Paper VI

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Recycle Potential of Externally Fibrillated Chemical Pulp

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ABSTRACT

The objective of the study was to clarify the recycle potential of externally fibrillated chemical pulp. To this end, internally fibrillated and externally fibrillated fibers were prepared using a Lampén mill and an ultra-fine friction grinder, respectively.

Fibers were hornified to roughly the same level by recycling, regardless of the degree of internal or external fibrillation of nonrecycled fibers, and this was restored to some extent by refining. Externally fibrillated fibers offer greater potential for restoring swelling and straightening by refining than internally fibrillated fibers. The higher degree of external fibrillation of externally fibrillated fibers was not changed during recycling and refining.

After five recycles, the tensile strength and internal bond strength of externally fibrillated fibers were higher than those of internally fibrillated fibers. The loss in tensile strength for internally fibrillated fibers was not fully restored by refining. However, the strength of externally fibrillated fibers was restored beyond their original strength. The potential for restoring their internal bond strength was even more enhanced by refining.

KEYWORDS

External fibrillation, Internal fibrillation, Lampén mill, Recycling, Refining, Ultra-fine friction grinder.

INTRODUCTION

The bonding potential of chemical pulp is significantly reduced by recycling which involves drying and rewetting. One of the main reasons is the partial loss of the fibers' swelling ability [1-5]. However, refining can compensate for the loss of fiber swelling to some extent by re-opening the pores in the fiber wall, although it is generally accompanied by fines generation [2-9]. The refining degree of virgin pulp is a key factor affecting the potential for restoring fiber bonding: the higher the refining degree of virgin pulp, the lower the potential for restoring sheet properties [1]. Therefore, refining, before or after recycling, plays an important role for the recycle potential.

Refining causes a variety of simultaneous changes in the fiber structure, such as internal fibrillation, external fibrillation and fines formation [10-11]. Among these effects, swelling is commonly recognized as an important factor affecting the strength of recycled paper. A study on the role of fines in the recycling process has also been carried out. The recycle potential of fines has been a controversial issue. One view is that hornified fines from recycled paper can not be recovered with refining [2, 12], so the fines act as a filler and disturb the drainage properties, not the strength of paper [2, 6-7, 12]. Another view is that the hornification of fines can be reversed with refining [13], enhancing the strength of paper [14-15].

Although external fibrillation is one of the main refining effects, its role in the recycling process has not been studied to any major extent. As the degree of refining increases, external fibrils attached to the fibers increase to a certain level, but the simultaneous occurrence of swelling makes it difficult to assess the role of external fibrillation for the recycle potential. Using an ultra-fine friction grinder, the external fibrillation can be increased, while changes in other properties are minimized [16], which makes it possible to evaluate the role of external fibrillation during drying and rewetting cycles. In the Lampén mill [17-18], internal fibrillation develops as a result of compressive forces, which provides a useful reference for comparison with pulp treated in the grinder.

The objective of the present study was to examine how external fibrils still attached to fibers behave in the

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recycling process and in refining. This study is not intended to simulate industrial recycling of paper, but to provide a deeper understanding of the fundamental role of fiber morphology during drying and rewetting cycles and during refining.

EXPERIMENTAL

Pulp Used

Once-dried bleached kraft softwood pulp, consisting of a mixture of Scots pine (*Pinus sylvestris*, 56%) and Norway spruce (*Picea abies*, 44%), was obtained from a Finnish pulp mill. The pulp was disintegrated for 10 min in a Valley beater (ISO 5264-1:1979).

Recycling Experiment

The disintegrated pulp was refined either in a Lampén mill or in an ultra-fine friction grinder (Masuko Sangyo Co. Ltd., Japan). Pulp with a consistency of 3 % was refined to 8000 rev in the Lampén mill, which is a rotating-ball mill consisting of a 10-kg ball and a housing. This is shown in Figure 1 (A). Pulp with a consistency of 5% was recirculated up to 30 times through a 240 µm gap in the grinder, which consists of two grinding stones with a diameter of 250 mm: a lower rotating one and an upper stationary one. This is shown in Figure 1 (B). The rotating speed used was 1500 rpm, and the grit class of the grinding stones was 46 (grit size of 297-420 µm). The operating principle of the grinder is described in detail in an earlier publication [19].

