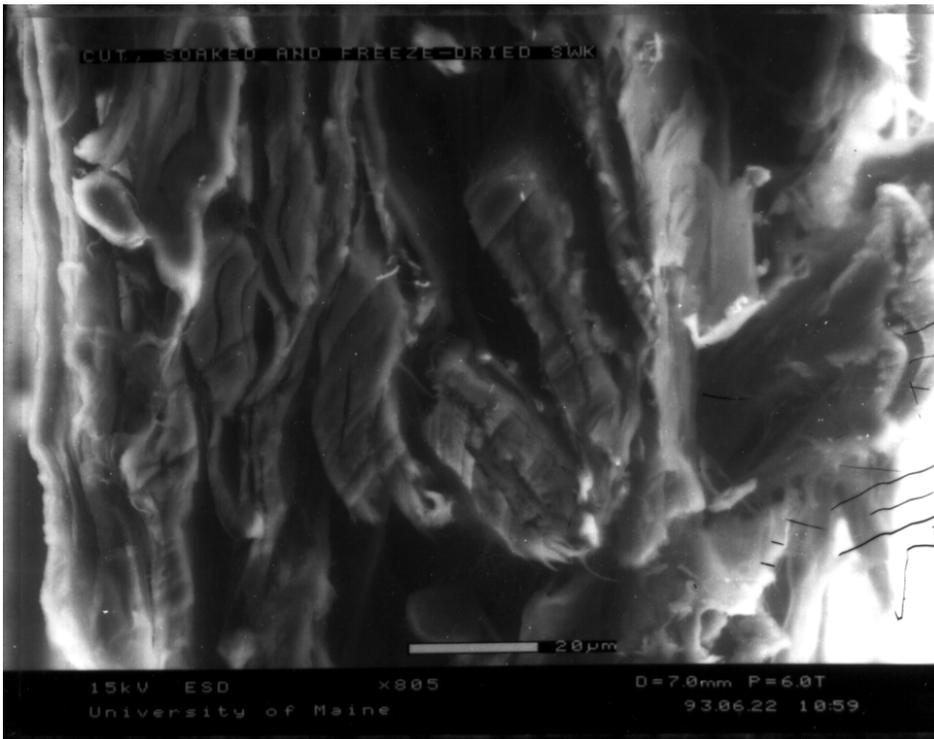
*Photo. 4 Freeze-dried fibers from softwood kraft pulp soaked, defibrated and beaten.*



*Photo. 5 Freeze-dried fibers of never-dried softwood kraft pulp.*



*Photo. 6 Cross-section of handsheet made of softwood kraft pulp.*



*Photo. 7 Cross section of handsheet cross-sectioned, soaked and freeze-dried.*