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Doctoral Thesis

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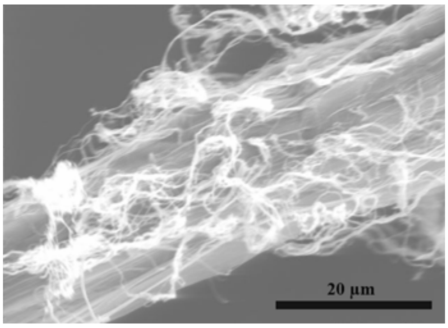

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Taegeun Kang

Dissertation for the degree of Doctor of Science in Technology to be presented with due permission of the Department of Forest Products Technology, for public examination and debate in Auditorium Puu II at the Helsinki University of Technology (Espoo, Finland) on the 28th of September, 2007, at 12 noon.

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Abstract

The purpose of this thesis was to clarify the role of external fibrillation in pulp and paper properties. If the degree of external fibrillation could be controlled and the role of external fibrillation were known, this would provide better possibilities for utilizing the full potential of fibrous material.

External fibrillation caused by refining is inevitable, but its role has not been clearly explained, because conventional refiners produce a variety of simultaneous effects, such as internal fibrillation, external fibrillation, straightening or curling of fibers and fines formation. Therefore, a special type of refiner was required for evaluating the specific role of external fibrillation. An ultra-fine friction grinder consisting of two grinding stones was found to overcome the limitation related to conventional refiners.

Using the grinder, it was possible to promote external fibrillation while keeping the internal fibrillation and curl constant. An increase in external fibrils attached to fibers was found to increase the retention of filler during sheet forming in a high-vacuum dewatering device. Promoting the degree of external fibrillation contributed to strengthening the bonding layer between fibers, resulting in improved tensile strength and internal bond strength. Recycling reduced fiber swelling and paper strength, but for externally fibrillated fibers this loss was restored by refining more than for internally fibrillated fibers, even beyond the swelling and strength of the nonrecycled fibers. Increasing external fibrillation alone was found to play an important role in fiber and paper properties, so it can be used as means to control those properties more effectively.

Keywords External fibrillation, Ultra-fine friction grinder, Refining, Strength, Recycling, Retention	
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PREFACE

This study was carried out in the Laboratory of Paper and Printing Technology at the Helsinki University of Technology during 2002-2005 as a part of the International Doctoral Program in Pulp and Paper Science and Technology (PaPSaT). Financial support from the Ministry of Education, the Academy of Finland, the National Technology Agency of Finland (TEKES) and the Finnish Paper Engineers' Association (PI) is gratefully acknowledged.

I would like to express my sincere thanks to my supervisor, Professor Hannu Paulapuro for his encouragement and the advice throughout my thesis work. I would also like to thank my co-author, Phichit Somboon for his contribution. All members of the Laboratory of Paper and Printing Technology are appreciated for their support. Special thanks go to Leena Nolvi and Eija Korhonen for meausuring the fiber saturation point, Mika Sirviö for CLSM measurement, Kaarlo Nieminen for measuring the degree of external fibrillation, Alexey Kononov for the discussions and cooperation, and to Jaesung Eom and Eunyoung Kim for conducting part of the experimental work. I wish to thank Etsuzou Tatsuguchi and Keiji Takahashi at Masuko Sangyo Co. Ltd., Japan for their great help with the grinder. Olof Andersson is acknowledged for revising the language. I would also like to thank Professors Byoungmuk Jo and Byungho Yoon at Kangwon National University, Korea for their kind support during my stay in Finland.

Finally, I wish to express my thanks to my parents, my mother-in-law, my wife Tomoko, and my children Hanna and Noah for their love and support.

Espoo, April 18, 2007

Taegeun Kang

LIST OF PUBLICATIONS

The thesis is based on six original publications listed below, which are referred to in the text by Roman numerals.

I. Kang, T. and Paulapuro, H., New Mechanical Treatment for Chemical Pulp, *Journal of Process Mechanical Engineering.* 220(2006)3, p.161-166.

II. Kang, T. and Paulapuro, H., Effect of External Fibrillation on Paper Strength. *Pulp & Paper Canada.* 107(2006)7/8, p.51-54.

III. Kang, T. and Paulapuro, H., Characterization of Chemical Pulp Fines. *Tappi Journal.* 5(2006)2, p.25-28.

IV. Kang, T., Somboon, P. and Paulapuro, H., Fibrillation of Mechanical Pulp Fibers. *Paperi ja Puu.* 88(2006)7, p.409-411**.**

V. Kang, T. and Paulapuro, H., Effect of External Fibrillation on the Retention of Filler. *Japan Tappi Journal.* 60(2006)8, p.88-92.

VI. Kang, T. and Paulapuro, H., Recycle Potential of Externally Fibrillated Chemical Pulp. *Progress in Paper Recycling.* 15(2006)2, p.11-17.

THE AUTHOR'S CONTRIBUTIONS

In the above publications I-VI, the author completed the following:

I, II, III, V, VI	All experiments, analysis and first version of manuscript
IV	Experiments in part, main part of analysis, first version of manuscript

LIST OF ABBREVIATIONS

C-PAM	Cationic polyacrylamide
CSF	Canadian standard freeness
ECF	Elemental chlorine-free
FSP	Fiber saturation point
MBF	Moving belt former
PCC	Precipitated calcium carbonate
P14/R50	Pulp fraction passed through 14 mesh wire and retained on 50 mesh wire
P200	Pulp fraction passed through 200 mesh wire
R30, R50, R100, R200	Pulp fraction retained on 30, 50, 100, or 200 mesh wire
SEM	Scanning electron microscope
TMP	Thermomechanical pulp
WRV	Water retention value

CONTENTS

1. INTRODUCTION

Refining is an essential step in developing pulp fibers to their desired quality level in the papermaking process, and it causes a variety of simultaneous structural changes, such as internal fibrillation, external fibrillation, fines formation, fiber shortening or cutting, and fiber curling or straightening [1,2,3,4,5]. The simultaneous occurrence of these effects in conventional refiners has limited the possibilities of utilizing the full potential of fibrous material. If some of the structural properties of fibers could be better controlled, this could improve the possibilities for utilizing the inherent potential of the fiber material.

Several attempts have been made to produce a homogeneous refining effect using a shear refiner [6,7] and a compressive refiner [6,8,9], allowing the relative importance of different refining effects to be examined. Among these effects, the internal fibrillation of fibers is gaining more attention, because it improves the flexibility and collapsibility of fibers, which are essential for inter-fiber bonding. External fibrillation has attracted less attention, firstly, because it was not believed to improve paper strength even with increased density [6], secondly, because it generates fines [10], and thirdly, because it is difficult to assess the specific role of external fibrillation. External fibrillation, however, presumably contributes to some papermaking properties, such as the retention of fine particles. Although this previous research has expanded the knowledge of controlled fibrillation of fibers, the potential role of external fibrillation has not been studied to any major extent. Therefore, further research on the role of external fibrillation is justified.

This study focused on examining how to control external fibrillation while minimizing changes in other properties, and on clarifying the contribution of external fibrillation to the properties of pulp and paper.

2. OBJECTIVES AND STRUCTURE OF THE STUDY

The objective of the study was to find a way of controlling the external fibrillation of fibers and to evaluate the role of external fibrillation for the properties of pulp and paper. This was done using an ultra-fine friction grinder, controlling the degree of fibrillation. The thesis work was carried out in the following steps:

1. Reviewing current knowledge on the role of external fibrillation and the difficulties in controlling fibrillation.
2. Selecting a special piece of equipment and developing a new method for controlling fibrillation.
3. Evaluating the role of external fibrillation for the properties of pulp and paper.

Based on earlier studies, the following hypotheses were put forward to fulfill the objectives:

- External fibrillation can be produced to some extent independently of other refining effects such as swelling and straightening of fibers and fines formation.
- External fibrillation alone increases retention of fine particles and strength properties of a sheet.
- Improved paper property combinations can be obtained by controlling external fibrillation alone.
- External fibrillation plays a role even at commercial scale.

The structure of the study is illustrated in Fig. 2.1.

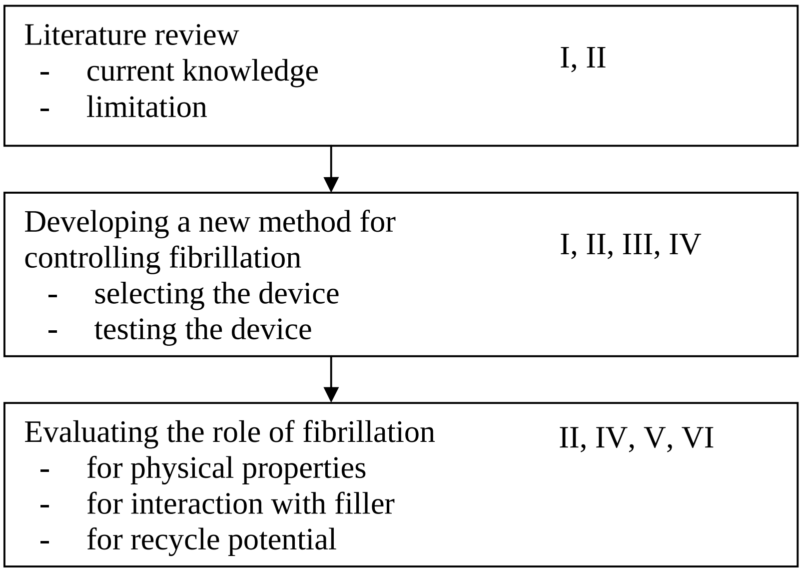

Fig. 2.1 Structure of the study. The roman numerals refer to publications.