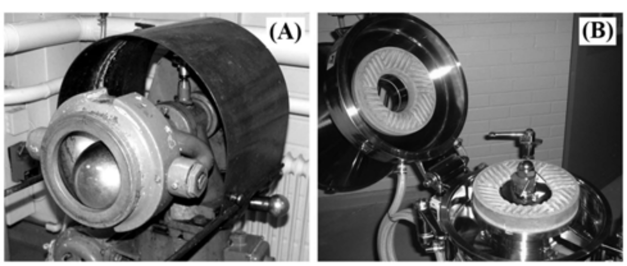


Figure 1. **A Lampén mill (A) and an ultra-fine friction grinder [19] (B).**

After refining in the Lampén mill and the grinder, the fines fraction passing through a 100-mesh screen was removed using a Bauer-McNett classifier, and the long-fiber fraction retained on the 100-mesh screen was used for the recycling experiment. A handsheet with a basis weight of 65 g/m^2 was prepared according to ISO 5269-1, and the pulp left from handsheet preparation was used to prepare a pulp pad with a basis weight of about 300 g/m^2 using a standard sheet mould, dried at 80 °C for 6 hours, and re-slushed. This procedure was repeated five times. The five-times recycled pulp, originally refined in the Lampén mill and the grinder, was refined separately to 8000 rev in the Lampén mill, and a handsheet with a basis weight of 65 g/m^2 was then prepared according to ISO 5269-1. The procedure for the recycling experiment is illustrated in Figure 2.

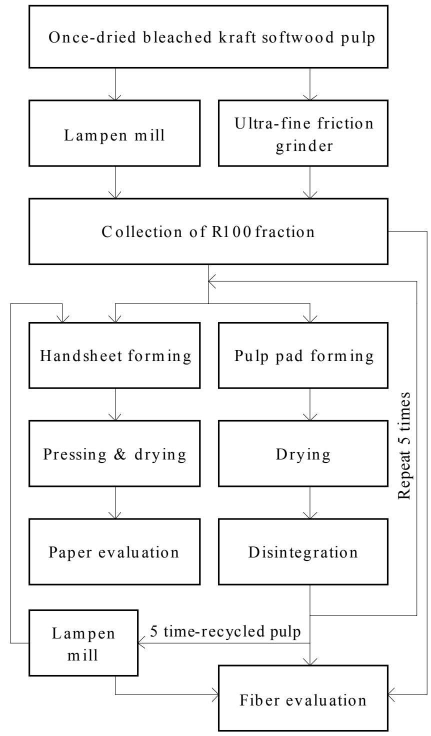


Figure 2. **Procedure for recycling experiment.**

Although only the long-fiber fraction was recycled five times and then refined in the Lampén mill, some fines were generated during recycling and Lampén refining. However, the fiber properties were measured for the long-fiber fraction which probably contained some fines. Fiber length (length weighted) and curl were measured using a FiberLab 3.0 (Metso Automation). The fiber saturation point (FSP) was measured with the solute exclusion technique using a 2 x 10^6 Dalton dextran polymer (Amersham Biosciences AB, Uppsala, Sweden) to evaluate the change in internal fibrillation of fibers [13]. The average of two measurements for the FSP was reported in this study. Fines-free pulp was also used in some cases for the FSP measurement in this study. A scanning electron microscope (SEM, DSM 962, Zeiss) and a light microscope with a phase contrast mode (DM LAM, Leica) were used for measuring the external fibrillation of fibers qualitatively and quantitatively, respectively. For the quantitative measurement, at least 20 images, each containing several fibers, taken with a light microscope were further analyzed using an image processing tool to calculate the proportion of the area of external fibrils to fibers, which allowed a quantitative measurement of the degree of external fibrillation. Paper properties were evaluated according to standard methods.

RESULTS AND DISCUSSION

Mechanical Treatment before Recycling

Figure 3 shows the changes in internal and external fibrillation with refining in the Lampén mill (A) and in the grinder (B). The FSP of fibers develops similarly both in the Lampén mill and in the grinder, increasing initially and then reaching a plateau. However, the external fibrillation develops differently in these two refining devices, as was to be expected. The Lampén mill does not increase the degree of external fibrillation, while the grinder increases the amount of external fibrils still attached to the fibers. In the plateau of the FSP curve, from pulp passage 6 to passage 30, as shown in Figure (B), the change in internal fibrillation seems to remain constant, though external fibrillation seems to be a major effect. Therefore, pulp with the passages of 21 and 30 are considered as externally fibrillated fibers while pulp refined with 8000 rev in the Lampén mill is considered as internally fibrillated fibers in this study.

