Fibre Suspension Flocculation under Simulated Forming Conditions

by

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To my parents and my wife
Copy of hand painting from book, Tiangong Kaiwu, published in 1637 written by Yingxing Song in Ming Dynasty in China illustrating production technology of making bamboo paper.

Photo of flow loop system built by Huawei Yan et al. in 2001 at Royal Institute of Technology in Sweden for study of fibre suspension flocculation under simulated forming conditions.
ABSTRACT

A flow loop system for study of fibre flocculation in suspensions has been developed. The system is designed to simulate the flow conditions in a paper machine headbox. It is equipped with a radial distributor feeding a step diffuser pipe package, after which the flow is contracted in a 2-D nozzle. The flow system is also equipped with a secondary flow contraction with an area reduction ratio of 2:1 after the headbox nozzle, mimicking accelerations that may take place during forming. The flow system is equipped with heating and cooling devices for the study of temperature effects on fibre suspension flocculation. An online dosage device for the study of chemical effects on fibre suspension flocculation is also included. The maximum flow velocity in the system is 16 m/s.

Flowing fibre suspensions were studied using a high speed CCD video camera and transmitted infra-red laser light pulse illumination. Images were taken either separately before and after or along the secondary contraction. Images of fibre flocculation were evaluated by power spectrum analysis, and the mean floc size and the flocculation index were calculated. A concept of mean floc area reduction, based on power spectrum, has been introduced to characterise the fibre network in suspensions. By comparing the fibre flocculation before and after the secondary contraction, or by following the fibre flocs along the secondary contraction, floc rheology information can be obtained. The effects of chemical additives and fibre surface modification can also be studied by comparing the corresponding fibre flocculation.

For a bleached softwood kraft pulp suspension at a fibre concentration of 5 g/l, the fibre flocs along the secondary contraction have been manually evaluated, and the results confirm that the power spectrum analysis is applicable. Ca 1/5 of the flocs were broken into two by the contraction. The mean floc size increases in MD while in CD it decreases during the flow contraction. Both the floc aspect ratio and the floc orientation in MD increase during the flow contraction. The net fibre floc area is decreased and the fibre flocs are concentrated by the flow contraction, which is confirmed by an increase of gray value of the flocs in the light transmission images. The dewatering of fibre flocs may thus have already been started in the suspension before reaching the wires in the forming zone.

Some physical influences on fibre suspension flocculation have been investigated. The results confirm that fibre concentration and fibre length are the dominating factors affecting fibre suspension flocculation. Increasing absolute flow velocity has an insignificant effect on fibre flocs in the flow contraction. Suppressing turbulence, by increasing suspension viscosity via a decrease of medium temperature, shows a clear effect on reducing fibre suspension flocculation.

Some chemical influences on fibre suspension flocculation have also been investigated. A retention aid, flocculant, cationic polyacrylamide, C-PAM, increases fibre suspension flocculation by a bridging mechanism, and a formation aid, class II, anionic polyacrylamide, A-PAM, decreases fibre suspension flocculation by suppressing turbulence. Fibre
suspension flocculation can also be reduced by surface modification with carboxymethyl cellulose, due to a reduction of the friction between fibres. The amount of fibre dispersion depends on the ionic form of the grafted CMC, due to the electrostatic repulsion between negatively charged groups on the grafted CMC moieties. Xyloglucan, a non-ionic polymer, which is strongly adsorbed on cellulosic fibre surfaces, shows a similar influence on reduction of fibre suspension flocculation by decreasing the friction between fibres.

The fibre flocculation data in the flow loop system were also compared with the corresponding paper formation data in the sheets produced on a pilot paper machine, both with and without chemical additives. The results show that the fibre suspension flocculation is well correlated with the paper sheet formation: when the fibre suspension flocculation is increased, the corresponding paper sheet formation deteriorates, especially in the large scale range.

*Keywords*: Flow loop system, fibre suspension, fibre flocculation, fibre floc, fibre concentration, fibre length, floc size, flow contraction, flow acceleration, flow velocity, CCD video camera, laser light pulse illumination, image analysis, power spectrum, wavelet transform, manual evaluation, refining, sheet formation, temperature, suspension viscosity, turbulence suppressing, friction between fibres, retention aid, cationic polyacrylamide, formation aid, anionic polyacrylamide, surface modification, carboxymethyl cellulose, xyloglucan.
LIST OF PUBLICATIONS

This thesis is based on the following papers:

I. **Huawei Yan, Tom Lindström and Bo Norman**
   “A Flow Loop System for Study of Fibre Suspension Flocculation under Simulated Forming Conditions”

II. **Huawei Yan and Daniel Söderberg**
   “Two-Dimensional Continuous Wavelet Transform Analysis for Study of Fibre Suspension Flocculation”

III. **Huawei Yan and Bo Norman**
    “Fibre Floc Behaviour of Softwood Kraft Pulp in Flowing Suspensions under Simulated Forming Conditions”

IV. **Huawei Yan, Bo Norman and Tom Lindström**
   “Effects of Refining on Fibre Flocculation of Bleached Softwood Kraft Pulp Suspensions under Simulated Forming Conditions”

V. **Huawei Yan and Tom Lindström**
   “Some Ways to Decrease Fibre Suspension Flocculation”

VI. **Huawei Yan, Tom Lindström and Maria Christiernin**
    “Some Chemical Technologies for Reduction of Fibre Suspension Flocculation”
Other relevant publications not included in the thesis:


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1. INTRODUCTION

Fibre suspension flocculation phenomena are essentially important for papermaking. They directly affect paper formation, which influences paper strength, paper opacity, paper coating and printing quality, machine runnability and efficiency, and so forth. Fibre properties that relate to formation can be affected by stock preparation, and its influence on paper forming efficiency has been studied by Hallgren and Lindström (1988). Formation and retention are competing in papermaking. On one hand, retention aids deteriorate the formation of paper, which is not wanted. On the other hand, they improve the retention of fines and fillers, which is wanted. How to get both good formation and retention is a vital and unsolved problem in papermaking.

Since the beginning of the 1970s, with the development of different imaging devices and laser light sources, several groups have concentrated their efforts on constructing equipment for the study of fibre suspension flocculation. Examples include the studies of fibre flocculation in flow channels, Nerelius et al. (1972), Wågberg (1985), Kaji et al. (1991), in decaying turbulence cells, Kerekes and Schell (1992), Zhao and Kerekes (1993), Kerekes and Schell (1995), and in turbulence generated flow channels, Beghello et al. (1996), Raghem-Moayed and Kuhn (2000), Huber et al. (2004).

Several image analysis methods on fibre suspension flocculation measurement and characterisation have also been developed, such as threshold analysis for floc size distribution measurement, Kaji et al. (1991), Karema et al. (1999), Kellomäki et al. (1999), binarization analysis for fibre network structure characterisation, Onabe et al. (1987), tomography analysis for inter and intra fibre floc concentration estimation, Ringnér and Rasmuson (2000), quantitative analysis for fibre floc surface size distribution measurement, Pierre (2000), and power spectrum analysis for fibre flocculation evaluation, Wågberg (1985), Beghello et al. (1996). Modern wavelet transforms have also been recently introduced into papermaking, in order to describe the paper formation, Keller et al. (1999a; c), Keller et al. (1999b), Bouydain et al. (1999).

In order to explain the physical mechanisms that cause fibres to agglomerate to form flocs in suspensions, several mathematical models of fibre flocculation in suspensions have been introduced, such as fibre flocculation in turbulent flow, Steen (1991), crowding factor concept, Kerekes and Schell (1992), Zhao and Kerekes (1993), flowing suspensions of rigid and flexible fibres, Ross and Klingenberg (1998), and fibre flocs with frictional and attractive interfibre forces in suspensions, Schmid and Klingenberg (2000a; b), Schmid et al. (2000). However, due to the limitation of observation possibilities, fibre flocculation in the forming zone, which is the most essential part in papermaking, has only been studied in a few cases, such as Bergström (2003). What really happens to fibre flocs during forming is still unclear in many respects.

Apart from obvious factors such as fibre concentration, fibre length, and so on, chemical additives can also affect the state of fibre dispersion in suspensions. In modern paper-
making, retention aids are always used to retain fines and fillers in fibre suspensions by different mechanisms, such as patching, bridging, network flocculation, and so forth, Eklund and Lindström (1991). The study of retention aids has also been focused on dual and microparticle systems, such as Wågberg and Lindström (1987c), Swerin and Ödberg (1996), Gruber and Mueller (2001), Huber et al. (2004). Formation aids may be grouped into the following classes: additives increasing the dispersion medium viscosity, Soszynski and Kerekes (1988), Zhao and Kerekes (1993); formation aids: class I, gums and mucilages believed to decrease the coefficient of friction between fibres, De Roos (1958); formation aids: class II, high molecular weight polymers affecting the rheological properties of the suspending media, Wasser (1978), Lee and Lindström (1989). Formation aids are, however, seldom used in commercial paper manufacture to reduce fibre suspension flocculation, because of their negative effects on drainage.

Over the years, the development of more sophisticated cameras and computers has allowed the construction of more advanced equipment. A flow loop system has been constructed during this thesis work for the further study of fibre suspension flocculation. The system is designed to simulate the flow conditions in a modern paper machine headbox, and has a secondary contraction after the headbox nozzle to mimic flow accelerations that may take place during forming. The system is equipped with heating and cooling devices to control the suspension temperature. It is also equipped with a chemicals dosing device, which permits online dosages in the flow loop system. Image sequences of fibre suspension flocculation are taken by a high speed CCD video camera with transmitted infra-red laser light pulse illumination, either separately before and after or along the secondary contraction.

Several evaluation methods for studying the fibre suspension flocculation have also been developed during this thesis work. The traditional power spectrum method has been slightly modified to characterise the overall fibre suspension flocculation, mainly the mean floc size and the flocculation index. Additionally, a concept of mean floc area reduction, based on the power spectrum, has been introduced to characterise the fibre network in suspensions. The modern wavelet transform method has been adapted to describe inner floc structure, and to trace individual fibre flocs along the flow contraction. Moreover, a concept of mean floc structure index, based on the wavelet transform, has been introduced to evaluate fibre floc number in suspensions. Furthermore, manual evaluation of fibre flocs along the secondary contraction has been applied to study the floc behaviour during flow acceleration.

The goal of this thesis work was to investigate both physical and chemical influences on fibre suspension flocculation under simulated forming conditions. Physical influences included fibre concentration, refining effects, absolute flow velocity, and medium temperature. Chemical additives included retention and formation aids, and fibre surface modifications. The vision of this thesis work was to elucidate the fibre floc rheological behaviour in the forming zone, to study the mechanisms of some medium property and fibre surface modifications that prevent fibres to agglomerate in suspensions. The final aim of this thesis work was to develop technologies for paper formation improvement in modern papermaking.
2. FLOW LOOP SYSTEM (PAPER I)

The design of the flow loop system is shown in Figure 2.1. The idea of the design is to simulate realistic headbox flows. The flow loop system includes a pulp storage tank equipped with appropriate agitation and heating and cooling devices, a flow distributor, and a headbox with a step diffuser pipe package (see Figure 2.2). It also includes a secondary contraction (contraction after the headbox nozzle contraction) aimed at studying the fibre floc rheology, mimicking flow accelerations during forming in practical papermaking (see Figures 2.3 and 2.4). Moreover, the system is equipped with a dosage system including a static mixer, where suitable retention/formation aids can be added online.