The objectives of the thesis are addressed in the publications, as shown in Table 2.1.

Table 2.1 Appearance of the thesis objectives in earlier publications.

	Publication					
Objective	I	II	III	IV	V	VI
1	x	x				
2	x	x	x	x		
3		x		x	x	x

3. LITERATURE REVIEW

3.1 Controversial view on external fibrillation

External fibrillation is one of the indispensable refining effects in conventional refiners, with concomitant occurrence of structural changes, such as internal fibrillation, fines formation etc. External fibrillation can be defined as a peeling off of fibrils from the fiber surface, while leaving them attached to the fiber surface [4,5]. Fig. 3.1 shows an example of external fibrils still attached to the fibers. Over the years, the role of external fibrillation has been controversial, while there has been no doubt about the role of internal fibrillation.

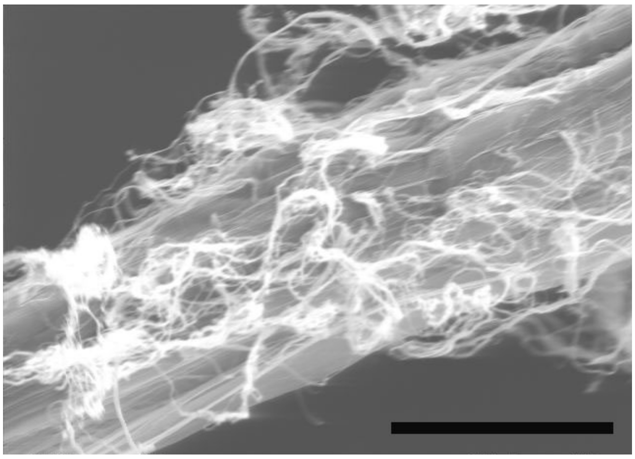

Fig.3.1 Example of external fibrillation of once-dried bleached kraft softwood pulp beaten for 90 min in a Valley beater, Scale bar of 20 µm [11].

Strachan [12,13] emphasized that the fiber surface can be fibrillated even in the early beating stage, and that the external fibrils play an essential role for the cohesion between fiber surfaces. Clark [14] also supported the view that external fibrils attached to the fiber serve as bonding agents for inter-fiber bonding. Casey [15] pointed out that maximum strength is obtained when the fibers are relatively intact, but still have a high degree of external fibrillation compatible with the longest fiber length. Jayme and Hunger [16,17], and Page and Sargent [18], and Buchanan and Lindsay [19] showed microscopic images of external fibrils, which were believed to contribute to paper strength by spanning between the fibers. Claudio-da-Silva et al. [20] claimed that the geometry of the fibrils and macro-fibrils loosened from the outer layers of the fibers is an important factor affecting web strength, and that the role of external fibrillation in bond formation can be considered secondary to internal fibrillation. Nanko [21] has recently shown a bonding layer between fibers composed of micro- and macro-fibrils and this layer seems to be very important for strong bond formation.

The role of external fibrillation in strength development is likely to be more significant in mechanical than in chemical pulp [22,23,24,25]. The mechanism governing the development of mechanical pulp fibers is explained as a peeling-off mechanism; the primary wall and S_1 layer are peeled off, and the S_2 layer is exposed to the inter-fiber bond. The ribbons, membranes and fibrils induced by the peeling-off mechanism act as binding agents for the bonding.

Mason [26] and Emerton [27] pointed out that the external surface contributes to the drainage properties of the pulp, and to the retention of fine particles such as fillers, dyes and other additives. Strachan [12] concluded from his experiment that the increased external surface induced by beating contributes to the retention of filler particles. Silenius [28] recently introduced the SuperFill Technology, according to which a fibril-filler composite provides a better combination of light scattering and paper strength than a conventional filler.

Fibrillation of chemical and mechanical pulp developed by refining influenced the light scattering coefficient differently. The light scattering coefficient of chemical pulp decreases with refining, while that of mechanical pulp increases [29]. Both types of fiber collapse and fines in chemical pulp contribute to the densification of the sheet. This decreases the unbonded surface area, which accounts for the decreased light scattering coefficient. Mechanical pulp fines behave as loosely bonded filler material, creating new light scattering surfaces and open structures [30]. An increased degree of external fibrillation of the long-fiber fraction in mechanical pulp gave a higher light scattering coefficient and surface smoothness [31]. The quality of external fibrils attached to the fiber influences the light scattering differently, with narrow ribbon-shaped and thread-like fibrils making a positive contribution to light scattering. Broad ribbon-shaped fibrils have a negative effect on light scattering [32].

The inter-fiber bond structure and fiber morphology play an important role in the stress transfer mechanism. The wrapped-around-type bonds in the sheets, which can be found in bonds of highly flexible fibers, give a much higher stress transfer ratio, and thus increase the hygroexpansivity of sheets [33]. Fines contribute to making a tight and dense bond structure, and thus increases stress transfer, which results in increased hygroexpansivity [34,35]. Chemical pulp fines are more efficient in stress transfer than mechanical pulp fines because of their high bonding potential [34]. External fibrils attached to fibers may behave similarly to loose fibrils in the stress transfer mechanism, because external fibrils contribute to the formation of “skirt” or a “covering layer” at the edge of the fiber bond [21].

External fibrillation is closely related to fines formation. As external fibrillation increases due to rubbing against other fibers or a moving part of a metallic bar, external fibrils start to peel off from the fiber, which results in increased fines formation. A considerable economic advantage is believed to be obtained by promoting external fibrillation rather than fines formation [5]. As the amount of fines increases, it improves paper strength properties, but has some negative effects such as dewatering. Although no research has been for a direct comparison of the role of loose fibrils and fibrils attached to fibers, these may be similar to each other, because loose fibrils are the detached form of external fibrils.

Harrison [36] claimed that external fibrillation does not improve paper strength at all, because the fibrillation develops at a later stage of beating when the strength remains constant. Gallary [37] and Cottrall [38] emphasized that external fibrillation does weaken the inter-fiber bond in spite of the increased bonded area. Tasman [39] pointed out that external fibrillation decreases tensile strength due to a possible loss of fiber strength.

Despite the many controversial opinions, the relative role of external fibrils in combination with other important properties, such as internal fibrillation, could not be verified due to difficulties in separating the simultaneous refining effects. To gain a better understanding of the relative importance of these effects, Hartman [6] developed an abrasion refiner in which pulp was abraded between rotating parallel disks. In this device, the roughness of the disk surfaces can be changed by attaching silicone carbide sandpaper with various grit sizes. In Hartman’s experiments, the degree of external fibrillation was found to increase at a fixed gap of 4 mm, while the changes in fiber length and zero-span breaking length were minimized, and different degrees of external fibrillation could be achieved by varying the grit size and refining time. Only one gram of pulp was used for the treatment. Hartman concluded that external fibrillation did not produce any increase in tensile strength, in spite of a slight increase in sheet density.

Although the role of external fibrillation has been controversial for many decades, its role in combination with internal fibrillation has been considered important [2,3,4,5,40].

3.2 Method measuring the degree of external fibrillation

External fibrillation involves the creation of a new surface, and it can be evaluated by measuring the external specific surface area [10,22,24,41,42,43,44,45,46,48,49]. There have been several techniques available to measure the specific surface, which are based on permeability, filtration, silvering and settling.

The liquid permeability method [26,41] measures the water permeability of a pulp pad under different levels of compression, and the specific surface can be calculated using a modified Kozeny-Carman equation. The filtration method [42,43], which is based on the same principle as the permeability method, except that it uses a pulp suspension, measures the filtration resistance of pulp under constant-rate filtration, which is then used to calculate the specific surface with the same equation.

In the silvering method [44,45], an ammoniacal solution of $AgNO_3$ is used to deposit a thin layer of silver on the fibers, and the specific surface of the pulp is determined from the decomposition of H_2O_2 solution. However, it is not recommended for finely fibrillated or well-beaten fibers, because silvering causes external fibrils to collapse onto the fiber, which gives a lower value for the surface area.

The settling method [46] can also be used for measuring the specific surface. The settling rate is obtained from the initial slope of the settling curve. The settling rate was found to be strongly dependent on the specific surface area, although several other factors affect the settling rate, e.g. coarseness, fiber length and flexibility. The specific surface can be calculated from the settling rate using Stoke's law [47,48].