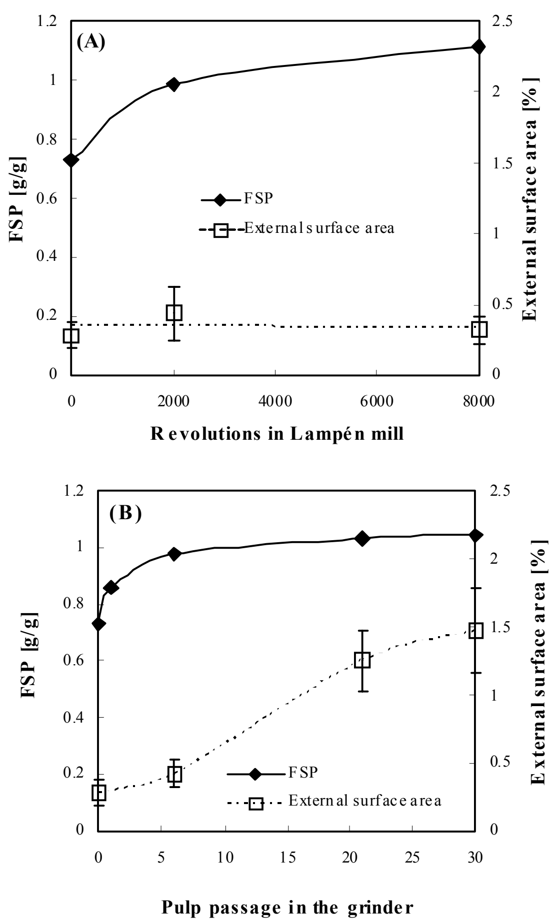


Figure 3. **Development of internal and external fibrillation of fibers refined in the Lampén mill (A) and in the grinder (B).** Pulp passage is defined as the number of times that the pulp is recirculated through the gap in the grinder. Error bars represent the 95% confidence interval, otherwise the average of two measurements is shown.

Other important properties such as fiber length and fiber curl seem to undergo little change during refining in both devices, which is shown in Table 1. A curl index of about 10 % is considered to represent straight fibers, an index of about 20% curly fibers [20].

Table 1. **Changes in fiber length and curl with refining in the Lampén mill and in the grinder.**

Grinder	Fiber length	Curl	Lampén mill	Fiber length	Curl
[Pulp passage]	[mm]	[%]	[rev]	[mm]	[%]
0	2.10	22.9	0	2.10	22.9
6	2.08	21.5	2000	1.99	20.7
30	2.13	21.8	8000	2.11	19.9

Effect of Recycling

As the number of drying and rewetting cycles increases, the swelling ability of fibers is reduced because of their hornification. This is shown in Figure 4 (A)-(B). The FSP of unrefined fibers decreases slower than that of highly refined fibers in both refining devices. This agrees with

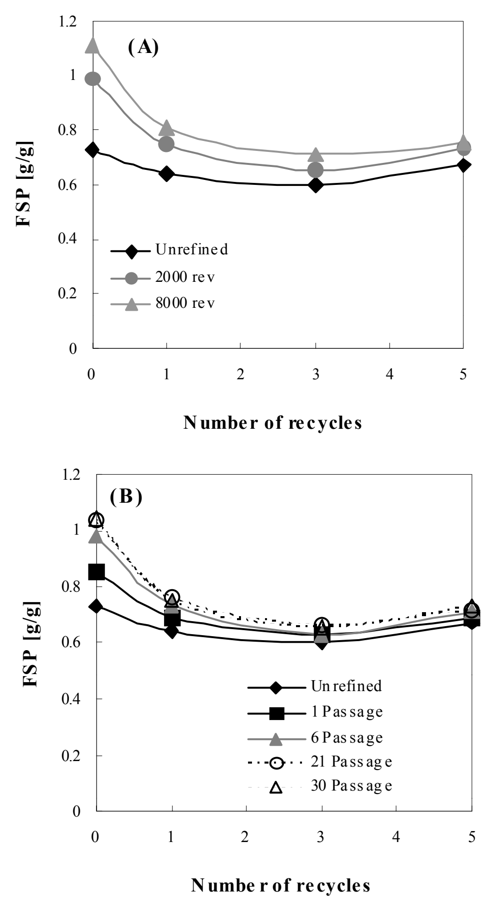


Figure 4. **Effect of recycling on internal fibrillation. Nonrecycled pulp was refined in the Lampén mill (A) and the grinder (B).**

