FRACTIONATION OF MICROFIBRILLATED CELLULOSE AND ITS EFFECTS ON TENSILE INDEX AND ELONGATION OF PAPER

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ABSTRACT

In this work we examine the fractionation of microfibrillated cellulose (MFC) and its effect on the physical properties of paper. In the first part of this work, we evaluate the fractionation of commercially available MFC using multiple stages of traditional fractionation unit operations, namely a hydrocyclone, a pressure screen, and a novel technique based upon the control of the threshold for motion in a weak gel. The results indicate that a smaller fibre length average fraction could be obtained using the gel separation technique than using multiple stages of separation in both the hydrocyclone and the pressure screen. With the gel technique we were able to reduce the average fibre length of the MFC from 221 µm to 100 µm. In the second part of the work, composite paper samples were formed by addition of fractionated and non-fractionated MFC to chemical wood fibres and the strength of the resulting composites were studied. The results showed 25% improvement in tensile index by addition of MFC and an additional 10% improvement in tensile strength by addition of fractionated MFC using the gel technique.

INTRODUCTION

The potential of nanotechnology is far-reaching. One promising area is the development of nanocomposite materials that are light-weight with high-strength. In particular, microfibrillated cellulose (MFC) reinforced polymer composites have recently found applications in the automotive, aerospace and construction industries due to their strength as well as their active or “smart” properties [1,7]. One of the remaining open questions is that of a reliable manufacturing method to fractionate or separate MFC from cellulose fibre for large scale manufacturing purposes. This class of problem is gaining interest rapidly in the Pulp & Paper industry.

Before we discuss the relevant separation techniques available, it is instructive to first review the source of MFC and its potential benefits on the physical properties of paper. It is well known that an excellent source of MFC is the cellulose fibrils found in the walls of either hardwood vessel elements or pulp fines from chemical pulps. . Turbak et al. [17] has explained the process that results in production of MFC in details. Once these elements have been separated and processed to create MFC, small additions of this to traditional polymer-reinforced composites have been shown to increase the elastic modulus more than 1000 times [5, 6, 9]. For MFC reinforced paper, the application which motivates the present study, Iwatake et. al. [8], demonstrates a 25% increase in the tensile strength at a 10% (wt/wt) MFC content.

The MFC recovered from most pulping operations has an inherent length distribution. It is commonly known that the length and aspect ratio distributions of the reinforcing MFC dramatically affect the strength properties of the resulting composites [2, 4,7]. The motivation of the work stems from the need to remove the larger particles, or fibrils, from MFC gels to improve its inherent properties. A number of unit operations currently exist for fractionation purposes, i.e. hydrocyclones and pressure screens, which are able to separate papermaking fibres suspension, but efficiency of these processes known to be low. This has been discussed previously by Madani et al [13]. As a result, in this work we report on the fractionation of a commercially-available MFC suspension in a sequence of hydrocyclone and pressure screening steps, as well as in a novel technique advanced by Madani et. al. [13]. The novel methodology involves centrifugation in a viscoplastic carrier fluid. Following this we assess the reinforcing ability of these purified samples by assessing the tensile strength of MFC reinforced paper. This would follow a similar approach to the work of Nakagaito et. al. [14] which produced MFC-reinforced composite sheets in a process similar to papermaking. Their results show increase in physical properties of produced composite as MFC is added.