Freeness measurement, which gives a measure of the specific surface and the compressibility of pulp, can be simply used for determining the specific surface, if the compressibility of the pulp remains the same [50]. Freeness has been found to correlate well with the specific surface area [22,40], but it does not reflect this relationship constantly, because the specific surface varies even at the same freeness value [51].

The water retention value (WRV) by centrifugation is normally used to measure the swelling of pulp. A modified WRV was used to evaluate external fibrillation. WRV is the sum of the extra-fiber bonded water fraction (WRV_E) and the intra-fiber retention water fraction (WRV_I). WRV_E obtained by subtracting WRV_I from WRV can be used to evaluate external fibrillation [52,53].

External fibrillation has been observed qualitatively using a microscope such as a light microscope [1,2,3,10,22,54], electron microscope [6,16,17,21,22,55,56] and confocal laser scanning microscope [57,58]. Wet fibers can be used for observation with a light microscope and confocal laser scanning microscope, but for examination with an electron microscope the external fibrils attached to the fibers should be dried down onto the fiber surface during sample preparation, which is not favorable for the observation. A special treatment such as hexamethyldisilazane treatment [59], critical point drying [6,56,60] or freeze drying [60] can prevent this problem. Selective staining of external fibrils using a metal colloid makes it possible to observe the external fibrils in the cross-section of the sheet [21,55]. Some attempts using microscopic observation have been made to quantify the degree of external fibrillation by calculating the pixel size of external fibrils over the whole fiber area [58,61,62]. Laamanen measured the degree of external fibrillation by counting the occurrence of external fibrillation along the fiber length under a light microscope [63]. A more simplified method was used by Koljonen and Heikkurinen to classify fibers into “not fibrillated” and “fibrillated”, after examining fibers under a light microscope [64].

Although many attempts have been made, none of these techniques seem to have been widely accepted for measuring the degree of external fibrillation.

4. EXPERIMENTAL

4.1 Pulp used

For chemical pulp, 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 and used for the experiments. The pulp was elemental chlorine free (ECF) bleached.

For mechanical pulp, thermomechanical pulp (TMP) with a freeness of 545 ml was taken from the reject line of a Finnish pulp mill. The pulp, made from Norway spruce (*Picea abies*), was supplied at 40 % dry solids and stored at -25 °C before use.

4.2 Method controlling fibrillation

The ultra-fine friction grinder (Supermasscolloider®, Model MKZA 10-15J, Masuko Sangyo Co.Ltd., Japan) shown in Fig. 4.1 (a) - (b) consists of two grinding stones with a diameter of 250 mm: a lower rotating one and an upper stationary one. The grinder has a nominal motor power of 11kW. The gap clearance between the two stones can be adjusted by operating a handle which moves the rotating stone vertically in 20 µm increments. Before feeding the pulp, the gap should be large enough so that no contact occurs between the two grinding stones. As soon as the pulp has been fed into the large enough gap, the gap is reduced to the desired level. The pulp passing through the gap between the stone plates is subjected to compressive and shear forces. The gap between the plates varies between 0 and 250 µm. A smaller gap results in closer contact. The grinding stones are composed of either Al_2O_3 (grit class 80) or SiC (grit class 46). The surface of the outer periphery of the stone is rather flat and the tapered inner part is grooved, which allows the gap to be narrowest at the outlet of the grinder and widest at the inlet or inner part. This is shown in Fig. 4.1 (b). Water can be circulated through the cooling chamber to cool down the grinding zone. Treated pulp is discharged by centrifugal force and can be recirculated manually to the inlet of the grinder, if necessary. The rotating speed can be raised to about 3000 rpm with a frequency converter.

Fig. 4.1 (c) shows the surface of the grinding stone. The surface image was scanned using a Form Talysurf Series 2 (Taylor Hobson Ltd., England). It may be useful to compare the scanned images for evaluating the changes in the degree of wear of the stone surface.

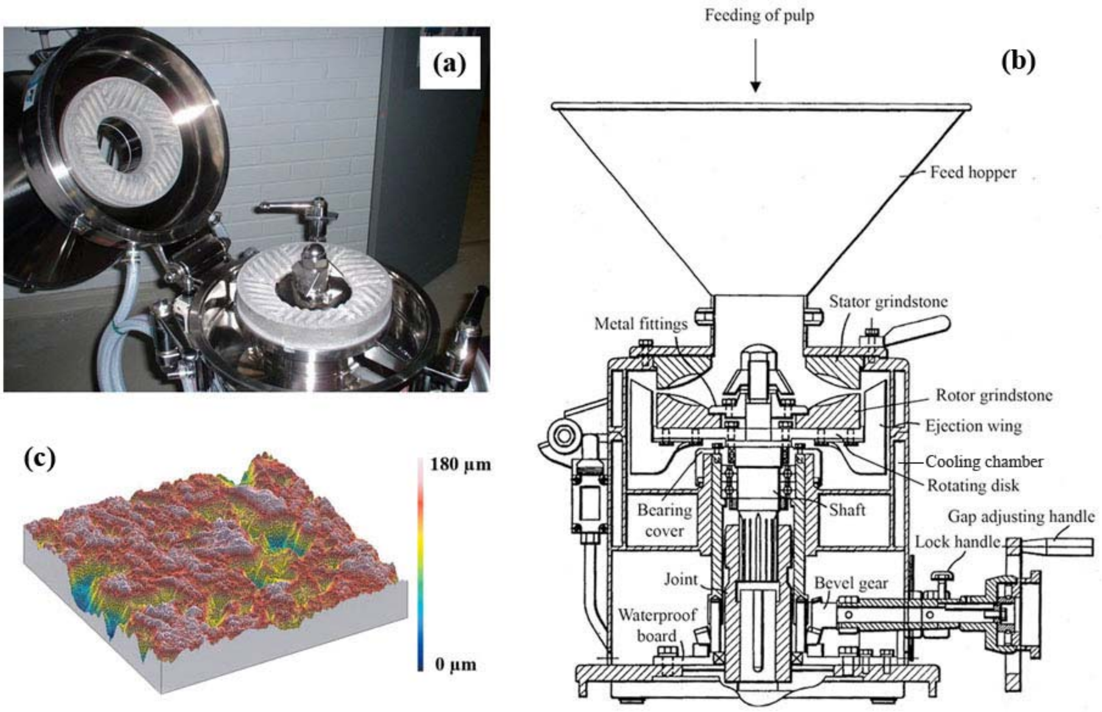

Fig. 4.1 An ultra-fine friction grinder (a) [I], longitudinal sectional view (b) [65] and surface of stone plate of grit class of 46, Scan size 2 mm x 2 mm (c) [I].

The pulp was disintegrated for 10 min, and it was treated in an ultra-fine friction grinder under the conditions shown in Table 4.1.

Table 4.1 Conditions for mechanical treatment in ultra-fine friction grinder.

Variables	Conditions
Gap (µm)	60-250
Rotating speed (rpm)	900 & 1500
Initial pulp consistency (%)	3 - 6
Grit class	46 (grit size 297-420 µm) & 80 (grit size 149-210 µm)

Pulps beaten separately in a Lampén mill and a Valley beater were used as reference pulps for the purpose of comparison.

4.3 Evaluation of fibrillation

The degree of internal fibrillation was measured as the fiber saturation point (FSP) with solute exclusion [66] using a 2×10^6 Dalton dextran polymer (Amersham Biosciences AB, Uppsala, Sweden). The WRV was also used for mechanical pulp in addition to FSP measurement.

The measurement of the external fibrillation index was based on analysis of images acquired from phase contrast microscopy. The phase contrast technique provides a clear differentiation between the fiber body, external fibrils and surrounding medium. At least 20 images, each containing several fibers from each sample, were taken using a phase contrast microscope (DM LAM, Leica) with an NPLAN 10xPH1 objective. The microscope was equipped with a Leica DC300 digital camera. Image processing using the Matlab software was then carried out as follows: The RGB images were converted into gray-scale, and a specially developed image processing algorithm was used to distinguish the pixels belonging to the background, fiber bodies and external fibrils from each other. Detected pixels of fibers and external fibrils were used to calculate the external fibrillation index expressed in percent, as the ratio of the sum of external fibril pixels to the sum of the fiber body pixels. The procedure is described in more detail in a separate publication [62]. In addition to this quantitative method, qualitative observation of external fibrillation was also made using a scanning electron microscope (SEM, DSM 962, Zeiss) [II, IV, V, VI].

Fig. 4.2 shows the degree of internal and external fibrillation developed in a Lampén mill or Valley beater, and measured with the methods mentioned above. As expected, internal and external fibrillation develop simultaneously in the Valley beater, while only internal fibrillation develops in the Lampén mill. This result validates the method measuring the degree of external fibrillation used in this study.