the findings of the previous study, according to which the swelling ability of more refined fibers is more difficult to restore in the recycling process [1]. The FSP of both internally fibrillated fibers and externally fibrillated fibers decreases to a similar level, although that of internally fibrillated fibers at recycle 0 is slightly higher.

It would be important to examine the changes in the fiber surface during recycling. Fibers refined in the Lampén mill have a smaller amount of external fibrils attached to their surface, regardless of the degree of refining, and their degree of external fibrillation is not changed by the number of recycles. This is shown in Figure 5 (A). A similar trend with recycling is seen for the fibers treated in the grinder, as shown in Figure 5 (B). The external fibrils might have collapsed onto the fiber under milder drying conditions, but they may be able to rise from the fiber surface again under given wetting conditions.

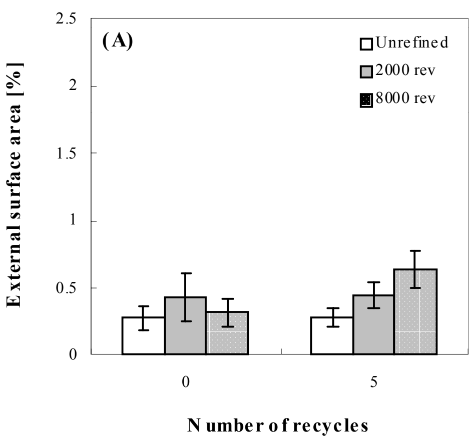
Table 2-1 shows the changes in average fiber length during recycling. No changes in fiber length are found with recycling, except that the length of internally fibrillated fibers decreased a little during the first cycle. Although in an earlier study [5] the recycling process was found to straighten fibers, this was not found in the present study, as shown in Table 2-2. This may be due to the differences in recycling conditions. In the present study, milder conditions were used in recycling, which may not cause the fibers to straighten.

Table 2-1. **Effect of recycling on fiber length, mm.**

No. Recycles	Unrefined	2000 rev	8000 rev	6 passage	30 passage
0	2.10	1.99	2.11	2.08	2.13
1	2.05	1.99	1.93	2.09	2.09
3	2.07	2.01	1.92	2.07	2.08
5	2.08	2.02	1.95	2.07	2.07

Table 2-2. **Effect of recycling on fiber curl, %.**

No. Recycles	Unrefined	2000 rev	8000 rev	6 passage	30 passage
0	22.9	20.7	19.9	21.5	21.8
1	23.0	21.8	20.0	22.5	21.9
3	22.4	20.7	20.0	21.5	21.9
5	21.9	20.6	19.8	21.2	21.9



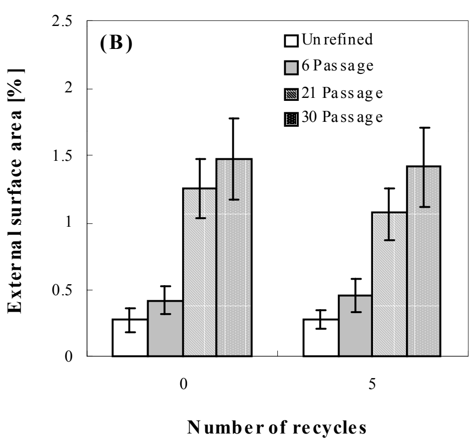


Figure 5. **Effect of recycling on external fibrillation. Nonrecycled pulp was refined in the Lampén mill (A) and the grinder (B). Error bars represent the 95% confidence interval.**

Figure 6 shows the changes in tensile strength during recycling. The tensile strength of unrefined pulp did not change during recycling, while the more refined fibers lost more of their strength. A significant reduction in strength for refined fibers occurred during the first cycle, which agrees with the findings of earlier studies [2, 5]. The tensile strength of externally fibrillated fibers is similar to that of internally fibrillated fibers at recycle 0. As the number of recycles increases up to 5 times, the tensile strength of internally fibrillated fibers decreases more than that of externally fibrillated fibers. One of the reasons for this can be found in the difference in the degree of external fibrillation. The changes in internal bond strength during recycling are similar to that of the tensile strength, which is shown in Figure 7.