METHODS AND MATERIALS

Microfibrillated cellulose (MFC) samples, obtained from JRS (www.jrs.de), were used in this work work (with the commercial name of NFC). The samples had an average fibre length of 221 µm as determined using an L & W STFI Fibremaster [10]. The MFC sample was then fractionated using one of the three techniques. In the first technique, the MFC was fractionated using four hydrocyclone stages. Here we employed a C-1201 Y Microspin polypropylene hydrocyclone (www.natcogroup.com) with 10mm diameter and operated at 5 bar at a feed flow rate of 4.2 lpm; the initial consistency was set at 0.2% and accept to reject ratio was set to 6 to 4. After each pass, the reject fraction was collected, its fibre length distribution measured, and then re-fractionated by passing it through the hydrocyclone again. As reported by Bliss [3], Paavilainen [15] and Wood and Karnis [18,19], in small size hydrocyclones, the average fibre length increases in the “accept” stream while this average reduces in the “reject” stream. In normal conventional size hydrocyclones an opposite trend has been observed [11, 12 and 16]. In the second technique we employed a Metso FS-03 pressure screen to fractionate the initial MFC sample. Here, we collected the accept and progressively fractionated the sample by passing it through screens with progressively smaller slot sizes, namely 0.13, 0.09 and 0.06 mm. It should be noted that for traditional papermaking suspensions fractionation generally proceeds with screens operating with screen baskets with holes. In our cases the slotted screen baskets are extremely small and we anticipate some fractionation, albeit not optimal. To compensate for this, in total five stages of screening were performed and we screened the sample through the smallest slot size three times. The screen was operated with five different initial concentrations, that is 0.1%, 0.3%, 0.5%, 0.6% and 1%, with a foil type rotor rotating at 3500 rpm and at a reject ratio of 60%. After each step the collected samples were diluted to reach the minimum required volume of 15 L. As a result, along the stages the samples became very dilute and effect of initial concentrations became insignificant.

The last set of fractionation experiments was based on controlling the criteria of motion of particle suspended in a weak gel during centrifugation; see Madani et. al. 2010 for details of the technique. Here an Eppendorf 5804 centrifuge was used with six 50 mL cylindrical containers and the rotational velocity up to 5000 rpm. In the experiments a 0.16% (wt/wt) Carbopol solution (Noveon) was dissolved gently in DI (deionized) water and then neutralized using *NaOH* solution. The resulting fluid was a clear fluid with density of water and a yield stress of approximately 1 Pa. The MFC suspension was then mixed into the Carbopol solution and subjected to a prescribed rotational rate; four different initial concentrations were tested, i.e. 0.1%, 0.2%, 0.4%, and 0.6% (wt MFC/wt suspension). The top two third of the samples in each test tube was collected as fractionated MFC. Finally, the fractionated MFC (the retained fraction) was mixed into a chemical pulp (bleached hardwood) at 4 different mass fractions (0, 5, 10 and 15%). Standard handsheets (Scandinavian SCAN-C standard) were then formed using both initial and fractionated MFC and the tensile strengths of the paper samples were evaluated.

RESULTS AND DISCUSSION

The results of the fractionation studies are shown in Figure 1 and Figure 2. The first observation that can be made from these figures is that the pressure screen was relatively ineffective in removing the larger size particles in comparison to the other techniques. The fractionation in the pressure screen was attempted over a number of different initial concentrations and we find almost identical fractionation performance due to dilution of the samples. Caution must be used in interpreting these results as we employed slotted screen baskets; a more efficient fractionation may have occurred with screen baskets with holes. This speculation needs to be confirmed as to the best knowledge of these authors, no results are available in the literature indicating the efficiency of fractionation of MFC in pressure screens.