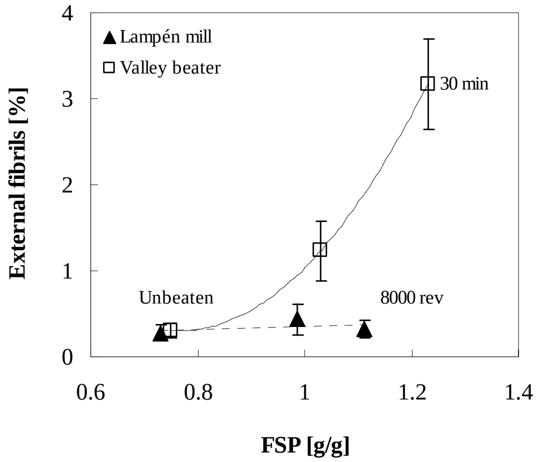

Fig. 4.2 Fibrillation of once-dried bleached kraft softwood pulp developed in a Lampén mill and Valley beater. R100 fraction was used for the measurement. Error bars represent the 95% confidence interval. Redrawn from [II,VI].

4.4 Evaluation of pulp and paper properties

The detailed methods for measuring the properties of pulp and paper can be found in the enclosed publications [I-VI].

5. RESULTS AND DISCUSSION

5.1 Effect of gap clearance on fibrillation

The gap clearance of the grinder was found to be an important variable in controlling the fibrillation of fibers, as shown in Fig. 5.1. The amount of fines generated from the grinding action varies with the gap, and is closely related to the internal and external fibrillation of fibers. At a smaller gap, a larger quantity of fines can easily be generated within several pulp passages, which makes it possible to study fines in detail [III] and also to produce either micro-fibrils or nano-fibrils [67,68,69].

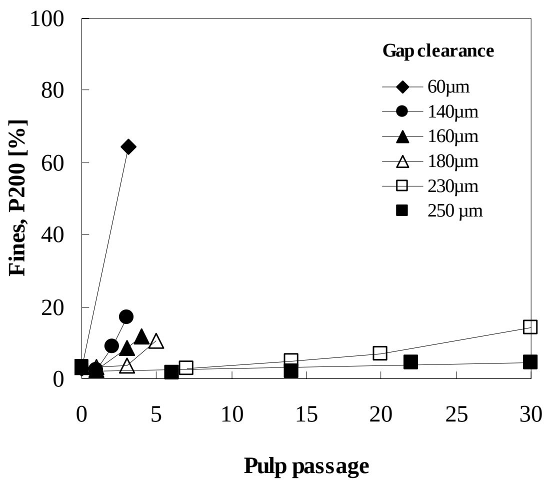

Fig. 5.1 Effect of gap clearance on fines formation. Pulp passage is defined as the number of pulp recirculations through the gap. Once-dried bleached kraft pulp with 6% consistency treated at a rotating speed of 1500 rpm [I].

Fig. 5.2 shows the effect of gap clearance on the internal fibrillation of fibers. In the same way as fines formation, also internal fibrillation varies with the gap. As the gap decreases, the FSP increases rapidly, while at a larger gap (230 - 250 µm) the FSP reaches a plateau. Fibers in the plateau seem to continue to promote external fibrillation, and this plateau is a focus area in this study, because it allows examining the effect of external fibrillation on various pulp and paper properties.

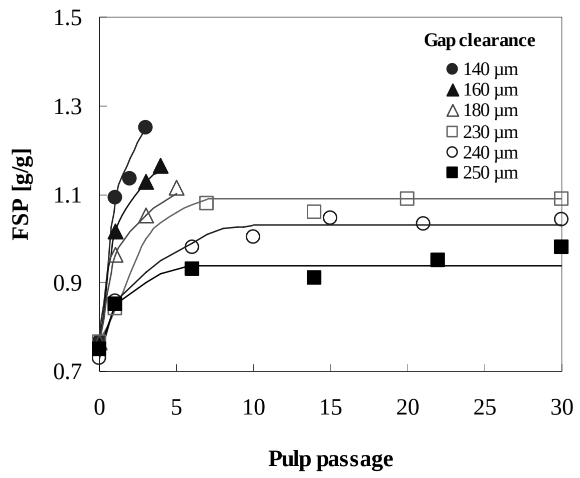

Fig. 5.2 Effect of gap clearance on internal fibrillation of fibers (R100 fraction). FSP of the reference pulp: 1.03 g/g for 10 min, 1.23 g/g for 30 min and 1.42 g/g for 60 min beaten in a Valley beater. Once-dried bleached kraft pulp with 6% consistency treated at a rotating speed of 1500 rpm. Redrawn from [I,II,VI].

The increase in the degree of external fibrillation shown in Fig. 5.3 supports what happens in the plateau of the FSP curve shown in Fig. 5.2. External fibrillation continues to increase while internal fibrillation remains constant.

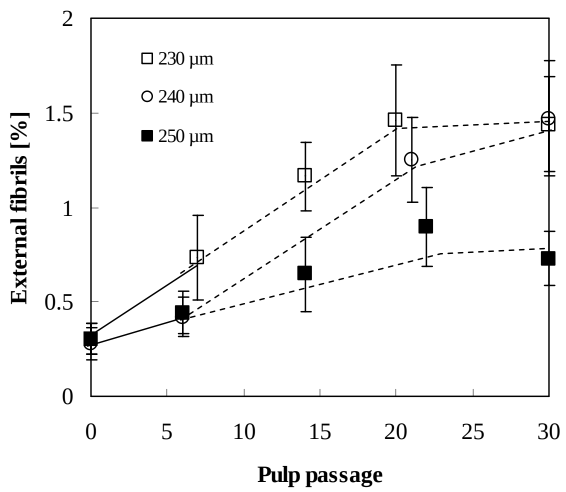

Fig. 5.3 Effect of gap clearance on the degree of external fibrillation of once-dried bleached kraft softwood pulp (R100 fraction). The solid line represents the region where internal fibrillation is dominant, the dotted line the region where external fibrillation is dominant. This is also shown in some other figures in this thesis. Redrawn from [II,VI].

External fibrillation induced by the grinder appeared at the beginning of beating, and it was accompanied by internal fibrillation and fines formation. It has been suggested that external fibrillation is a natural consequence of swelling [1]. In other words, no external fibrillation can occur without developing internal fibrillation. When pulp is beaten in non-swelling liquids such as fuel oils and butyl alcohol, fibers are cut and not externally fibrillated [70]. This is explained by the fact that fibers become soft with swelling, and beating fibrillates the softened fibers, while without swelling, fibers are stiff and brittle, so the beating action would result in sharp cutting. Therefore, swelling seems to be prerequisite for developing external fibrillation. For mechanical pulp, the external fibrillation mechanism is different from that of chemical pulp. Swelling may not be needed to allow external fibrillation.

When pulp passes through the gap, the consistency inside the gap will increase, because the water in the pulp suspension is squeezed out and the pulp consistency between the plates increases. Pulp is then compressed and sheared in the gap, and the shear forces between fibers become dominant, which may cause external fibrillation. As the gap decreases, the pulp consistency in the gap will further increase and only a limited number of fibers will be trapped in the gap. As a result, the friction between the grit and fibers becomes dominant and will produce mainly fibrils. External fibrillation can occur without any mechanical treatment such as ultrasonic treatment [71,72]. Weak points at the fiber surface were found to be favorable for external fibrillation in ultrasonic treatment. The partial removal of the outer layer is likely to be favorable for developing external fibrillation with a larger gap in the grinder.

Fig. 5.4 shows a light microscopic image of the development of external fibrillation with an increased number of pulp passages. External fibrillation was found to develop well in the later stage of beating in the grinder, while internal fibrillation, as shown in Fig. 5.2, increases rapidly in the early stage of beating.

Fig. 5.4 Light microscopic observation of external fibrillation of fibers (R100 fraction). Once-dried bleached kraft softwood pulp with 5% consistency treated at the gap of 240 µm in the grinder. Scale bar of 50 µm. [V].

External fibrillation correlates well with fines formation in the Valley beater, while in the grinder it does not increase despite a doubling in the amount of fines formed in the later stage of grinding at a gap of 230 µm [II]. This is probably due to the different mechanisms of fines formation in these two mechanical treatments. The mechanism of fines formation in the grinder was found to be different from than in a Valley beater, as shown in Fig. 5.5. In the grinder, mainly surface shear seems to cause external fibrillation and fines formation [73], rather than fiber straightening. In a Valley beater, cyclic compressions under the metallic bar seem to cause fiber straightening and swelling through internal delamination of the fiber wall [73,74]. This may be the reason why the grinder produces more fines with a given longer fiber fraction. However, it would be beneficial to promote external fibrillation, while producing a lower amount of fines [5].