Figure 2.1. Diagram of the flow loop system. All dimensions are properly scaled.

1. Transparent plastic headbox, vertically installed. Details see Figure 2.2.
2. PVC pipe, Ø=6.5 cm, roughly 320 cm long, connecting the headbox and the stock tank.
3. Two PVC valves. If chemical additives are online dosed in the system, the fibre suspension is not returned to the stock tank.
4. PVC stock tank, L=140 cm, D=60 cm, H=100 cm. Equipped with four electrical heaters, 4x1.5 kW, and a copper heat exchanger fed by cooling water.
5. Agitator, 1.15 kW.
6. PVC valve.
7. PVC pipe, Ø=6.5 cm, roughly 110 cm long between the stock tank and the pump.
8. Centrifugal pump, ABS FB 80/80-26, max 16.2 m³/h, max 1450 r/min, horizontal input, vertical output. Pump motor frequency controller, ABB ACS 300, 0.55 to 11 kW
9. PVC pipe, Ø=6.5 cm, roughly 50 cm long between the pump and the flow meter.
11. Dosage inlet, Ø=9 mm PVC tube, with PVC valve. Chemicals are dosed in the middle of the pipe along with the flow direction roughly 35 cm before the static mixer. The chemical dosage system is shown in Figure 2.5.
12. Inline static mixer, Komax Motionless Mixer M series, installed roughly 70 cm after the flow meter. Transparent PVC pipe, Ø=6.5 cm, 40 cm long, with 5 changeable mixing elements.
13. PVC pipe, Ø=6.5 cm, roughly 250 cm between the flow meter and the radial distributor including an inline static mixer.
14. Radial distributor, vertical input, horizontal output. Eight output PVC tubes, Ø=9 mm, are perpendicularly connected to the input PVC pipe, Ø=6.5 cm, evenly distributed around the pipe perimeter.
15. Eight plastic hoses with the same diameter and length, Ø=1.5 cm, L=235 cm, connecting the radial distributor and the headbox.

![Figure 2.2. Dimensions of the headbox with step diffuser pipe package, nozzle, and nozzle extension. Measures in mm.](image)

The fibre suspension is pumped into the flow loop system by a frequency controlled centrifugal pump from the stock tank, which has a maximum volume of 840 l. The flow rate is measured by a vertically installed electromagnetic flow meter, and the maximum flow rate in the system is approximately 300 l/min (5x10⁻³ m³/s). The system uses a simple radial distributor, sometimes applied as a cross-direction distributor for industrial headboxes. The headbox is made transparent, and is installed vertically to reduce the space requirement. There are eight equal-length feed hoses between the radial distributor and the headbox. Each feed tube in the headbox is of step diffuser type, simulating the flow in a typical industrial headbox. Figure 2.2 shows the details of the headbox.
The headbox nozzle contraction ratio is 16.7, and at the exit of the nozzle contraction, turbulence has decayed. Following the headbox nozzle contraction, there is a 110-mm-long straight channel, with a cross-section of $40 \times 15 \text{ mm}^2$, here called "extended nozzle", 15 mm being adopted as a normal slice opening in a real paper machine. A removable secondary contraction, shown in Figures 2.3 and 2.4, made of two plastic inserts inside of the extended nozzle, mounted perpendicular to the headbox nozzle contraction, is designed for studies of fibre floc rheology. It simulates flow accelerations that can happen during forming. The secondary contraction is replaceable, which means that the contraction ratio can be changed in order to generate different flow accelerations. By the secondary contraction used in this thesis work illustrated in Figure 2.4, the width of the extended nozzle is reduced from 40 mm to 20 mm. The curvature of the contraction is specially designed so that the acceleration of the flow suspension is constant. The maximum flow velocity after the secondary contraction is 16 m/s (960 m/min).

![Figure 2.3](image1)

*Figure 2.3. Diagram illustrating the 3-D structure of the secondary contraction inside of the extended nozzle after the headbox nozzle contraction. Dimensions are not scaled.*

![Figure 2.4](image2)

*Figure 2.4. Diagram illustrating one example of the secondary contraction. The curvature of the contraction blocks follows the equations: $y = 20x/(x+40)$, $0 \leq x \leq 40$, and $y = 10$, $40 < x \leq 85$. Measures in mm.*
Chemicals can be added online into the flow system by a piston instead of a pump, besides being directly added into the stock tank. This arrangement is beneficial, because some polymers are sensitive to high shear rates. Figure 2.5 shows the dosage system. The maximum volume of the piston cylinder is 5 l. Chemicals are mixed with the fibre suspension in the inline static mixer (see Figure 2.1). The mixer elements can be inserted in different numbers and in different patterns to generate different mixing effects. Tap water pressure is used as the dosing force. A water pressure regulator is used to stabilize the water pressure, and a needle valve is used to control the dosing speed. The dosing speed can be measured by a linear sensor, which is attached to the piston. When chemicals are online added, the suspension after the headbox is not returned to the stock tank.

Figure 2.5. Diagram of the chemical dosage system. All dimensions are properly scaled.

1. Tap water inlet.
2. Needle valve.
3. Water pressure regulator with water pressure meter, max 600 kPa.
4. Ball valve.
5. Water room.
6. Piston.
7. Chemicals room.
8. Transparent PVC piston cylinder, L=50 cm, A=100 cm², V=5 l.
9. Piston cylinder outlet, to the flow system.
10. Connection piston-sensor.
11. Linear sensor, Regal KTC 525.
12. Signal converter with display.
3. EXPERIMENTAL METHODS (PAPER I)

The fibre suspension flocculation was studied using an optical detection method. Images are taken through the extended nozzle by a high speed CCD video camera with transmitted infra-red laser light pulse illumination (see Figure 3.1). The camera control unit triggers the laser light, so the camera catches images synchronously with the laser light flashing. Since the laser light is very strong, stray light does not affect the images. And since the laser pulse length used in the experiments is only around a few microseconds, images remain sharp in spite of the fast flow. After the experiment, images are saved as 8-bit monochrome uncompressed Tiff-file in an Apple Power Mac computer.

![Figure 3.1. Imaging equipment. Dimensions are not scaled.](image)

1. **Transparent plastic headbox.**
2. **Infra-red laser head, Oxford Lasers HSI1000, wavelength 805 nm, peak power 150 W, individual pulse length 1 to 80 µs, max frequency 1000 Hz.**
3. **Infra-red laser control unit, Oxford Lasers HSI1000, external trigger input.**
4. **High speed CCD video camera control/display unit, Redlake Imaging, Motion Scope HR 1000, S-Video output, trigger output.**
5. **High speed CCD video camera, Redlake Imaging, Motion Scope HR 1000, 480x420 pixels, max recording speed 1000 frames/s.**
6. **Apple Power Mac, S-Video input, images treated by NIH Image 1.62 software, and saved in 8-bit monochrome uncompressed Tiff format.**

The aperture of the camera is fixed (f=22), and the laser light output is expanded by two pieces of lens (F=25 and F=150), which gives an illuminated area of approximately 75x75 mm². The laser light pulse width and intensity are kept constant in the experiments. Images of fibre suspension flocculation can be taken through the extended nozzle either separately before and after or along the secondary contraction, as illustrated in Figure 3.2.
Figure 3.2. Experimental methods. (A) Images can be taken respectively before and after the secondary contraction. (B) Images can be taken along the secondary contraction. All dimensions are properly scaled.

The images taken before the secondary contraction are 400×400 pixels or 40×40 mm², since the resolution of the images is 0.1 mm/pixel, which covers the whole nozzle width. The images taken after the secondary contraction are 400×200 pixels or 40×20 mm², which covers the whole channel width. Because of the boundary layer effects on both sides of the channel, only the middle parts of the images in CD are used. The cut images before and after the secondary contraction are 400×200 pixels and 400×100 pixels respectively. In each suspension, 400 successive images are taken at a camera recording speed of 50 frames/s.

The images taken along the secondary contraction cover the 40-mm-long contraction plus 10-mm-long upstream and 20-mm-long downstream, i.e. totally 70 mm, as illustrated in Figures 3.2 B and 4.7. The camera recording speed is set at 1000 frames/s, and it is possible to follow the fibre flocs manually along the secondary contraction as the maximum flow velocity after the contraction is 16 m/s.

Figure 3.3. Possible observation areas A–D in the extended nozzle without and with the secondary contraction. All dimensions are properly scaled.
For geometrical reasons (limited observation area in MD between nozzle outlet and secondary contraction), images representing the condition before and after the secondary contraction (C and D in Figure 3.3) are taken at the same position without and with the secondary contraction (B and D in Figure 3.3). Since the gravity difference between before the secondary contraction (C) and without the contraction (B) was negligible, and secondary flow was not found near the channel wall before the secondary contraction, the flow condition difference between the areas (C) and (B) was negligible.

Reflection and refraction from the interface between the nozzle wall and the flowing suspension can be neglected, because the camera detects no signal in the direction perpendicular to the laser beam. Suspensions of certain pulp species, such as SGW and TMP pulps, which have a high amount of fines material, are not suitable for this optical detection method, as illustrated in Figure 3.4. The light refracted from the fines fraction suppresses the visualisation of the fibre flocs in the suspension. The method may, however, be applicable if fines material is removed from the SGW and TMP pulps.

Figure 3.4. Different pulp suspensions at a fibre concentration of 5 g/l at a flow velocity of 8 m/s after the headbox nozzle. Image size: 40x20 mm². (A) Bleached softwood kraft pulp. (B) Bleached hardwood kraft pulp. (C) Unbleached TMP pulp. (D) Unbleached SGW pulp.
4. IMAGE ANALYSIS (PAPER I, II, III & V)

Images of fibre suspension flocculation were analysed in an Apple Power Mac computer using NIH 1.62 and Matlab 5.2.1 software, either automatically or manually. After the laser light compensation, different evaluation methods were applied for different purposes.

4.1. Calibration of Light Intensity – Fibre Concentration

If the fibre suspension is well mixed in the headbox, the average of successive images taken through the extended nozzle should be constant. There is, however, an unevenness of the average of 400 successive images, caused by an uneven laser light illumination. Such a kind of uneven illumination should be subtracted from each image as light compensation, as illustrated in Figure 4.1.