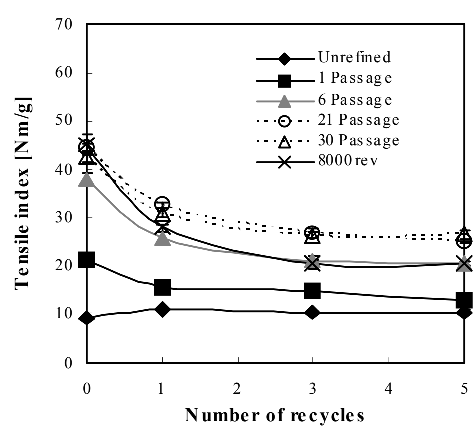


Figure 6. **Effect of recycling on tensile strength of paper. Error bars represent the 95% confidence interval.**

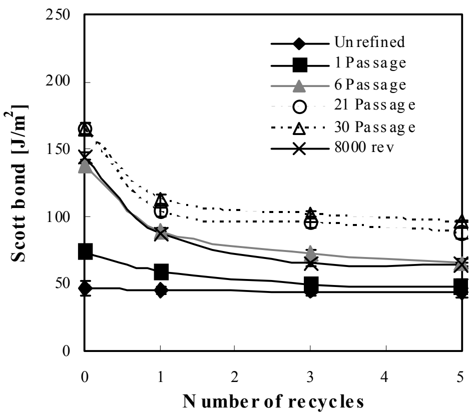


Figure 7. **Effect of recycling on internal bond strength of paper. Error bars represent the 95% confidence interval.**

Effect of Re-refining

The loss in fiber swelling caused by recycling can be restored by refining. When five-times recycled fibers are refined in the Lampén mill, their swelling is restored to some extent as shown in Figure 8. Refining, however, does not fully restore the FSP of fibers refined in the Lampén mill. For externally fibrillated fibers, the FSP was restored more than for the others, even beyond the FSP of nonrecycled fibers. The FSP was 1.16 g/g for the pulp passage of 21 and 1.13 g/g for the pulp passage of 30. The FSP of fibers without fines was 1.14 g/g for the pulp passage of 21 and 1.13 g/g for the pulp passage of 30. Therefore, according to this study, the swelling of fines does not seem to be restored by refining. Unlike the FSP restored by refining, the degree of external fibrillation was not found to be restored by refining. This is shown in Figure 9.

Refining of the five-times recycled fibers does not seem to change the fiber length, except for the externally fibrillated fibers. This is shown in Table 3-1. The shortening of the fiber length for the externally fibrillated fibers is probably closely related to the generation of fines caused by refining.

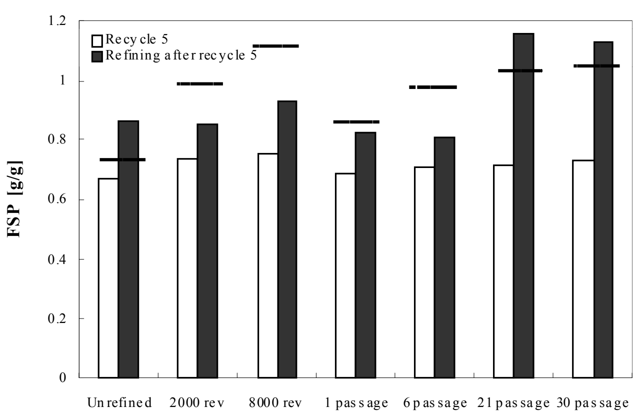


Figure 8. **Effect of refining on internal fibrillation. The FSP for nonrecycled fibers is marked with a solid line.**