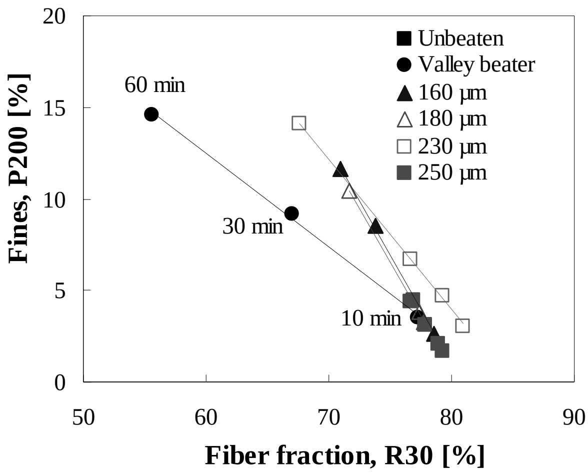

Fig. 5.5 Fines generation in Valley beater and grinder. Once-dried bleached kraft softwood pulp with 6% consistency treated at rotating speed of 1500 rpm [I].

To be able to evaluate the role of external fibrillation, other properties such as internal fibrillation and fiber curl should remain constant during refining in the grinder. No changes in internal fibrillation were found in the plateau of the FSP curve as shown earlier in Fig. 5.2. Fiber curl seems to undergo hardly any change during mechanical treatment in the grinder, while fibers are more straightened in the Valley beater, as shown in Table 5.1. It was also found that fiber curl changes less with a larger gap clearance. A curl index of about 10 % is considered to represent straight fibers, an index of about 20% curly fibers [75]. The changes in fiber curl induced by the grinder can be considered as a minor change during mechanical treatment.

Table 5.1 Changes in curl index of once-dried bleached kraft softwood pulp (R100 fraction) with beating in the grinder and a Valley beater. Rewritten from [I,II,V].

Pulp passage (160 μm)	Curl (%)	Pulp passage (180 μm)	Curl (%)	Pulp passage (230 μm)	Curl (%)	Pulp passage (240 μm)	Curl (%)	Pulp passage (250 μm)	Curl (%)	Valley beating (min)	Curl (%)
0	22.4	0	22.4	0	22.4	0	22.4	0	22.4	0	22.4
1	22.1	1	23.2	1	22.1	1	22.8	1	22.5	10	18.4
2	-	2	21.5	7	21	6	22.1	6	21.6	30	17.5
3	19.8	3	-	14	21.1	14	21.3	14	21.6	60	16
4	19.6	4	20.6	20	20.9	22	20.7	22	20.6		
5	-	5	20.6	30	19.7	30	-	30	20.2		

The morphological changes in fibers with the progress of grinding, based on the results obtained, can be summarized as shown in Fig. 5.6. With the larger gap, internal and external fibrillation increase with fines formation in the beginning of the treatment. As the grinding continues, external fibrillation continues to increase and much more fines are formed, while internal fibrillation remains constant. With the smaller gap, fines are generated in the beginning of the treatment, so no fiber shapes will remain.

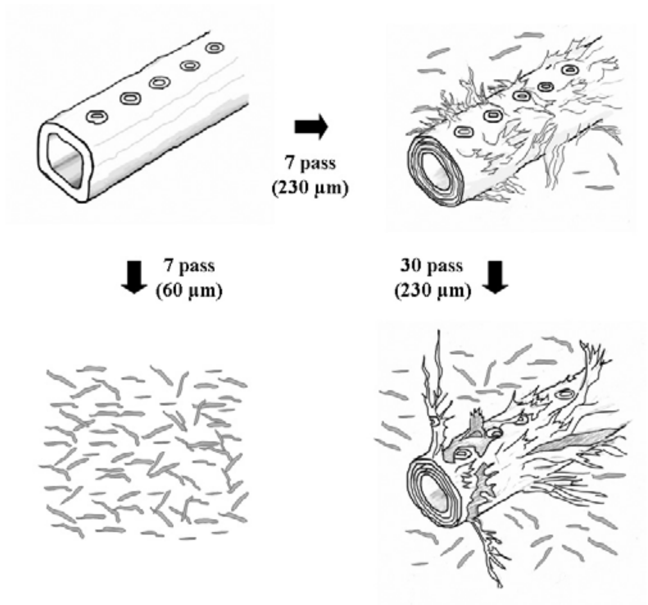

Fig. 5.6 Morphological changes in fibers with the progress of grinding.

The fibrillation of mechanical pulp is different from that of chemical pulp. Internal fibrillation develops well with beating in chemical pulp fibers, while in mechanical pulp fibers it is restricted by the lignin-rich structure of the fiber wall [66,76]. Neither the compressive forces generated in a Lampén mill nor the shear forces generated in the grinder promote the internal fibrillation of mechanical pulp fibers, as shown in Fig. 5.7. The WRV shows a higher value than the FSP, which may be due to external fibrils between the fibers. This overestimation of the cell wall water measured by the WRV was also shown by Maloney et al. [77].

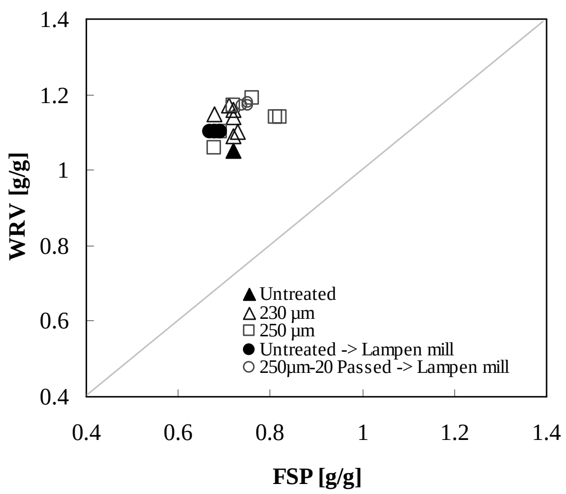

Fig. 5.7 Internal fibrillation of TMP long fiber fraction (R50). Pulp treated up to 20 passages in the grinder and 8000 revolutions in Lampén mill [IV].

The changes in the weights of pulp fractions caused by mechanical treatment using the grinder and the Lampén mill are shown in Table 5.2-5.3. Shear forces generated in the grinder increased the fines content (P200), probably due to the fibrillation of the fiber surface, while the compressive force generated in the Lampén mill had little effect on the fines content. Shives were effectively disintegrated into fibers in the grinder, while they remained unchanged in the Lampén mill.

Table 5.2 Properties of TMP treated in grinder. Pulp with 5% consistency treated at a rotating speed of 1500 rpm [IV].

Gap, µm	Pulp passage	CSF, ml	Bauer McNett fractions, %			Shives, %
			R50	R200	P200	
	0	545	84.9	8.8	6.3	4
230	1	451	81.5	9.6	8.9	2.8
	3	339	-	-	-	-
	6	214	-	-	-	0.7
	10	118	68	12.6	19.4	0.5
	15	78	-	-	-	-
	20	47	61.3	13.9	24.8	0.1
250	1	489	82.7	9.1	8.2	3.1
	3	401	-	-	-	-
	6	267	-	-	-	1.4
	10	167	71.7	11	17.3	1.1
	15	110	-	-	-	-
	20	80	67.7	12.1	20.2	0.6

Table 5.3 Properties of TMP treated in Lampén mill. 3% consistency used [IV].

Rev	CSF, ml	Bauer McNett fractions, %			Shives, %
		R50	R200	P200	
0	545	84.9	8.8	6.3	4
2000	505	86.6	8.9	4.5	2.9
5000	508	-	-	-	-
8000	492	81.3	9.1	9.6	3.5
0*	80	67.7	12.2	20.2	-
2000	76	67.1	13.2	19.6	-
5000	74	-	-	-	-
8000	69	64.4	14.4	21.2	-

** Pulp treated with 20 passages at the gap of 250 µm in the grinder.*

The degree of fibrillation seemed to be sensitive to changes in the gap clearance between the plates. A smaller gap in the grinder was found to produce more external fibrillation of fibers, also resulting in more fines.

The results reported in this chapter prove the suitability of the grinder for the purposes of this study, i.e. for examining the role of external fibrillation in pulp and paper properties.

5.2 Effect of external fibrillation on the retention of filler

The retention of filler during sheet forming in the paper machine is critical for the efficiency of papermaking. To be able to control the retention more effectively, the role of fiber properties in the interaction of filler particles during sheet forming needs to be known in detail. The interaction between fibers with different degrees of external fibrillation and filler was evaluated during sheet forming in a high-vacuum dewatering device, a moving belt former (MBF), which simulates the dynamic dewatering phenomena in the wire section of a paper machine [78].