![Figure 4.1. A bleached softwood kraft pulp suspension at a fibre concentration of 5 g/l at a flow velocity of 8 m/s after the headbox nozzle. Image size: 40x20 mm². (A) An uneven distribution of the laser light illumination based on 400 successive images at a recording speed of 50 frames/s. (B) Original image. (C) Compensated image of (B).](image)

After the laser light compensation, the gray value on the images is proportional to the fibre concentration in the suspensions in the range of 2.5 to 7.5 g/l as shown in Figure 4.2. It should be pointed out that it is also possible to study the higher fibre concentrations.
roughly up to 10 g/l in this optical method, while for the lower fibre concentrations, further improvement in the imaging detection technique has to be implemented.

![Figure 4.2. The observed relationship between mean fibre concentration and mean gray value for a suspension of bleached softwood kraft pulp. The mean gray value was calculated from the average of 400 images after the laser light compensation.](image)

### 4.2. Power Spectrum

The theory of frequency analysis was introduced during the early 1970s, Norman and Wahren (1972), and was subsequently applied to determine the degree of fibre floculation, Nerelius et al. (1972). In the 1980s, Wågberg and Lindström (1987b) introduced the concept of floculation index for studying polymer-induced fibre floculation. In the 1990s, Beghello et al. (1996) applied the concept of mean floc size. In the traditional wavelength power spectrum, presented using log-scales on both x- and y-axis, it is difficult to appreciate the contribution from larger wavelengths. Johansson and Norman (1996) therefore suggested multiplying the spectral density by the wavelength \( \lambda \). A better impression of the actual area below the different parts of the spectrum will then be obtained.

#### 4.2.1. Wavelength Spectral Density

Frequency power spectra are transformed into wavelength spectra by

\[
\frac{E(u)}{n} = E(\lambda) = \frac{n^2}{u} P(n),
\]

where \( \lambda \) is wavelength, \( u \) is flow velocity, \( n \) is frequency, \( E(\lambda) \) is wavelength spectral density, and \( P(n) \) is frequency spectral density.
In the power spectrum analysis method, the 8-bit monochrome image is converted to a matrix, where columns are in MD and rows are in CD. Then the matrix is normalized by subtracting the mean value, and making the maximum amplitude to one. Power spectral density is estimated in MD and CD respectively, and each spectrum is normalized with the mean squared amplitude of the input signal. It should be pointed out that the power spectral density in this thesis work is the average obtained from 400 successive images. Figure 4.3 illustrates an example of the power spectra for the suspensions of bleached softwood and hardwood kraft pulps as shown in Figures 3.4 A and B.

![Figure 4.3. Spectral density distribution as a function of wavelength in MD for softwood pulp suspension (dotted line) and hardwood pulp suspension (solid line) at a fibre concentration of 5 g/l at a flow velocity of 8 m/s after the headbox nozzle.](image)

### 4.2.2. Mean Floc Size

The evaluation method of the mean floc size is extended in this thesis work, and the concepts of mean floc aspect ratio and mean floc area reduction are introduced.

In MD, the mean wavelength, \( \lambda_{\text{mean}} \), is defined as the value to separate the integrated spectrum area in the range of 2-32 mm into two equal halves, which is calculated from

\[
\int_{2\text{mm}}^{\lambda_{\text{mean}}} E(\lambda)d\lambda = \int_{\lambda_{\text{mean}}}^{32\text{mm}} E(\lambda)d\lambda. \tag{4.2}
\]

The mean floc size is interpreted as half the value of the mean floc wavelength \( \lambda_{\text{mean}} \). The floc size is not an absolute value, but for better understanding, it can be regarded as the scale index of a floc.

The image MD length is 40 mm, and the selected floc size range in MD is chosen from 1 mm to 16 mm. The signal below 1 mm is regarded as noise. The maximum size 16 mm is due to the limited image width in MD.
In CD, the lengths of images before and after the secondary contraction are different. The selected floc size ranges before and after the contraction are chosen from 1 mm to 8 mm and from 1 mm to 4 mm respectively, equaling to the wavelength range of 2-16 mm and 2-8 mm respectively in Equation [4.2]. The maximum sizes 8 and 4 mm are due to the limited image widths in CD, which are 20 and 10 mm respectively before and after the contraction.

The mean floc aspect ratio is defined as the mean floc size in MD divided by the mean floc size in CD. The mean floc area reduction in the MD-CD plane is defined as the percentage cross-section area reduction for fibre flocs passing through the secondary contraction. To evaluate the floc volume reduction, it would be necessary to observe also the dimensions in the z-direction, which was not possible in this thesis work.

### 4.2.3. Flocculation Index

The flocculation index is a kind of relative value describing the fibre flocculation intensity in suspensions. The definition of flocculation index is extended in this thesis work as

$$ F = \frac{V_A^2 - V_B^2}{V_A^2 - V_B^2} \sqrt{V_A^2 - V_B^2}, $$

where $F$ is the flocculation index, $V_A$ is the coefficient of variation of the target suspension, and $V_B$ is the coefficient of variation of the reference suspension. Since the square of the coefficient of variation equals to the area below the spectral curve, $V_A^2 - V_B^2$ equals to the area between curve A and B in Figure 4.4.

**Figure 4.4. Illustration of power spectra. The area between curve A (solid line) and curve B (dash-dot line) is painted in gray.**

It should be pointed out that the flocculation index can be negative, which means that the target suspension is deflocculated compared with the reference suspension. For instance, if in Figure 4.4 the curve B represents the target suspension and the curve A represents the reference suspension, the corresponding flocculation index is negative. Since usually the variations of fibre distribution at larger wavelengths are especially interesting for com-
paring the effects on fibre suspension flocculation, the flocculation index in this thesis work is only calculated in the wavelength range of 8-32 mm.

4.3. Wavelet Transform

The wavelet transform was first recognized as a specialised field by the work of Grossman and Morlet (1984). Since then, the wavelet theory has been mathematically developed, and has been applied in various fields with different purposes. In this thesis work, a two-dimensional continuous wavelet transform with a two-dimensional Mexican hat wavelet as a mother wavelet, Mao and Bopardikar (1998), is used to describe fibre suspension flocculation. The applications of the wavelet transform are discussed in details in Section 6, thus only a brief summary of the theory will be given in the section.

4.3.1. Two-Dimensional Continuous Wavelet Transform

If the integral of a function is zero, and it is quadratic integrable or equivalently has finite energy, the function qualifies as a mother wavelet or wavelet, which is written as

$$\int_{-\infty}^{\infty} \psi(t) dt = 0, \quad [4.5]$$

and

$$\int_{-\infty}^{\infty} |\psi(t)|^2 dt < \infty, \quad [4.6]$$

where \( \psi(t) \) is a mother wavelet, and \( t \) can be e.g. a time or space variable.

Let \( f(t) \) be any square integrable function. The continuous wavelet transform of \( f(t) \) with respect to a mother wavelet \( \psi(t) \) is defined as

$$W(a,b) = \int_{-\infty}^{\infty} f(t) \frac{1}{\sqrt{|a|}} \psi^\ast \left( \frac{t-b}{a} \right) dt, \quad [4.7]$$

where \( a \) and \( b \) are real and \( \ast \) denotes complex conjugation.

If \( \psi_{a,b}(t) \) is defined as

$$\psi_{a,b}(t) = \frac{1}{\sqrt{|a|}} \psi\left( \frac{t-b}{a} \right), \quad [4.8]$$

Equation [4.7] can be written as
\[ W(a,b) = \int_{-\infty}^{\infty} f(t)\psi_{a,b}^*(t)dt. \]  \[4.9\]

Notice that
\[ \psi_{1,0}(t) = \psi(t). \]  \[4.10\]

The energy of \( \psi_{a,b}(t) \) remains the same for all \( a \) and \( b \) values, which is
\[ \int_{-\infty}^{\infty} |\psi_{a,b}(t)|^2 dt = \int_{-\infty}^{\infty} |\psi(t)|^2 dt. \]  \[4.11\]

For any given value of \( a \), the function \( \psi_{a,0}(t) \) is a shift of \( \psi_{a,0}(t) \), shown in Equation \[4.12\], by an amount \( b \) along the time axis. Thus, the variable \( b \) represents time/space shift or translation. Thus, \( \psi_{a,0}(t) \) is a time/space and amplitude-scaled version of \( \psi(t) \). Since \( a \) determines the amount of time/space scaling or dilation, it is referred to as the scale or dilation variable. If \( a > 1 \), \( \psi(t) \) is stretched along the time axis; whereas if \( 0 < a < 1 \), it is contracted. Negative values of \( a \) result in time reversal in combination with dilation.

The set of quadratic integrable functions forms a linear vector space under addition and scalar multiplication. This vector space comes with a well-defined inner product. Given two finite energy signals \( x(t) \) and \( y(t) \), their inner product is defined as
\[ \langle x(t), y(t) \rangle = \int_{-\infty}^{\infty} x(t)y^*(t)dt, \]  \[4.13\]
where the inner product is denoted by \( \langle x(t), y(t) \rangle \). The total energy in \( x(t) \) is given by
\[ \int_{-\infty}^{\infty} |x(t)|^2 dt = \langle x(t), x^*(t) \rangle. \]  \[4.14\]

As is shown in Equation \[4.9\], the continuous wavelet transform is essentially a collection of inner products of a signal \( f(t) \) and the translated and dilated wavelet \( \psi_{a,b}(t) \) for all \( a \) and \( b \) values
\[ W(a,b) = \langle f(t), \psi_{a,b}(t) \rangle. \]  \[4.15\]

The cross-correlation \( R_{x,y}(\tau) \) of the two functions \( x(\tau) \) and \( y(\tau) \) is defined as
\[ R_{x,y}(\tau) = \int_{-\infty}^{\infty} x(t)y^*(t-\tau) = \langle x(t), y(t-\tau) \rangle, \]  \[4.16\]
where \( \tau \) is the lag or shift parameter.

By using Equation \[4.8\], \[4.12\] and \[4.16\], Equation \[4.15\] can be written as
\[ W(a,b) = \langle f(t), \psi_{a,b}(t-b) \rangle = R_{f,\psi_{a,b}}(b), \quad [4.17] \]

which means that the continuous wavelet transform is the cross-correlation at lag \( b \) between \( f(t) \) and the wavelet dilated to a scale factor \( a \).

Similarly, the two-dimensional continuous wavelet transform can be written as a two-dimensional cross-correlation

\[ W(a_x,b_x,a_y,b_y) = \langle f(x,y), \psi_{a_x,0,a_y,0}(x-b_x,y-b_y) \rangle = R_{f,\psi_{a_x,0,a_y,0}}(b_x,b_y). \quad [4.18] \]

Images are typical two-dimensional signals. In this case, the two-dimensional continuous wavelet transform is simplified by reducing the translation vectors of two-dimensional wavelet and by setting the symmetrical two-dimensional wavelet in the middle of the co-ordinate system. The local maximum peaks obtained after the transform represent the positions where the original function (signal) is similar to the mother wavelet.