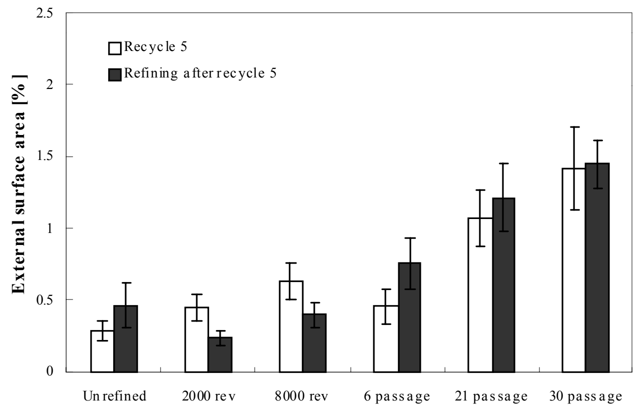


Figure 9. **Effect of refining on external fibrillation. Error bars represent the 95% confidence interval.**

The more external fibrils attached to the fibers, the easier the external fibrils are removed from the fiber surface by mechanical treatment.

Rather curly fibers can be straightened with refining as shown in Table 3-2, and more externally fibrillated fibers were found to be much more straightened than the others. As the fibrils are peeled from the fiber surface, the thickness of the fiber wall is probably reduced, making it susceptible to straightening during refining.

Table 3-1. **Effect of refining on fiber length, mm.**

	Unrefined	2000 rev	8000 rev	6 passage	21 passage	30 passage
Recycle 5	2.08	2.02	1.95	2.07	-	2.07
Refining after recycle 5	2.08	2.04	1.90	2.03	1.61	1.63

Table 3-2. **Effect of refining on fiber curl, %.**

	Unrefined	2000 rev	8000 rev	6 passage	21 passage	30 passage
Recycle 5	21.9	20.6	19.8	21.2	-	21.9
Refining after recycle 5	18.6	17.6	17.4	19.3	15.9	14.1

Figure 10 shows the effect of refining of 5 times recycled fibers on tensile strength. Refining was found to restore the bonding ability of recycled fibers having a different initial refining level. The strength of internally fibrillated fibers is not fully restored by refining. However, externally fibrillated fibers are restored to beyond the strength of nonrecycled fibers. This is similar to the restoration of the FSP, as shown in Figure 8, and the FSP seems to make a pronounced contribution to tensile strength. The potential for restoring their internal bond strength is more pronounced with refining, as shown in Figure 11.

Microscopic Observation

Figure 12 shows the surface of a handsheet made from internally fibrillated fibers at recycle 0 (A); a handsheet

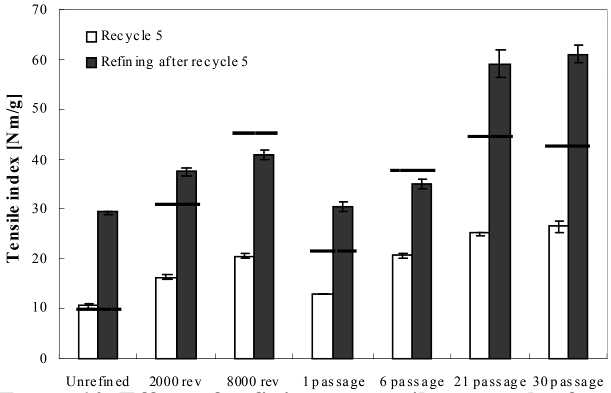


Figure 10. **Effect of refining on tensile strength of paper. Tensile strength for nonrecycled fibers is marked with a solid line. Error bars represent the 95% confidence interval.**

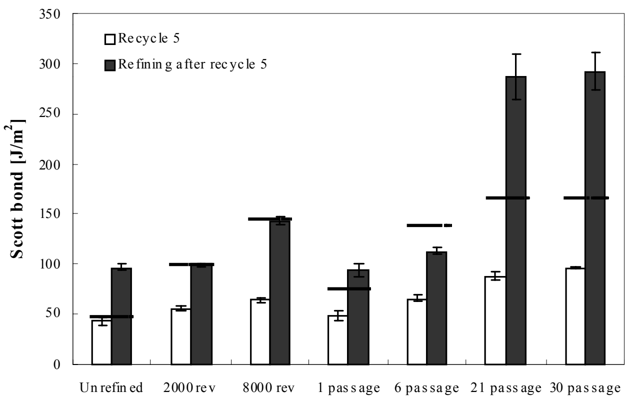


Figure 11. **Effect of refining on internal bond strength of paper. Internal bond strength for nonrecycled fibers is marked with a solid line. Error bars represent the 95% confidence interval.**

made from the same fibers at recycle 5 (B); and a handsheet made from recycled and subsequently refined fibers (C). It is difficult to distinguish between the images, in spite of the recycling and refining treatments.