Fig. 5.8 shows the effect of external fibrillation of fibers on the retention of filler in the absence and presence of cationic polyacrylamide (C-PAM). Filler retention in the absence of C-PAM is not influenced by the degree of external fibrillation of fibers under high-vacuum dewatering, whereas colloidal interaction caused by C-PAM enhances filler retention with an increased degree of external fibrillation of fibers. In the absence of cationic polymer, the deposition of a filler particles on the fiber surface is due to attractive electrostatic and van der Waals forces. The distance between the filler particle and fiber surface is not short enough for such colloidal forces to become important. In the presence of cationic polymer, the gap between the filler particle and fiber can be bridged by cationic polymer, so the electrostatic repulsion is reduced [79, 80]. This explains well the difference in filler retention in the absence and presence of C-PAM, as shown in Fig. 5.8. As the pulp passage increases, filler retention increases in the presence of C-PAM, while it remains constant in the absence of C-PAM. Previous studies [81, 82, 83] have shown that beating increases the deposition of filler particles on the fiber surface due to an increase in the external surface. However, the refiners used in these studies were conventional laboratory refiners in which the external surface will increase at the same time with internal fibrillation. The results did not reflect the relative role of internal and external fibrillation for filler retention. In the present study, internal fibrillation was kept constant during pulp passages 6 and 22, while external fibrillation was increased. Therefore, the increased number of external fibrils can be the main reason for adsorption of precipitated calcium carbonate (PCC) particles onto them. The increase in external fibrils with beating increases the external surface area and amount of ionizable groups for improving filler adsorption [81]. The structural properties of the fiber surface, such as the degree of external fibrillation, are important variables in controlling filler retention.

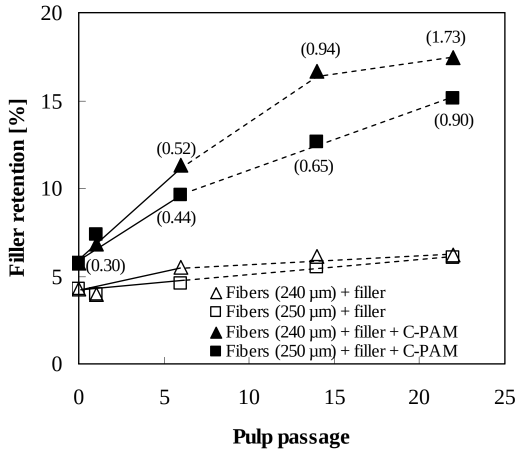

Fig. 5.8 Effect of external fibrillation of fibers (R100 fraction) on the retention of filler. Once-dried bleached kraft softwood fibers used. PCC used as a filler. Numbers shown in the figure indicate the degree of external fibrillation. Redrawn from [V].

The important role of external fibrils attached to the fibers for the interaction with filler particles is shown in Fig. 5.9. More filler is seen where more fibrils are available.

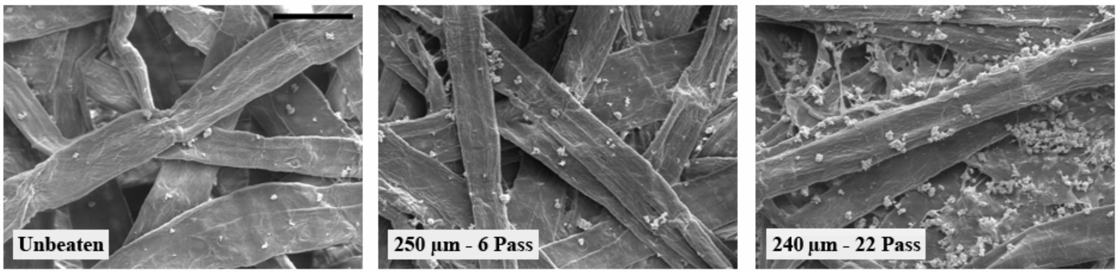

Fig. 5.9 SEM images of filler retained in the fiber network. Once-dried bleached kraft softwood fibers used. 30 % filler addition. Scale bar of 50 μm [V].

5.3 Effect of external fibrillation on paper properties

The collapsibility and conformability of fibers caused by internal fibrillation play an important role in the development of sheet density, shown as a solid line in Fig. 5.10, while an increase in the degree of external fibrillation, shown as a dotted line, increases the density slightly. The external fibrils attached to fibers can be expected to increase Campbell's forces, resulting in improved consolidation of the sheet, as suggested by Giertz [3,84].

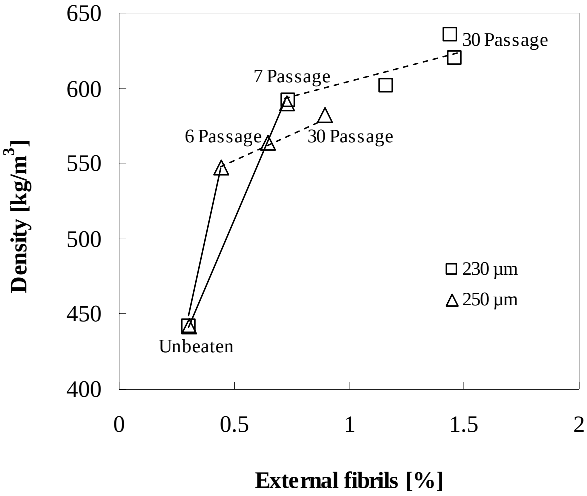

Fig. 5.10 Effect of external fibrillation of once-dried bleached kraft softwood fibers on sheet density. R100 fraction used for preparing a sheet. Redrawn from [II].

Fig. 5.11 shows the effect of external fibrillation on the light scattering coefficient. An increase in the degree of external fibrillation decreases the light scattering coefficient slightly. Similar results were shown by Retulainen [30], according to which light scattering decreases slightly when kraft fines are added to the long-fiber fraction of kraft pulp. In mechanical pulp, the increase in external fibrillation was found to increase the light scattering slightly [IV]. External fibrils still attached to the fiber seem to act in the same way as pulp fines.

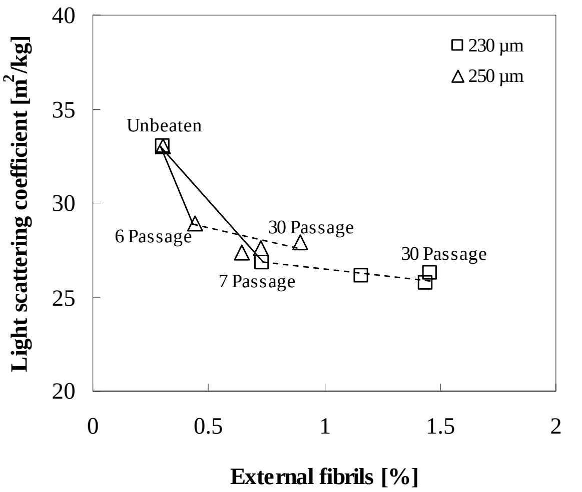

Fig. 5.11 Effect of external fibrillation of once-dried bleached kraft softwood fibers on light scattering coefficient. R100 fraction used for preparing a sheet. Redrawn from [II].

The linear relationship between the light scattering coefficient and density of both Valley-beaten and ground pulp is shown in Fig. 5.12. As the density increases, the difference in light scattering coefficient between Valley-beaten pulp and ground pulp becomes bigger. At the higher density, the light scattering coefficient of pulp treated in the grinder is higher. External fibrils still attached to the fibers might provide an open structure for scattering light. This is one of the potential roles of external fibrillation in promoting higher light scattering coefficient at a certain density.

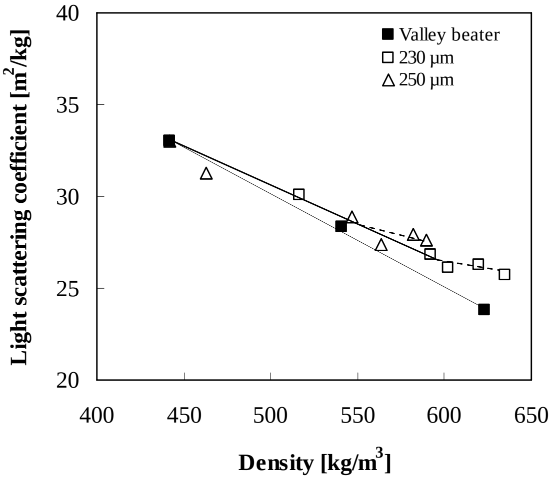

Fig. 5.12 Effect of external fibrillation on the light scattering coefficient – the tensile combination. R100 fraction of once-dried bleached kraft softwood fibers used for preparing a sheet. Redrawn from [II].

It is apparent that the degree of internal fibrillation strongly promotes paper strength properties, such as tensile index and internal bond strength, which are shown in Fig. 5.13 and Fig. 5.14. However, the strength of pulp treated at both 230 µm and 250 µm in the grinder is further increased by promoting external fibrillation, with the tensile index increasing by about 20 %, as shown in Fig. 5.13. This result does not agree with Hartman's work [6], according to which external fibrillation does not increase tensile strength in spite of increased sheet density. Hartman prepared a sheet by adding externally fibrillated fibers to unrefined fibers but did not report how much externally fibrillated fibers that were added, while in the present study 100 % externally fibrillated fibers were used to prepare the sheet. In a sheet consisting of 100 % externally fibrillated fibers, the bonding layer formed by external fibrils might be distributed more uniformly within the fiber network, which results in increased tensile strength.