### 4.3.2. Two-Dimensional Mexican Hat Wavelet

In this thesis work, the Mexican hat wavelet, the second derivative of the negative Gaussian function, shown in Equation [4.19], is used as the mother wavelet. Figure 4.5 shows the one-dimensional Mexican hat wavelet, which is given by

\[ \psi(t) = (1-t^2)e^{-\frac{1}{2}t^2}. \quad [4.19] \]

![Figure 4.5. One-dimensional Mexican hat wavelet.](image)

The two-dimensional Mexican hat wavelet is obtained by rotating the one-dimensional Mexican hat wavelet around the vertical axis at \( t = 0 \). Equation [4.20] shows the function of two-dimensional Mexican hat wavelet, and Figure 4.6 shows the samples of two-dimensional Mexican hat wavelets dilated to different scale factors with the view along the vertical axis.
\[ \psi(x,y) = \left[ 1 - \frac{1}{2} (x^2 + y^2) \right] e^{-\frac{1}{2} (x^2 + y^2)}. \] [4.20]

Figure 4.6. Two-dimensional Mexican hat wavelet. Symmetrical dilation, left: \( a_x = a_y = (10^{2.5}/\pi)^{1/2} \) mm, right: \( a_x = a_y = (10^{2.0}/\pi)^{1/2} \) mm. Illustrated image size: 20x20 mm².

4.4. Manual Evaluation

A manual evaluation was made by following the flow along the secondary contraction. The images cover, in MD, the 40-mm-long contraction plus 10-mm-long upstream and 20-mm-long downstream, i.e. totally 70 mm. Fibre flocs are identified by following them along the contraction. Floc boundaries are distinguished by adjusting the threshold levels on the images. Floc area is the area of the best fitting ellipse for the floc boundary drawn by hand. Floc MD and CD lengths are calculated from the corresponding ellipse axes with an orientation angle.

Figure 4.7. The observation zones for manual evaluation of fibre flocs along the secondary contraction. Measures in mm.
As illustrated in Figure 4.7, two observation zones were chosen to compare the fibre flocs at different positions in the contraction. In order to obtain images with a high resolution along the secondary contraction, the corresponding observation area in MD is limited, thus both the upstream and downstream zones cover part of the contraction. In order to study large flocs, the width of the observation zone in MD was chosen as 30 mm. Since the flow velocity is doubled by the contraction, doubled observation areas after the contraction were chosen in order to follow the fibre flocs.

Fibre flocs during the contraction were manually evaluated twice respectively in the upstream and downstream zones. Only the flocs with clear boundaries that could easily be recognized by the naked eye were selected for the evaluation. If a fibre floc appeared more than once in the observation zones, it was evaluated as early as possible in the upstream zone, and as late as possible in the downstream zone. The situation during the contraction was also simplified by assuming that a fibre floc could either remain intact or be broken into two parts when travelling from the upstream zone to the downstream zone. In order to statistically reduce the errors in the manual evaluation, as many as 1000 successive images, with 0.001 s interval, were analysed. More details of this application are described in Section 5.
5. FOLLOWING FIBRE FLOCS ALONG FLOW CONTRACTION (PAPER III)

Images of a bleached softwood pulp suspension at a fibre concentration of 5 g/l were taken along the secondary contraction at a camera recording speed of 1000 frames/s. The flow was accelerated from 8 m/s to 16 m/s in the contraction. The images were also treated by the laser light compensation. Fibre flocs were assumed to be either broken up (see Figure 5.1) or remain intact (see Figure 5.2) when subjected to the flow contraction.

Figure 5.1. A fibre floc broken during the secondary contraction for a bleached softwood kraft pulp suspension at a fibre concentration of 5 g/l. The flow was accelerated from 8 m/s to 16 m/s in the contraction. Time interval between images: 0.001 s. Image size: 70x40 mm².

Figure 5.2. A fibre floc unbroken during the secondary contraction for a bleached softwood kraft pulp suspension at a fibre concentration of 5 g/l. Data as in Figure 5.1.

It should be pointed out that the fibre flocs are already anisotropic when entering the secondary contraction due to the acceleration in the headbox nozzle. At a fibre concentration
of 5 g/l, 1000 successive images, representing a 1-second recording along the secondary contraction, were manually analysed. As described in Section 4.4, 511 fibre flocs were selected in the upstream zone, and 19% of them were found to be broken into two parts in the downstream zone. The distributions of the different floc properties in the upstream and downstream zones are shown in Figure 5.3. It should be pointed out that in Figure 5.3 the fibre floc number in the upstream and downstream zones are different. The unit of pixel was converted to the corresponding unit of mm. The gray value on the images of fibre flocculation was converted to the corresponding unit of g/l for fibre concentration in the suspensions, according to the relationship between mean gray value and mean fibre concentration, shown in Figure 4.2. By multiplication with the channel thickness 15 mm, the floc grammage in g/m² can be calculated.

![Figure 5.3](image1.png)

**Figure 5.3.** Distributions of fibre floc properties in the upstream and downstream zones for a suspension of bleached softwood kraft pulp at a fibre concentration of 5 g/l. The flow was accelerated from 8 m/s to 16 m/s in the secondary contraction.
From the distributions, it shows that the fibre floc size increases in MD and decreases in CD by the flow contraction. The net fibre floc area is decreased and the fibre floc is thus concentrated by the flow contraction. The fibre floc is more elongated and oriented in MD by the flow contraction. Since ca 1/5 of the fibre flocs are broken into two parts, the average stretching of the flocs cannot be evaluated simply from the length distributions in the upstream and downstream zones in Figure 5.3 A. The average values of the fibre floc properties are listed in Table 5.1.

<table>
<thead>
<tr>
<th>Fibre floc property</th>
<th>Upstream zone</th>
<th>Downstream zone</th>
</tr>
</thead>
<tbody>
<tr>
<td>Floc number</td>
<td>511</td>
<td>608</td>
</tr>
<tr>
<td>Floc size in MD (mm)</td>
<td>9.98</td>
<td>10.9</td>
</tr>
<tr>
<td>Floc size in CD (mm)</td>
<td>5.20</td>
<td>3.26</td>
</tr>
<tr>
<td>Floc area (mm²)</td>
<td>41.6</td>
<td>28.8</td>
</tr>
<tr>
<td>Mean grammage in a floc (g/m²)</td>
<td>131</td>
<td>156</td>
</tr>
<tr>
<td>Floc aspect ratio</td>
<td>2.00</td>
<td>3.49</td>
</tr>
<tr>
<td>Floc absolute orientation angle (˚)</td>
<td>16.7</td>
<td>6.41</td>
</tr>
</tbody>
</table>

Since the breaking of the fibre flocs conceals some of the information on the floc extension, the distributions of fibre floc size in MD are separated and shown in Figure 5.4, according to the floc behaviour in the contraction: remaining intact or being broken into two parts. When a fibre floc is broken into two parts, the smaller one was marked as I, and the larger one was marked as II. The distributions of the groups I and II are shown respectively in Figure 5.4 B. During the flow contraction, a fibre floc is stretched in MD. If a fibre floc breaks into two parts, the corresponding broken floc sizes in MD decrease. The summary of these two broken flocs size in MD is, however, still larger than the original floc size in MD, since a stretching is the cause for breaking up the fibre floc. Fibre flocs with a larger size in MD are more inclined to break by the flow contraction. The average value of the fibre floc size in MD is listed in Table 5.2.

![Figure 5.4](image-url)  
*Figure 5.4. Distributions of fibre floc size in MD in the upstream and downstream zones for a suspension of bleached softwood kraft pulp at a fibre concentration of 5 g/l. The flow was accelerated from 8 m/s to 16 m/s in the secondary contraction. (A) 414 flocs, remaining intact. (B) 97 flocs, being broken into two parts, group I (smaller part), group II (larger part).*
Table 5.2. Average fibre floc size in MD (mm).

<table>
<thead>
<tr>
<th>Floc behaviour in the contraction</th>
<th>Upstream zone</th>
<th>Downstream zone</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flocs remaining intact</td>
<td>9.48</td>
<td>12.0</td>
</tr>
<tr>
<td>Flocs being broken into two parts</td>
<td>12.1</td>
<td>7.05 &amp; 10.2</td>
</tr>
</tbody>
</table>

The fibre floc weight was simply calculated by multiplying the floc grammage and the floc area. Figure 5.5 shows the relation between the fibre floc weight in the downstream and upstream zone. If a fibre floc was broken into two parts in the downstream zone, the fibre floc weight in the downstream zone was plotted as the summary of these two flocs against the corresponding fibre floc weight in the upstream zone, which means there are 511 points illustrated in Figure 5.5. The equation of the linear trendline in Figure 5.5 shows that the fibre floc weight in the downstream zone statistically equals that in the upstream zone. This confirms that the fibre floc is concentrated by the flow contraction, due to the lower floc area in the downstream zone.

![Figure 5.5. Fibre floc weight in the downstream zone vs. the corresponding fibre floc weight in the upstream zone for a suspension of bleached softwood kraft pulp at a fibre concentration of 5 g/l. The flow was accelerated from 8 m/s to 16 m/s in the secondary contraction.](image)

The information mentioned above only considers compression in the MD-CD plane. At low concentrations, only fluid forces are relevant. Due to the fibre network elasticity, when a fibre floc is stretched in MD in the flow, it would be shrunken both in CD and in the z-direction. Compression in the z-direction could then be equal to compression in CD.
This means that an increase in floc concentration could be higher than that given by a calculation which is only concerned with the MD-CD plane.

Water is moved out from the fibre network by the flow contraction. The fibre floc area is decreased, and the fibre flocs are concentrated. The statistical survey of manually following fibre flocs along the secondary contraction proves the phenomenon, which is confirmed by an increase in floc gray scale level in the image analysis. Inside a forming zone, fibre flocs may pass through pressure pulse generated acceleration zones. They may then be stretched and broken, and the dewatering of the fibre flocs may already have been started in the suspension, before reaching the wires in the forming zone. Therefore, increasing the local accelerations during dewatering or the headbox nozzle contraction ratio might help to increase the dewatering efficiency, but at the same time, a grainy formation can be generated.

Manual evaluation of fibre flocs along the flow contraction has confirmed that the power spectrum analysis method is applicable. The corresponding results from the power spectrum analysis method before and after the secondary contraction for the same fibre suspension at the same fibre concentration are discussed in Section 7.1.
6. APPLICATIONS OF TWO-DIMENSIONAL CONTINUOUS WAVELET TRANSFORM (PAPER II)

Besides of the information of overall fibre suspension flocculation, the information of single fibre flocs is also needed in some cases, such as studying fibre floc distribution and fibre floc rheology in the suspension. The traditional power spectrum analysis method fails to evaluate the individual fibre flocs, thus, the wavelet transform analysis method, described in Section 4.3, is required. In this thesis work, two applications have been demonstrated.

6.1. Inner Floc Structure

Fibres begin to contact in a suspension when the suspension concentration is over a certain crowing factor levels, described by Kerekes and Schell (1992). In most papermaking cases, the fibre concentration in the suspension during forming is rather above this level, which means that the fibres are entangled to each other. Unlike the situation in paper sheets, floc networks in such suspensions are loose and comparatively large. Fibre flocs can be classified into different sizes with different scales, and there can be small flocs inside of large flocs.