Figure 13 shows the surface of a handsheet made from externally fibrillated fibers at recycle 0 (A); a handsheet made from the same fibers at recycle 5 (B); and a handsheet made from recycled and subsequently refined fibers (C). Externally fibrillated fibers at recycle 0, showing the external fibrils between the fibers, are very different from internally fibrillated fibers. The external fibrils at recycle 5 are still visible, but they seem to appear as a loose structure. Refining of the five-times recycled fibers seems to reactivate the external fibrils, which may contribute to the bonding between fibers. The amount of external fibrils for externally fibrillated fibers does not change during recycling and refining, but the quality of the external fibrils seems to differ, resulting in different paper strength.

CONCLUSIONS

Fiber swelling decreased to roughly the same level regardless of the degree of internal or external fibrillation of nonrecycled fibers after the drying and rewetting cycle was repeated 5 times, although the reduction ratio in fiber swelling was smaller for less refined fibers. The loss in fiber swelling caused by recycling was found to be restored by refining. Externally fibrillated fibers offered greater potential for restoring swelling and straightening by refining than internally fibrillated fibers. The higher degree of external fibrillation of externally fibrillated fibers was

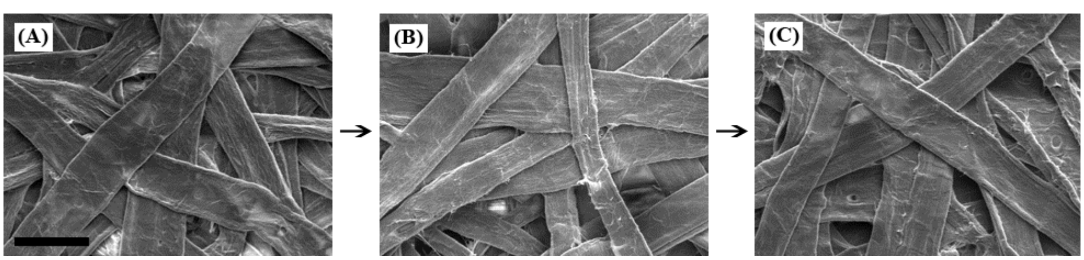


Figure 12. **SEM micrographs of a handsheet surface made from nonrecycled fibers (A), five-times recycled fibers (B) and five-times recycled, refined fibers (C). The pulp was refined to 8000 rev in the Lampén mill before recycling. Scale bar of 50 µm.**

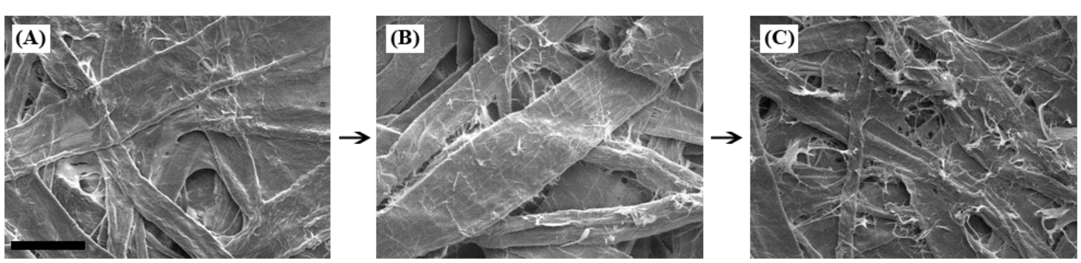


Figure 13. **SEM micrographs of a handsheet surface made from nonrecycled fibers (A), five-times recycled fibers (B) and five-times recycled, refined fibers (C). The pulp was recirculated 30 times in the grinder before recycling. Scale bar of 50 µm.**

not changed during recycling and refining, but the bonding ability of the external fibrils from nonrecycled, recycled and re-refined fibers seemed to vary.

The tensile strength and internal bond strength of externally fibrillated fibers decreased less than those of internally fibrillated fibers as the number of recycles increased. The loss in tensile strength for internally fibrillated fibers having a lower degree of external fibrillation was not fully restored by refining. However, the strength of externally fibrillated fibers was restored beyond their original strength. The potential for restoring their internal bond strength was even more improved by refining.

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