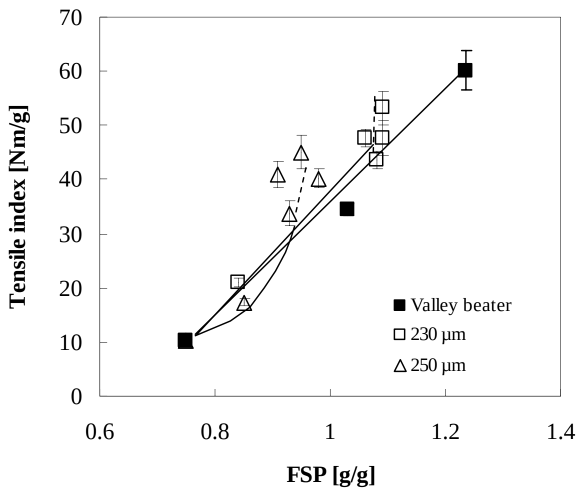

Fig. 5.13 Effect of internal fibrillation and external fibrillation on tensile strength of long-fiber fraction (R100) beaten in a Valley beater and an ultra-fine friction grinder. Once-dried bleached kraft softwood fibers used for preparing a sheet. 95 % confidence interval limits are included [II].

Fig. 5.14 shows the relationship between the external fibrillation of fibers and the internal bond strength of a sheet. The strength was increased by 33% and 46% for pulp treated at 230 μm and 250 μm, respectively, by promoting external fibrillation. The increase in internal bond strength caused by external fibrillation is more pronounced than the increase in tensile strength. Therefore, it is evident that external fibrils contribute more to internal bond strength than tensile strength. Fibers treated in the grinder show higher strength than Valley-beaten fibers at a given degree of internal fibrillation. One of the reasons is probably that the quality of fibrils attached to fibers treated in the grinder differs from that of fibrils attached to Valley-beaten fibers. As shown in Fig. 5.5, more fines are formed with the ground pulp than the Valley-beaten pulp. More fines from the outer wall layer might be more homogeneously removed by grinding, and the remaining external fibrils attached to the fiber are likely of better quality for bonding.

The FSP is linearly correlated with the water content of wet pulp after pressing [85]. In other words, the higher the FSP, the more difficult it is to remove water. The increase in tensile strength and internal bond strength at a certain FSP, as shown in Fig. 5.13 and Fig. 5.14, can be seen to represent the potential role of external fibrillation in achieving a better combination of dewatering and paper strength.

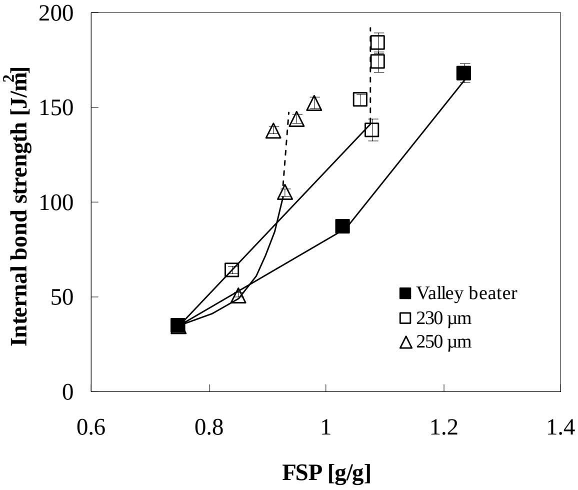

Fig. 5.14 Effect of internal fibrillation and external fibrillation on internal bond strength of long-fiber fraction (R100) beaten in a Valley beater and an ultra-fine friction grinder. Once-dried bleached kraft softwood fibers used for preparing a sheet. 95 % confidence interval limits are included [II].

An increase in external fibrillation by 0.3-0.7 % increased the tensile strength by about 20 % and internal bond strength by 46 %. Retulainen [30] has shown that adding 3 % kraft fines to a kraft long fiber fraction increases the tensile strength by 18 % and internal bond strength by 45 %. Therefore, the effect of a 3 % fines addition seems to be equivalent to that of 0.3-0.7% increase in external fibrillation. The addition of fines seems to compensate for the role of external fibrillation for paper strength, but as the amount of fines to be added increases, some limitations might occur in the retention of fines and their distribution in the fiber network. Instead of changing the amount of fines to be added, changing the degree of external fibrillation would be beneficial to overcome these limitations, if external fibrillation occurs with minimizing fines formation. The grinder used in this study increased the external fibrillation and produced a great amount of fines, but still allowed studying the effect of external fibrillation alone on paper strength.

The shear forces have been found to remove shives and to increase the external fibrillation of fibers. To eliminate the effect of shives on paper strength, a shive-free sheet made from the P14/R50 fraction of TMP was evaluated, as shown in Table 5.4. The increase mostly in external fibrillation caused by the shear forces further increases the density of the sheet, resulting in improved tensile and internal bond strength. External fibrillation of mechanical pulp therefore appears to be an important factor in controlling the quality of mechanical pulp fibers.

Table 5.4 Properties of a sheet made from P14/R50 fraction of TMP. Pulp with 5% consistency treated at a rotating speed of 1500 rpm in the grinder. 95 % confidence interval limits are included [IV].

Gap, μm	Pulp passage	Density, kg/m^3	Tensile index, Nm/g	Scott bond, J/m^2
	0	218 ± 3	11.2 ± 0.3	40.1 ± 0.9
230	1	212 ± 2	9.4 ± 0.4	42.2 ± 2.9
	6	249 ± 3	12.6 ± 0.5	47.9 ± 2.7
	10	265 ± 3	15.7 ± 0.6	47.9 ± 2.2
	15	294 ± 2	19.2 ± 0.4	50.0 ± 1.4
250	1	209 ± 3	8.9 ± 0.4	40.1 ± 1.8
	6	230 ± 1	11.4 ± 0.4	42.9 ± 2.3
	10	245 ± 2	12.5 ± 0.3	43.7 ± 2.3
	15	269 ± 2	15.3 ± 0.4	49.2 ± 2.3

5.4 Effect of external fibrillation on recycling potential

Fiber swelling plays a major role for the development of paper strength, as shown in the previous chapter, but its effect is reduced in the recycling process by hornification [86,87]. The swelling of internally (8000 rev in Lampén mill) and externally fibrillated fibers (pulp passage 21 and 30 in the grinder) is reduced with recycling to a similar level, and the loss in fiber swelling can be restored to some extent by refining in a Lampén mill. This is shown in Fig. 5.15. The preparation of internally and externally fibrillated fibers is described in [VI]. For externally fibrillated fibers, the swelling was restored more than for the others, even beyond the swelling of nonrecycled fibers.

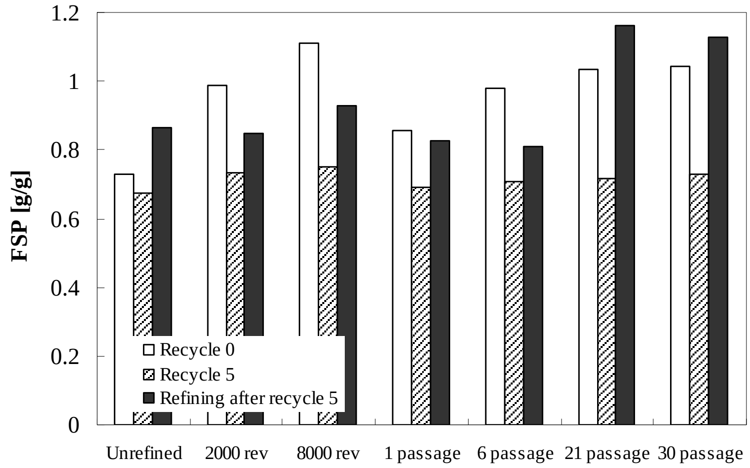

Fig. 5.15 Effect of recycling and refining on internal fibrillation of fibers. Once-dried bleached kraft softwood fibers (R100 fraction) were refined in a Lampén mill and a grinder before recycling. Redrawn from [VI].

Fibers refined in a 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 nor by refining after recycling. This is shown in Fig. 5.16. 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 [VI].

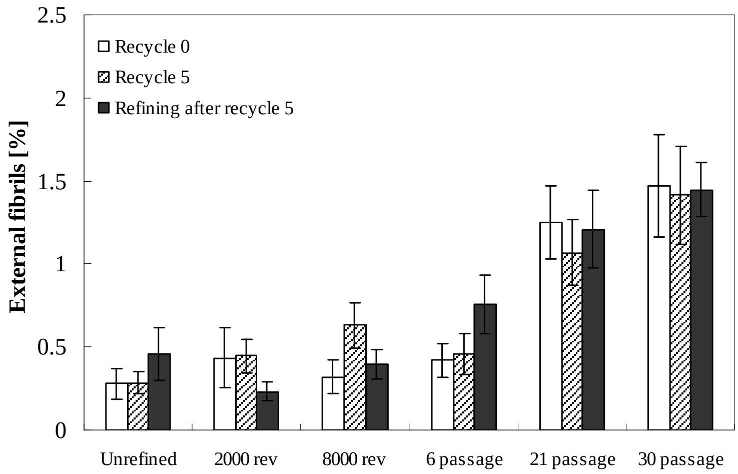

Fig. 5.16 Effect of recycling and refining on external fibrillation of fibers. Once-dried bleached kraft softwood fibers (R100 fraction) were refined in a Lampén mill and a grinder before recycling. Error bars represent the 95% confidence interval. Redrawn from [VI].