![Figure 6.1. Images of simulated flocs. Image size: 40x20 mm².](image-url)
Images of such flocs were simulated as shown in Figure 6.1. The flocs were generated by overlaying ellipses in three different colours, respectively 100%, 60% and 30% of the black colour in linear gray scale. These ellipses were then filtered using a Gaussian Blur. The outer structure of these 3 samples is the same, while the inner structure of these 3 samples is different.

By the classical wavelength power spectrum evaluation method, as described in Section 4.2, the mean floc sizes in MD obtained from the images of A, B and C in Figure 6.1 are 10.79, 10.81 and 10.84 mm respectively. That means by such an analysis method, only the outer floc information can be evaluated, while not the inner floc information. From a concept of power spectrum, the information on the “inside floc” size could also be obtained, but it would not be possible to see their actual location. However, by using different scaled or dilated two-dimensional Mexican hat wavelets, this information can be extracted from the images of simulated flocs shown in Figure 6.1.

Two different scales of Mexican hat wavelets were used, as shown in Figure 4.6, while the integral scales of those wavelets were set as the same as the simulated images, 40x20 mm². The signals after the transforms were converted into 5 contour levels with the same linear gray scale as that of the simulated images for better illustration. Figures 6.2 and 6.3 show the respective results. With the large scale Mexican hat wavelet, the local maximum peaks represent the centers according to the outer floc shapes, shown in Figure 6.2. With the small scale Mexican hat wavelet, the local maximum peaks represent the centers according to the inner floc shapes, shown in Figure 6.3. Thus, floc size can be classified, and inner floc structure can be studied.

The mean floc structure index is defined as the number of the peaks after the transform with the small scale Mexican hat wavelet divided by the number of the peaks after the transform with the large scale Mexican hat wavelet. The larger the mean floc structure index, the more sub-floc structure the floc has. For example, the mean floc structure indexes for image A, B and C in Figure 6.1 are 1, 1.5 and 2 respectively.

![Figure 6.2](image.png)

*Figure 6.2. Simulated flocs images after two-dimensional continuous wavelet transforms with two-dimensional Mexican hat wavelets dilated to the scale factors of $a_x = a_y = (10^{2.3}/\pi)^{1/2}$ mm. Image size: 40x20 mm².*
Supposing the whole image of a real fibre suspension was a large floc, the mean floc structure index could be regarded as a floc number index. The larger the mean floc structure index, the more flocs exist in the suspension. It should be pointed out that the mean floc structure index is also a value based on the concentration variation in the images. Such an application of wavelet transform can be used to evaluate the floc numbers in suspensions. For example, the mean floc structure indexes, calculated from 400 successive images after the laser light compensation, for the suspensions of bleached softwood and hardwood pulps at a fibre concentration of 5 g/l, as illustrated in Figures 3.4 A and B, are 2.08 and 2.83 separately. That means there are more fibre flocs in the hardwood pulp suspension than in the softwood pulp suspension.

6.2. Tracing Fibre Flocs

Another advantage of the continuous wavelet transform is that it can give the position of the fibre flocs. Figures 6.4 and 6.5 show the sample images, as illustrated in Figures 5.1 and 5.2, with the results of the wavelet transforms. The flocs centers, local maximum signal peaks after the two-dimensional continuous wavelet transforms, are marked on the original images. The mother wavelet was chosen as the two-dimensional Mexican hat wavelet dilated to the scale factors of \( a_x = a_y = (10^{2.0}/\pi)^{1/2} \) mm. Since the small scale Mexican hat wavelet is used, initially the broken floc has two centers, which can be regarded as the centers of the inner flocs. The inner structure caused the floc breaking during the flow contraction.
Figure 6.4. A fibre floc broken during the flow contraction for a bleached softwood kraft pulp suspension at a fibre concentration of 5 g/l. The flow was accelerated from 8 m/s to 16 m/s. Time interval between images: 0.001 s. Image sizes: 70x40 mm². Centers of the followed fibre floc(s) are marked according to the two-dimensional continuous wavelet transforms.

Figure 6.5. A fibre floc unbroken during the flow contraction for a bleached softwood kraft pulp suspension at a fibre concentration of 5 g/l. Data as in Figure 6.4.

Compared with the manual method of plotting the centers of flocs in the images, the two-dimensional continuous wavelet transform is more objective and reliable. If the ratio of broken to unbroken flocs by the flow contraction can be calculated automatically, the fibre floc strength in suspensions can be evaluated.

The floc mean velocities in MD in the center of the converging channel can be calculated from Figures 6.4 and 6.5, and are shown in Figure 6.6. The situation can be simplified assuming that fibre flocs travel straight in MD in the middle of the channel. The floc mean velocity was calculated by dividing the floc travel distance by the travel time. Figure 6.6 shows that the mean floc velocity is linearly increased in the middle of the channel during the flow contraction, conforming the design of the contraction curvature, illustrated in Figure 2.4. If fibre flocs at different zones along the contraction are traced, it is possible to obtain the complicated floc velocity profile in the flow contraction. Furthermore, the fibre floc rheology in suspensions can be studied.
Figure 6.6. The floc mean velocity in MD in the middle of the flow channel along the secondary contraction assuming fibre flocs travelling straight in MD. The distance is measured from the top on Figures 6.4 and 6.5. Floc A is the floc marked with plus in Figure 6.4, floc B is the floc marked with cross in Figure 6.4, and floc C is the floc marked with star in Figure 6.5.

6.3. Limitation of Wavelet Transform

The disadvantage of the wavelet transform is the calculation time required with a standard personal computer. Although a curve of the floc size distribution can be obtained by the wavelet transform, much more calculation is needed for the wavelet transform analysis compared to the power spectrum analysis. Usually, the mean size distribution of fibre flocs is more interesting than the exact number of fibre flocs of a certain size when characterising the overall fibre suspension flocculation in papermaking. Thus, the wavelet transform is less useful to study the effects on the overall fibre suspension flocculation. However, when following individual fibre flocs in a suspension, the wavelet transform can be used to describe the positions and the structure of the fibre flocs, which cannot be obtained from the power spectrum analysis. The programming technique should also be further developed in order to improve the intelligence of computerised analysis so that the fibre flocs can be distinguished and traced automatically.
7. PHYSICAL INFLUENCES ON FIBRE SUSPENSION FLOCCULATION (PAPER III, IV & V)

Fibres in a suspension have a strong tendency to entangle to form connected networks. Fibre flocs can be considered as the fragments of such networks that have a higher fibre concentration than the mean fibre concentration in the suspension. In this thesis work, a study was focused on some physical influences on fibre suspension flocculation under simulated forming conditions. Fibre suspension flocculation was evaluated by the power spectrum method, described in Section 4.2.

7.1. Fibre Concentration

Fibre concentration has a significant effect on fibre suspension flocculation. Two concepts, critical concentration, Mason (1954), and crowding factor, Kerekes and Schell (1992), have been applied to explain the mechanism. In this investigation, the concentration effect on fibre suspension flocculation of a refined bleached softwood kraft pulp, ca 20 °SR, and a refined unbleached softwood kraft pulp, 25 °SR, was studied using the secondary contraction. The flow velocities were 8 m/s and 16 m/s respectively before and after the contraction.

The mean floc size for bleached and unbleached softwood kraft pulp suspension before and after the contraction respectively is evaluated and plotted against the fibre concentration in MD and CD respectively in Figure 7.1. It shows that for both pulp suspensions, the mean floc size increases in MD and CD with increasing fibre concentration, both before and after the contraction. At all fibre concentrations, the mean floc size in MD increases during the contraction, while the mean floc size in CD decreases during the contraction. The reason why the fibre floc size in MD for bleached softwood pulp suspensions is larger than for unbleached softwood pulp suspensions might be that the fibre length of bleached pulp is longer than that of unbleached pulp in this case. Fibre length has a dominating effect on fibre floc size, especially in MD, as will be discussed in Section 7.2.

The mean floc aspect ratio for both pulp suspensions is plotted against the fibre concentration as shown in Figure 7.2. The mean floc aspect ratio after the contraction is higher than before the contraction, which means that the fibre flocs are elongated by the flow contraction. For different pulp suspensions, the longer the fibres, the larger the mean floc aspect ratio, as will be discussed in Section 7.2. The mean floc aspect ratio remains roughly at the same level with increasing fibre concentration both before and after the contraction.
Figure 7.1. Mean floc size in MD and CD respectively for suspensions of bleached and unbleached softwood kraft pulp at different fibre concentrations at flow velocities of 8 m/s and 16 m/s respectively before and after the secondary contraction. BSW: Bleached softwood kraft pulp. UBSW: Unbleached softwood kraft pulp.

Figure 7.2. Mean floc aspect ratio for suspensions of bleached and unbleached softwood kraft pulps at different fibre concentrations at flow velocities of 8 m/s and 16 m/s respectively before and after the secondary contraction. BSW: Bleached softwood kraft pulp. UBSW: Unbleached softwood kraft pulp.
The mean floc area reduction is plotted against the fibre concentration in Figure 7.3. It should be pointed out that this does not represent the total floc area reduction. Due to the fibre floc breakage, there are significantly more fibre flocs after the contraction than before the contraction. The figure shows that for both pulp suspensions, the mean floc area reduction increases with increasing fibre concentration. Since the extended nozzle width is reduced at the contraction, the fibre flocs may consequently be mechanically compressed in CD in order to pass through the contraction. At lower fibre concentrations, more free water exists between the fibre flocs, so the fibre flocs do not need to compress so much to pass the contraction. At higher concentrations, less free water exists between the fibre flocs, so the fibre flocs need to compress more to pass the contraction. It could be expected that when further increasing the fibre concentration, the mean floc area reduction could approach 50%, as the nozzle width is reduced by 50%. Björkman (1999) described such compression phenomena for fibre networks at a high suspension concentration. In his publication, a mechanical force was directly applied on the fibre networks.

Table 7.1. Comparison of manual evaluation and power spectrum analysis.

<table>
<thead>
<tr>
<th>Mean fibre floc property</th>
<th>Manual evaluation</th>
<th>Power spectrum analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Floc size in MD (mm) upstream</td>
<td>9.98</td>
<td>8.30</td>
</tr>
<tr>
<td>Floc size in MD (mm) downstream</td>
<td>10.9</td>
<td>8.56</td>
</tr>
<tr>
<td>Floc size in CD (mm) upstream</td>
<td>5.20</td>
<td>4.14</td>
</tr>
<tr>
<td>Floc size in CD (mm) downstream</td>
<td>3.26</td>
<td>2.38</td>
</tr>
<tr>
<td>Floc aspect ratio upstream</td>
<td>2.00</td>
<td>2.01</td>
</tr>
<tr>
<td>Floc aspect ratio downstream</td>
<td>3.49</td>
<td>3.60</td>
</tr>
<tr>
<td>Floc area reduction (%)</td>
<td>31.5</td>
<td>40.7</td>
</tr>
</tbody>
</table>
Table 7.1 shows a comparison between manual evaluation, discussed in Section 5, and power spectrum analysis for the same suspension of bleached softwood pulp at a fibre concentration of 5 g/l. The flow was accelerated from 8 m/s to 16 m/s in the secondary contraction. Although the observation areas and the flow conditions in the upstream and downstream zones are not the same as those before and after the secondary contraction, the trends in the results obtained by manual evaluation and by power spectrum analysis are correlated satisfactorily. The mean floc size obtained by manual evaluation is larger in MD and CD than that obtained by power spectrum analysis. This might be due to that the selected flocs were larger in the manual evaluation. The evaluated mean floc aspect ratios are approximately equal in the two methods. The mean floc area reduction evaluated by manual evaluation is smaller than that obtained by power spectrum analysis. This is mainly due to the breaking of fibre flocs, as discussed above. It could be compensated for in the manual evaluation, in which the area reduction is calculated from the total floc area, but not in the power spectrum analysis, in which the area reduction is calculated from the mean floc area.

### 7.2. Refining

Some studies have also been performed demonstrating the relationship between fibre properties and fibre flocculation, Kerekes and Schell (1995), Kerekes (1995), Beghello and Eklund (1997). Although the main target of refining is to improve the bonding ability between fibres in order to increase paper strength, refining is sometimes also applied to improve paper sheet formation. Since refining affects several fibre properties, such as shortening the fibres, curling/decurling of fibres, increasing the external fibrillation, increasing fibre flexibility, and so on, the corresponding effects on fibre suspension flocculation is hard to interpret for the papermaker. In this investigation, twelve pulp samples (four different pulps each with three different degrees of refining) were evaluated with respect to their flocculation propensity. The corresponding refining effect on paper formation is discussed in Section 9.

Four kinds of commercial bleached softwood pulps were refined at an edge length of 118 km/s and an idling effect of 65 kW to three different net refining energy levels respectively in an industrial low consistency double disc refiner (Voith Sulzer, type: TF2E, segment dimension: 3 mm wide, 4 mm deep, 40 mm long, 30˚ angle). Tables 7.2, 7.3 and 7.4 show the details.

<table>
<thead>
<tr>
<th>ID</th>
<th>Pulp industrial name</th>
<th>Mixture</th>
<th>Late wood</th>
<th>Chips</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Botnia verde88 TCF</td>
<td>80% pine &amp; 20% spruce</td>
<td>37%</td>
<td>Sawmill</td>
</tr>
<tr>
<td>B</td>
<td>Södra Cell Tofte 90S ECF</td>
<td>10% pine &amp; 90% spruce</td>
<td>34%</td>
<td>Roundwood</td>
</tr>
<tr>
<td>C</td>
<td>Billerud Kraft ECF</td>
<td>72% pine &amp; 28% spruce</td>
<td>42%</td>
<td>Sawmill</td>
</tr>
<tr>
<td>D</td>
<td>Södra 90 TZ TCF</td>
<td>55% pine &amp; 45% spruce</td>
<td>42%</td>
<td>Sawmill</td>
</tr>
</tbody>
</table>
Table 7.3. Refining conditions.

<table>
<thead>
<tr>
<th>ID</th>
<th>C, g/l</th>
<th>pH</th>
<th>T, °C</th>
<th>Flow, l/min</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>38</td>
<td>6.7</td>
<td>23</td>
<td>652</td>
</tr>
<tr>
<td>B</td>
<td>38</td>
<td>7</td>
<td>24</td>
<td>652</td>
</tr>
<tr>
<td>C</td>
<td>40</td>
<td>6.35</td>
<td>23.7</td>
<td>612</td>
</tr>
<tr>
<td>D</td>
<td>39</td>
<td>6.3</td>
<td>24</td>
<td>627</td>
</tr>
</tbody>
</table>

Table 7.4. Refining levels.

<table>
<thead>
<tr>
<th>ID</th>
<th>Specific load, Ws/m</th>
<th>Net refining energy, kWh/ton</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.87</td>
<td>70</td>
</tr>
<tr>
<td>2</td>
<td>1.37</td>
<td>110</td>
</tr>
<tr>
<td>3</td>
<td>1.99</td>
<td>160</td>
</tr>
</tbody>
</table>

The pulp samples were characterised using FiberMaster, Karlsson and Fransson (1994). Figure 7.4 illustrates the fibre length distribution of the pulp samples. It shows that the fibre population at smaller fibre lengths increases while the population at larger fibre lengths decreases when more energy is consumed during the refining, which means that the fibres are shortened by refining.

Figure 7.4. Fibre length distribution for the different pulp samples refined to different levels. A1-D3 see Tables 7.2 and 7.4.

The fibre properties for these pulp samples are plotted against the net refining energy in Figure 7.5. The fibre curl index and the fibre form factor are the same concept, just ex-
pressed in different ways. The fibre flexibility is defined as the difference in the fibre form factor between two different flow velocities in the measurement. The results indicate that the fibre length decreases when increasing the net refining energy. The other fibre properties, such as fibre width, fibre coarseness and fibre curl index, are not significantly affected by the refining. The fibre flexibility was expected to increase by refining, but it did not show up in this characterisation. Three pulps were made from sawmill chips, whereas one was made from roundwood, and has a slightly lower fibre width.

![Figure 7.5. Fibre properties vs. net refining energy for different pulp samples. A-D see Table 7.2.](image)

The fibre suspension flocculation of these pulp samples was studied using the secondary contraction. The flow velocities were 8 m/s and 16 m/s respectively before and after the contraction. Previous studies have shown that fibre length has a significant influence on fibre flocculation in suspensions, Kerekes and Schell (1995), Kerekes (1995), Beghello and Eklund (1997), which was confirmed in this investigation. Among all measurable fibre properties, fibre length is the only one showing an evident effect on fibre suspension flocculation, shown in Figures 7.6, 7.7 and 7.8. Although fibre aspect ratio, L/d, and fibre curl is expected to have an influence on fibre flocculation in suspensions, the fibre length
is clearly the dominating factor. The conclusion is that the variations in pulp properties, such as bleaching sequences, curl index, coarseness, have little effects on fibre suspension flocculation as long as the comparison is limited to Nordic softwood mixtures.

The mean floc size is plotted in Figure 7.6 against the fibre length in MD and CD respectively both before and after the secondary contraction. The mean floc size in MD increases with the fibre length both before and after the contraction. The mean floc size in CD increases slightly with the fibre length before the contraction, while it remains roughly at a constant level after the contraction. Figure 7.7 shows the mean floc aspect ratio as a function of fibre length, both before and after the secondary contraction. Since longer fibres give longer flocs in MD, they can be expected to stretch more during the flow contraction.

Figure 7.6. Influence of fibre length at a fibre concentration of 5 g/l on mean floc size in MD and CD respectively for bleached softwood kraft pulp suspensions at flow velocities of 8 m/s and 16 m/s respectively before and after the secondary contraction.
Figure 7.7. Influence of fibre length at a fibre concentration of 5 g/l on mean floc aspect ratio, for bleached softwood kraft pulp suspensions at flow velocities of 8 m/s and 16 m/s respectively before and after the secondary contraction.

Figure 7.8. Influence of fibre length at a fibre concentration of 5 g/l on mean floc area reduction during contraction passage, for bleached softwood kraft pulp suspensions. The flow was accelerated from 8 m/s to 16 m/s by the secondary contraction.

The mean floc area reduction increases with the fibre length, illustrated in Figure 7.8. According to the concept of the crowding factor, Kerekes and Schell (1992), there is more fibre flocculation in the suspension with longer fibres than with shorter fibres. Thus, less free water exists between the fibre flocs with longer fibres, so the flocs need to compress more in order to pass the contraction, as also discussed by Björkman (1999). The fibre length effect is similar to the fibre concentration effect, discussed in Section 7.1.
7.3. Flow Velocity

For the unbleached softwood kraft pulp suspension, used in Section 7.1, images were taken before and after the secondary contraction at fibre concentrations of 3, 5 and 7 g/l. The flow velocities were set at 6, 7 and 8 m/s before the secondary contraction and correspondingly 12, 14 and 16 m/s after the secondary contraction. The mean floc size in MD and CD respectively of the suspension at different flow velocities is plotted against the fibre concentration in Figure 7.9. It shows that an increase of the absolute flow velocity has an insignificant effect on the fibre floc size both in MD and CD.

Figure 7.9. Mean floc size in MD and CD respectively for a suspension of unbleached softwood kraft pulp at different fibre concentrations at different flow velocities respectively before and after the secondary contraction.

7.4. Medium Temperature

It has also been known since the late 1930s that high stock temperatures enhance fibre flocculation in suspension, and it has been confirmed by Hourani (1988). Once it was re-
garded that fibres became softer when the temperature increased, and it was assumed that such an increase of the fibre flexibility caused an increase of fibre flocculation. However, it has been known that cellulose gel structures will not appreciably soften when the temperature increases, Lindström et al. (1987). Although lignin becomes softer when the temperature increases, overall there will be no or less effect of temperature on fibre flexibility in the present study, because softwood kraft fibres contain only little lignin. Moreover, both the experiment by Wikström and Rasmuson (1998) and the simulation by Ross and Klingenberg (1998) suggested that an increase of the fibre flexibility should decrease fibre network strength. Therefore, the mechanism of temperature influence on fibre suspension flocculation is presumably not caused by fibre flexibility. Beghello (1998) attempted to study the effects of medium viscosity on fibre flocculation but he did not notice any effect. In this investigation, we also decided to vary the medium viscosity through a change of medium temperature. The unbleached softwood kraft pulp suspension, used in Section 7.1, was studied at a temperature range of 25-50 °C.

The water viscosities at different temperatures were obtained from the Handbook of Chemistry and Physics, and were converted to SI units. The mean floc size in MD is plotted against the water viscosity in Figure 7.10. The mean floc size increases with an increasing fibre concentration, and decreases with an increasing water viscosity. The water viscosity increases with a decreasing temperature, so the mean floc size decreases with a decreasing medium temperature. The flocculation index in MD is also plotted against the water viscosity in Figure 7.11, setting the suspensions at 50 °C as the corresponding references for each series. Firstly, the mechanism of the flocculation reduction may be that the turbulence in suspensions is suppressed by an increase of water viscosity. Secondly, the inertia/viscosity ratio increases at a higher temperature, which increases fibre-fibre collisions leading to floc generation. Although the water temperature range in this investigation was quite narrow, the effect was obvious.

![Figure 7.10. Influence of water viscosity on mean floc size in MD for an unbleached softwood kraft pulp suspension at different fibre concentrations at a flow velocity of 8 m/s after the headbox nozzle.](image)

38
Figure 7.11. Influence of water viscosity on flocculation index in for an unbleached soft-wood kraft pulp suspension at different fibre concentrations at a flow velocity of 8 m/s after the headbox nozzle. The suspensions at 50 °C were set as the corresponding references for each series.