Refining of the five-times recycled fibers does not seem to change the fiber length, except for the externally fibrillated fibers as shown in Table 5.5.

Table 5.5 Effect of refining on fiber length [VI].

	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

(mm)

Fig. 5.17 shows the changes in tensile strength during recycling and refining. The tensile strength of externally fibrillated fibers is similar to that of internally fibrillated fibers at recycle 0. As the number of recycles increases, the tensile strength of internally fibrillated fibers decreases more than that of externally fibrillated fibers. 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 beyond the strength of nonrecycled fibers. This may be expected from increased fines formation, as shown in Table 5.5, but the FSP with and without fines for refined pulp after 5 times' recycling shows no difference. The FSP was 1.16 g/g for the pulp passage 21 and 1.13 g/g for pulp passage 30. The FSP of fibers without fines was 1.14 g/g for pulp passage 21 and 1.13 g/g for pulp passage 30. Therefore, according to this study, the swelling of fines does not seem to be restored by refining. The recovery of tensile strength may be due to the better swelling ability and straightening of externally fibrillated fibers with recycling.

The changes in internal bond strength during recycling and refining are similar to the change in tensile strength, which is shown in Fig. 5.18. The potential for restoring the internal bond strength of the fibers becomes more pronounced with refining. The curl index of all pulps remains constant during the recycling process. Rather curly fibers can be straightened by refining, and externally fibrillated fibers were found to be much more straightened than the others [VI].

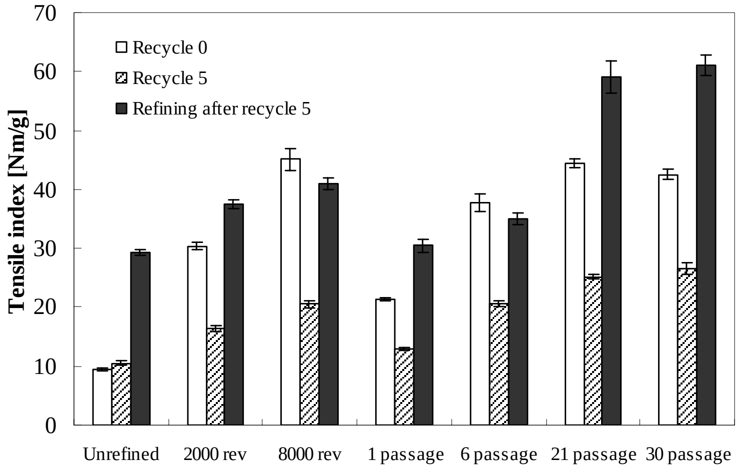

Fig. 5.17 Effect of recycling and refining on tensile strength of paper. Once-dried bleached kraft softwood fibers (R100 fraction) used for recycling, refining and preparing a sheet. Error bars represent the 95% confidence interval. Redrawn from [VI].

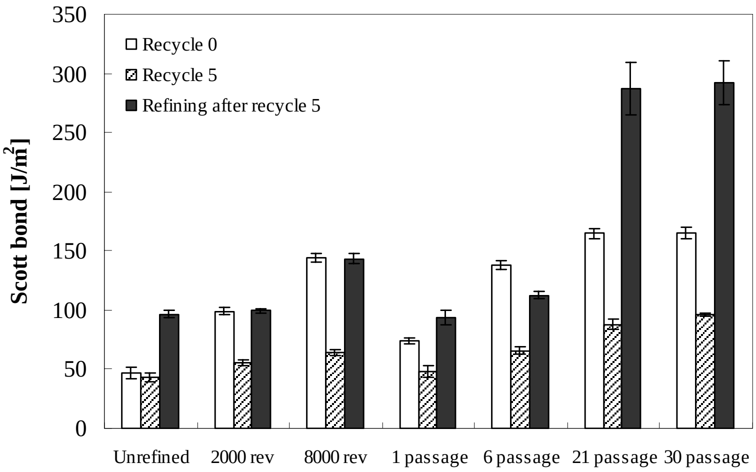

Fig. 5.18 Effect of recycling and refining on internal bond strength of paper. Once-dried bleached kraft softwood fibers (R100 fraction) used for recycling, refining and preparing a sheet. Error bars represent the 95% confidence interval. Redrawn from [VI].

Fig. 5.19 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 show the external fibrils between the 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, which may contribute to the paper strength differently, as shown in Fig. 5.17-5.18.

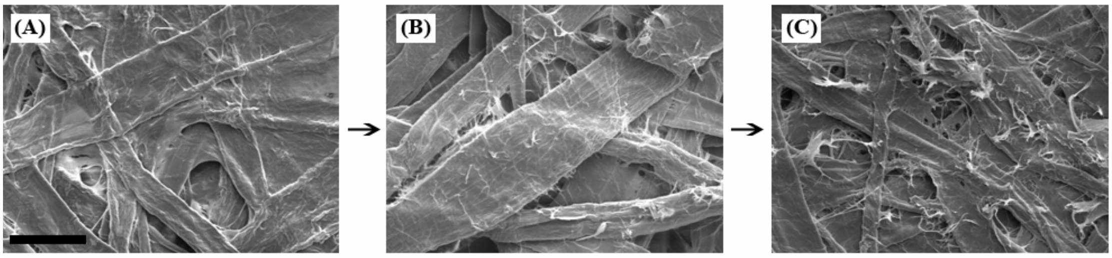

Fig. 5.19 SEM micrographs of a handsheet surface made from nonrecycled fibers (A), five-times recycled fibers (B) and five-times recycled, refined fibers (C). The pulp was recirculated 30 times in the grinder before recycling. Once-dried bleached kraft softwood fibers (R100 fraction) used for recycling, refining and preparing a sheet. Scale bar of 50 μm [VI].

6. CONCLUSIONS

The objective of the study was to evaluate the role of external fibrillation in pulp and paper properties. The capacity of conventional refiners, such as a Valley beater and a Lampén mill, for achieving this objective was found to be limited by the occurrence of simultaneous changes in a variety of fiber properties, or the absence of changes in external fibrillation. Therefore, a special type of refiner had to be found for the purposes of this study. An ultra-fine friction grinder was found to promote external fibrillation, while keeping other properties constant, which made it possible to evaluate the role of external fibrillation for pulp and paper properties. By using a larger gap, 230-250 µm, in the grinder, the degree of external fibrillation of chemical pulp fibers was promoted, while other properties, such as internal fibrillation and curl, were kept constant. This was achieved after the pulp passage of about 6. In mechanical pulp, the shear forces generated in the grinder at the gap of 230-250 µm fibrillated fibers externally throughout the pulp passages. Fines formation, however, was inevitable throughout the grinding.

Promoting external fibrillation of chemical pulp fibers (R100 fraction) while keeping internal fibrillation constant was found to increase tensile strength and internal bond strength by 20% and 46%, respectively, whereas light scattering was slightly decreased. The increase in both the paper strength properties at a certain FSP might imply a better combination of dewatering and paper strength. The light scattering coefficient of pulp treated in the grinder was higher than that of Valley-beaten pulp at a certain density. The role of external fibrillation of mechanical pulp fibers for the development of paper strength was found to be similar to that of the fibrillation of chemical pulp fibers.

When fibers with different degrees of internal or external fibrillation were dried and rewetted, the reduction in swelling was found to be at a similar level for both internally and externally fibrillated fibers. However, the paper strength properties, such as tensile strength and internal bond strength, of externally fibrillated fibers were better after recycling than those of internally fibrillated fibers. Hornified fibers can be restored to some extent by refining. Externally fibrillated fibers offer greater potential for restoring swelling and straightening of fibers by refining than internally fibrillated fibers. Refining restored the paper strength properties, such as tensile strength and internal bond strength, of externally fibrillated fibers beyond their original strength.

Increasing the degree of either internal or external fibrillation did not influence the retention of filler in the absence of a chemical retention aid during sheet forming under high-vacuum dewatering in a device which simulates the dynamic dewatering phenomena in the wire section of a paper machine. However, the retention of filler was increased with increased internal fibrillation of fibers in the presence of a chemical retention aid, and was further increased by an increase in the degree of external fibrillation of fibers during sheet forming under high-vacuum dewatering. Increasing the degree of external fibrillation alone without changing the internal fibrillation can be used as means to control the retention of filler even at a commercial scale.

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