It was traditionally believed that fibre flocs could be disintegrated by turbulent shear, however, it is now known that such turbulent shear can actually create fibre flocs, as stated by Norman and Söderberg (2001). It is possible to fluidize a fibre suspension by a high input of turbulent energy, but it is not possible to avoid fibre network generation when the turbulence decays. The fibres strive to recover their natural shapes and become restricted and interlocked by the surrounding fibres. The reason why Beghello (1998) had not seen the viscosity effect might have been that the suspensions he observed were highly turbulent, not as in this investigation that the suspensions are observed after the headbox nozzle contraction, where turbulence has decayed, described in Section 2.

The papermaker has generally little opportunity to change the temperature in the forming section of the paper machine, since the temperature is set by designs for system closure and energy conservation. Thus, the temperature effects on papermaking are usually neglected. As shown in this investigation, there is, however, a rather significant effect of temperature on fibre suspension flocculation, pointing to one obstacle of modern paper-making.
8. CHEMICAL INFLUENCES ON FIBRE SUSPENSION FLOCCULATION (PAPER V & VI)

Besides the physical influences such as fibre concentration, fibre length, and so on, chemical additives and fibre surface modifications also have an appreciable impact on fibre suspension flocculation and paper sheet formation. The most common case is the significant impact of retention aids, which are always used in modern papermaking to retain fines, fillers, and functional chemical additives. The chemical aspects of retention aids have been reviewed by Lindström (1989), Swerin and Ödberg (1997). There are different types of flocculation generation mechanisms, such as patching, bridging, network flocculation, Eklund and Lindström (1991). Since retention aids are flocculants, such additives generally deteriorate sheet formation. Thus, the papermaker is seeking to balance formation and retention, as illustrated in Figure 8.1. In spite of claims in the conference and the trade literature, there is little or no information as to whether different retention aids give different formation/retention relationships. In this thesis work, the research was aimed on the potential chemical ways to disperse fibres in suspensions. No attention was thus given to retention/formation relationships.

![Figure 8.1. Formation number vs. filler retention for fine paper containing different amounts of ground calcium carbonate (GCC) produced in the FEX pilot paper machine trials at STFI-Packforsk. Courtesy of EKA Chemicals, Sweden.](image)

Formation aids, contrary to retention aids, are seldom used in papermaking. The formation aids may be grouped into the following classes: additives increasing the dispersion medium viscosity, Soszynski and Kerekes (1988), Zhao and Kerekes (1993); formation aids:
class I, gums and mucilages believed to decrease the coefficient of friction between fibres, De Roos (1958); formation aids: class II, high molecular weight polymers affecting the rheological properties of the suspending media, Wasser (1978), Lee and Lindström (1989). Although the use of viscosity modifiers, such as gums and mucilages, have been known by hand-sheet makers in the far-east for centuries, the use of formation aids have never really taken off commercially in modern paper manufacture. In this thesis work, the research was more focused on the recently developed chemical technologies for the formation improvement. The magnitude of the effect was also compared to a common used flocculant, cationic polyacrylamide, and a formation aid, anionic polyacrylamide. Fibre suspension flocculation was evaluated by the power spectrum method, described in Section 4.2.

8.1. Retention Aid

The effect of a cationic polyacrylamide, C-PAM, Percol 292, a type of bridging retention aid, on fibre suspension flocculation of an unbleached softwood kraft pulp refined to different levels was studied at a fibre concentration of 5 g/l in this investigation. C-PAM was online dosed into the flow loop system by the piston after the pump, and was mixed with the fibre suspensions by the inline static mixer.

The mean floc size increases with an increasing addition level of C-PAM, as shown in Figure 8.2. The mean floc size decreases with an increasing refining level, which is mainly due to refining shortening the fibre length, discussed in Section 7.2.

![Figure 8.2](image)

*Figure 8.2. Influence of C-PAM addition level on mean floc size in MD for suspensions of an unbleached softwood kraft pulp at different refining levels at a fibre concentration of 5 g/l at a flow velocity of 7 m/s after the headbox nozzle.*

The flocculation index in MD is also plotted against the addition levels of C-PAM in Figure 8.3, setting the suspensions without C-PAM as the corresponding references for each
series. The flocculation indexes are positive, which means that the suspensions with the addition of C-PAM were flocculated compared with the corresponding suspensions without C-PAM. The flocculation index also increases with an increasing addition level of C-PAM at different refining levels, confirming previous results, such as those reported by Wågberg and Lindström (1987a), Solberg and Wågberg (2003).

![Figure 8.3](image-url)  
*Figure 8.3. Influence of C-PAM addition level on flocculation index in MD for suspensions of an unbleached softwood kraft pulp at different refining levels at a fibre concentration of 5g/l at a flow velocity of 7 m/s after the headbox nozzle.*

### 8.2. Formation Aid

High molecular weight non-adsorbing polymers, formation aids, class II, can improve the dispersion of fibre suspension. In this investigation, the effect of such a formation aid, anionic polyacrylamide, A-PAM, Percol 156, on fibre suspension flocculation of the refined unbleached softwood kraft pulp, 25 °SR, used in Section 8.1, was studied at different fibre concentrations. A-PAM was online dosed into the flow loop system by the piston after the pump, and mixed with the fibre suspension by the inline static mixer. The corresponding A-PAM effect on paper formation is discussed in Section 9.

The mean floc size increases with an increasing fibre concentration, while it decreases with an increasing addition level of A-PAM, as shown in Figure 8.4. The flocculation index in MD is also plotted against the addition levels of A-PAM in Figure 8.5, setting the suspensions without A-PAM as the corresponding references for each series. The flocculation indexes are negative, which means that the suspensions with the addition of A-PAM were deflocculated compared with the corresponding suspensions without A-PAM. The flocculation index also decreases with an increasing addition level of A-PAM at different fibre concentrations.
Figure 8.4. Influence of A-PAM addition level on mean floc size in MD for an unbleached softwood kraft pulp suspension at different fibre concentrations at a flow velocity of 7 m/s after the headbox nozzle.

Figure 8.5. Influence of A-PAM addition level on flocculation index in MD for an unbleached softwood kraft pulp suspension at different fibre concentrations at a flow velocity of 7 m/s after the headbox nozzle.

The mechanism of A-PAM on flocculation has been discussed before by Lee and Lindström (1989). A-PAM is not adsorbed onto chemical pulps and the addition levels are too low to affect medium viscosity. Hence, it is not expected that the friction between fibres is affected, and as the viscosity is constant, there will be no effect on the inertia/viscosity ratio. Instead, high molecular weight polymers are known to suppress the turbulence in suspensions, commonly recognized as the drag reduction effect, Mark et al. (1986), decreasing the likelihood of building in stresses in the fibre suspension networks.
8.3. Surface Modification: Ionic

Recently, it has been observed that carboxymethylation of cellulosic fibres can improve formation through a decreased fibre/fibre friction coefficient, Beghello and Lindström (1998). This is, however, not a very practical method of improving sheet formation, but clearly indicated that surface charges may have a strong influence on fibre flocculation. Subsequently, a new surface grafting method was developed, Laine et al. (2000), by which it is possible to graft carboxymethyl cellulose, CMC, onto cellulosic fibre surfaces with a very high surface specificity. This method allows studies of how the surface charge affects sheet consolidation and strength properties, Laine et al. (2002a; b), Laine et al. (2003a; b). This investigation was specifically aimed on the role of surface charge and ionic form on fibre flocculation.

A refined ECF-bleached softwood kraft pulp was used for the surface modification. CMC was grafted onto the fibre surfaces, in accordance with the previous published method, Laine et al. (2003a). The pulp was grafted with CMC at 120 °C in 0.05 M CaCl₂ for 2 hours. The addition of CMC was 40 mg/g according to the dry fibre weight. The pulp grafted with CMC was first washed with deionised water in its Ca-form, and then transferred to its H-form and Na-form respectively, in accordance with the previously published method, Laine et al. (2002a). The amount of irreversibly grafted CMC was determined by conductometric titration of the washed pulp, and is listed in Table 8.1 as well as the surface charge for the pulp in its Ca-form. As shown in Table 8.1, the surface charge has been appreciably increased by CMC-grafting. Some desorption of CMC occurs, when washed in the sequence of Ca-form, H-form and Na-form, as has been discussed before, Laine et al. (2000).

<table>
<thead>
<tr>
<th>Pulp</th>
<th>Grafted CMC (mg/g)</th>
<th>Surface Charge (µmol/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Without CMC</td>
<td>0</td>
<td>2.5</td>
</tr>
<tr>
<td>CMC H-Form</td>
<td>29.9</td>
<td>–</td>
</tr>
<tr>
<td>CMC Na-Form</td>
<td>28.4</td>
<td>–</td>
</tr>
<tr>
<td>CMC Ca-Form</td>
<td>34.7</td>
<td>40.1</td>
</tr>
</tbody>
</table>

Three pulp samples grafted with CMC in Ca-form, H-form and Na-form respectively as well as one pulp sample treated at the same conditions without CMC were studied in this investigation. Images of fibre suspensions for these four pulp samples were taken at a fibre concentration of 5 g/l both before and after the secondary contraction at flow velocities of 8 m/s and 16 m/s respectively.

The mean floc size and the flocculation index in MD for the suspensions of the CMC-grafted pulps in different ionic forms and the reference pulp treated without CMC are illustrated in Figure 8.6. It shows that both the mean floc size and the flocculation index are in the order of without CMC > CMC Ca-form > CMC H-form > CMC Na-form, which indicates that the reduction of fibre flocculation by CMC-grafting is in the order of Na-form > H-form > Ca-form.
Figure 8.6. Mean floc size and flocculation index in MD for suspensions of the CMC-grafted pulps in different ionic forms and the reference pulp cooked without CMC at a fibre concentration of 5 g/l at a flow velocity of 8 m/s after the headbox nozzle. The columns indicate the mean floc size. The dots and lines indicate the flocculation index.

The mutual repulsion between the surface charges leads to an electrostatic repulsion, resulting in a decreased friction coefficient between the fibres, which in turn, decreases the flocculation. The more dissociated the fibre carboxyl groups are, the higher is the repulsion and the more deflocculated the fibre suspension will be. The surface conformation of the attached CMC will become more expanded when in its Na-form, as has been discussed in detail before, Laine et al. (2003a). This also provides an efficient electrosteric barrier between the dispersed fibres, resulting in a lower fibre friction coefficient.

Figure 8.7. Mean floc aspect ratio for suspensions of the CMC-grafted pulps in different ionic forms and the reference pulp cooked without CMC at a fibre concentration of 5 g/l at flow velocities of 8 m/s and 16 m/s respectively before and after the secondary contraction.

The mean floc aspect ratio for suspensions of different pulp samples remained roughly at the same level, as illustrated in Figure 8.7. While the floc area reduction is in the order of
without CMC > CMC Ca-form > CMC H-form > CMC Na-form, as illustrated in Figure 8.8. The reason might be that the fibre flocculation is decreased in such an order that fibre flocs with grafted CMC are compressed to a lesser extent simply because the flocs are smaller. This effect is similar to the effects of fibre concentration and fibre length, discussed in Sections 7.1 and 7.2 respectively. The reason might also be that CMC-grafting decreases the friction between fibres in such an order. When the friction is decreased, the elasticity of the fibre network is also decreased. When fibre flocs with grafted CMC passed through the contraction, the stretching of the flocs in MD by the pressed-out water causes less corresponding shrinkage in CD.

![Graph](image.png)

**Figure 8.8.** Mean floc area reduction for suspensions of the CMC-grafted pulps in different ionic forms and the reference pulp cooked without CMC at a fibre concentration of 5 g/l. The flow was accelerated from 8 m/s to 16 m/s in the secondary contraction.

### 8.4. Surface Modification: Non-Ionic

There has been some recent attention in exploiting xyloglucan, a non-ionic polymer extracted from tamarind gum, for various applications in papermaking. Interestingly, this non-ionic polymer has a strong affinity to cellulosic surfaces, and it has also been found that xyloglucan improves hand-sheet formation, Christiernin et al. (2003). This prompted us to study the effects of xyloglucan on fibre flocculation. A comparison of the different treatments with cationic polyacrylamide, C-PAM, discussed in Section 8.1, and anionic polyacrylamide, A-PAM, discussed in Section 8.2, was also made in order to compare the magnitude of the dispersing effects.

The refined unbleached softwood kraft pulp, 25 °SR, was used for the study of xyloglucan, the same as for the study of C-PAM and A-PAM. Xyloglucan was directly dosed and mixed in the stock tank at a fibre concentration of 5 g/l and at different addition levels of 2.5, 5, 10, 20 and 40 mg/g according to the dry fibre weight. Images of the fibre suspensions were taken at different times after the dosage from 0.5 h to 30 h, both before and after the secondary contraction at flow velocities of 8 m/s and 16 m/s respectively.
The flocculation index in MD for suspensions with different xyloglucan addition levels is plotted against the time after dosage in Figure 8.9, setting the suspension without xyloglucan as the reference for each series. The flocculation indexes are negative, which means that xyloglucan functions as an effective dispersant. These results confirm the previous observation that sheet formation was improved by the addition of xyloglucan, Christiernin et al. (2003), in accordance with the fact that the reduction of fibre suspension flocculation leads to an improvement of paper formation, discussed in Section 9.

![Figure 8.9](image)

Figure 8.9. Influence of xyloglucan addition level on flocculation index in MD vs. time after dosage for an unbleached softwood kraft pulp suspension at a fibre concentration of 5 g/l at a flow velocity of 8 m/s after the headbox nozzle.

The adsorption of xyloglucan onto cellulosic fibres is quite slow, Christiernin et al. (2003), but the effect on the flocculation index is immediate. This effect of xyloglucan on fibre flocculation is presumably an effect of xyloglucan on reduction of the friction between fibres, and the friction is a surface effect. The slow adsorption of xyloglucan is due to the slow penetration of xyloglucan molecules into the cell wall of fibres, Christiernin et al. (2003), whereas the fibre surface is immediately saturated with xyloglucan. Therefore, there is no time effect of xyloglucan on fibre flocculation.

The mean floc size for the suspension is plotted against the addition levels of xyloglucan in Figure 8.10. It shows that the mean floc size decreases when increasing the addition level of xyloglucan. The flocculation index for the suspension with xyloglucan is plotted against the addition levels in Figure 8.11, as well as the flocculation index for the suspension with a retention aid, flocculant, C-PAM, and a formation aid, class II, A-PAM, for a comparison. The mechanism of C-PAM on increasing fibre flocculation is linked to the ability of C-PAM to bridge the fibres in the suspension, and the mechanism of A-PAM on decreasing fibre flocculation is to suppress the turbulence. C-PAM and xyloglucan are being adsorbed onto the fibre surface, whereas A-PAM acts in the solution phase. If the same level of fibre flocculation reduction is required, more xyloglucan is needed compared with A-PAM.
Figure 8.10. Influence of xyloglucan addition level on mean floc size in MD for an unbleached softwood kraft pulp suspension at a fibre concentration of 5 g/l at a flow velocity of 8 m/s after the headbox nozzle.

Figure 8.11. Comparison of A-PAM, C-PAM, and xyloglucan effects with different addition level on flocculation index in MD for an unbleached softwood kraft pulp suspension at a fibre concentration of 5 g/l after the headbox nozzle. The suspension without chemicals was set as the reference for each series.

The mean floc aspect ratio does not change much when increasing the addition level of xyloglucan, similar to the case of CMC-grafting, discussed in Section 8.3. While the mean floc area reduction remains roughly at the same level when increasing the addition level of
xyloglucan. This might be that the breakage of fibre flocs during the flow contraction increases when the friction between fibres is decreased by adding xyloglucan. The mean floc area reduction does not represent the total floc area reduction, discussed in Section 7.1.
9. RELATIONSHIP BETWEEN FIBRE SUSPENSION FLOCCULATION AND PAPER FORMATION (PAPER IV & V)

The concepts of fibre suspension flocculation and paper formation are not the same, and the situations for fibre flocs in a suspension and in a sheet are different. In a paper sheet, a fibre floc is a two-dimensional entity, and is tightly fastened by hydrogen bonds. While in a flowing suspension, a fibre floc is a three-dimensional network, and is loosely entangled by mechanical forces. Moreover, during the dewatering, there is an inherent self-healing effect, which improves the formation, Norman and Söderberg (2001). However, there still is a close relationship between fibre suspension flocculation and paper formation, which has also been found in previous research, such as Linhart et al. (1987).

Paper sheets of the pulp samples, used in Section 7.2, were produced on the FEX pilot paper machine at STFI-Packforsk, Röding and Norman (1986), at a forming concentration of 4.5 g/l with a slice opening of 14 mm, at a machine speed of 600 m/min. The paper sheets (60 g/m²), produced in the trials, were characterised by their formation numbers, Johansson and Norman (1996). The formation numbers of the paper sheets are plotted against the fibre length in Figure 9.1. When the fibre length is increased, the formation number is increased, which means that the paper formation deteriorates. It should be observed that large scale formation increases more than small scale formation with an increasing fibre length. Similar results may be found in previous research, such as Mohlin (2001). Comparing Figure 9.1 with Figure 7.6, the conclusion can be made that fibre suspension flocculation and paper formation show the same trend with an increasing fibre length.

![Figure 9.1. Influence of fibre length on formation numbers of paper sheets produced in the FEX pilot paper machine trials. 0.3-3 mm represents small scale formation, 3-30 mm represents large scale formation, and 0.3-30 mm represents total formation.](image)
Paper sheets of the pulp sample, used in Section 8.2, with the A-PAM addition levels of 1 and 1.7 mg/g according to the dry fibre weight, were produced on the FEX pilot paper machine at STFI-Packforsk, Röding and Norman (1986), at a forming concentration of 10 g/l with a slice opening of 10 mm, at a machine speed of 400 m/min. The paper sheets (100 g/m²), produced in the trials, were characterised by their formation numbers, Johansson and Norman (1996). The effect of A-PAM on the paper sheet formation numbers is shown in Figure 9.2. When increasing the addition level of A-PAM, the formation number decreases, which means that the paper formation is improved, confirming the previous result reported by Lee and Lindström (1989). The effect of A-PAM is more significant at larger wavelengths than at smaller wavelengths, which validates the feasibility of calculating the floculation index merely at larger wavelengths in our analysis method, described in Section 4.2.3. Comparing Figure 9.2 with Figures 8.4 and 8.5, the conclusion can be made that when the fibre suspension flocculation is decreased, the paper formation is improved.

It should be noted that the addition of 1.7 mg/g A-PAM according to the dry fibre weight resulted in an appreciable change in the flocculation index of fibre suspension from 0 to 0.3, discussed in Section 8.2. This change in the flocculation index resulted in an improved formation number at larger wavelengths of paper sheet from 19.5 to 13.5, which is a very significant improvement (see also Figure 9.3). Hence, the fibre surface modifications, CMC-grafting and xyloglucan addition, discussed in Sections 8.3 and 8.4, are expected to have a very significant effect on paper sheet formation.

Since the good correlation between fibre suspension flocculation and paper formation, it is feasible and sensible to study the corresponding fibre suspension flocculation in the flow loop system instead of to study the paper formation in the pilot paper machine trials for the reasons of convenience and economy. If the results obtained from the corresponding fibre suspension are remarkable and attractive, further experiments can then be focused on the paper sheet in the pilot paper machine trials for verification.
Figure 9.3. Beta radiography of paper sheets. Grammage: 100 g/m², size of paper samples: 50x50 mm², MD in vertical axis. Left: without A-PAM. Right: 1.7 mg/g A-PAM.
10. FUTURE WORK

In this thesis work, the extra flow contraction after the headbox nozzle has been proved able to partially break up fibre flocs in a suspension. Since the secondary contraction is changeable, further research could be focused on the effect of different flow contraction ratios on the breakage of fibre flocs. The headbox feeding part in the flow loop system can be modified, in order to introduce a single fibre floc in a water suspension. Thus, the behaviour of a single fibre floc in the flow contraction can be studied. Simultaneous observations in both the MD-CD and MD-Z planes would make it possible to study the flocculation phenomena in three dimensions.

The wavelet transform has been introduced to evaluate fibre suspension flocculation, while such an analysis method can be further developed. Although currently too time consuming, with the development of fast computers and programming techniques, it will be possible to evaluate individual fibre flocs in a suspension in future. Thus, the distributions of fibre floc size, fibre floc location, and even fibre floc orientation in suspensions can be investigated. If the ratio of broken to unbroken fibre flocs during the flow contraction can be calculated automatically by a computer, the strength of fibre flocs in suspensions can be characterised.

The relations between fibre properties and fibre suspension flocculation have been studied. However, since fibres have a rather wide distribution of length, width, and so on, relations for the average values are not always comprehensive. For that reason, rayon fibres, also swelling in water, may be used to simulate wood fibres, because rayon fibres can be synthesized to an exact width and cut to an exact length. Thus, the relations between fibre properties and fibre suspension flocculation could be further clarified.

Several methods for reduction of fibre suspension flocculation have been investigated. The compatibility between retention and formation aids can be further studied using the flow loop system. Experiment can be performed, by dosing retention aids in the stock tank and dosing formation aids online using the piston after the pump, to study the combined effects on fibre suspension flocculation. Further research can be focused on fibre surface modifications, which decrease fibre suspension flocculation by reducing the friction between fibres, while still allowing a good filler retention. Hence, it may be possible to solve the conflict between good paper formation and high filler retention in papermaking.
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12. REFERENCES