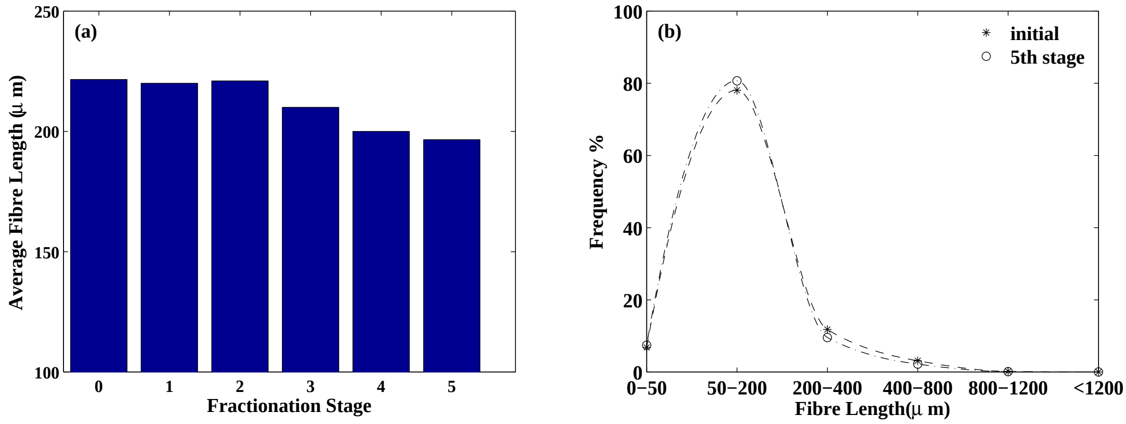
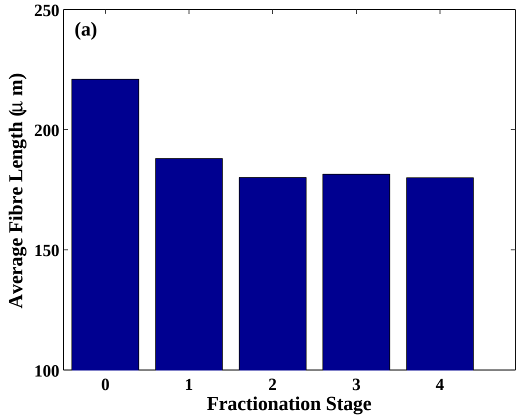


Figure 1-Fractionation of a 0.1% MFC suspension after five stages in a pressure screen. In each stage the accept stream was fed to the subsequent screen. The sizes of the slots in each fractionation stage are as follows: stage 1 - 0.13 mm, stage 2 - 0.09 mm and stages 3-5 - 0.06 mm. In panel (a), the average fibril length is shown. Stage 0 represents the initial fibre length distribution. In (b) a representative fibre length distribution is shown initially and after the fifth stage of separation.



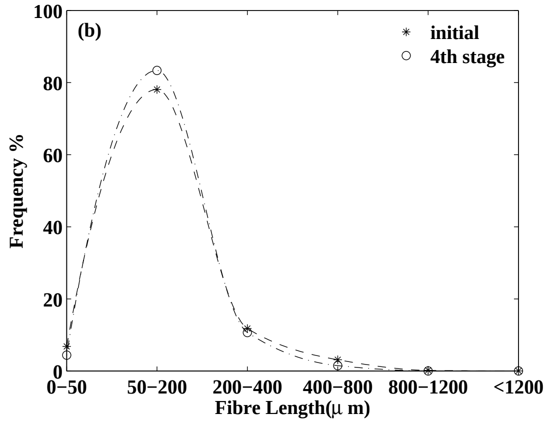


Figure 2- Fractionation of a 0.2% MFC suspension after four stages of a hydrocyclone. In each stage the reject stream was fed to the subsequent hydrocyclone. In panel (a), the average fibre length is shown. Stage 0 represents the initial fibre length distribution. In (b) a representative fibre length distribution is shown initially and after the fourth stage of separation.

As a result we display only one result which is representative of all cases tested. The second observation that can be made from these results is that the average fibre length diminished from 221 μm to approximately 180 μm after four fractionation stages in the hydrocyclone, see Figure 2. Finally, the largest reduction of fibre length was found using the gel fractionation technique (Figure 3); we observed very efficient fractionation as only one stage of this technique was used for all concentrations tested. The variation of fibre length distribution and frequency is shown in Figure 3 for 0.1% fibre consistency. It is important to note that increasing the fibre consistency in the gel fractionation has a significant effect on the fractionation result and as the consistency is increased, more fibres are moved to the bottom of the test tube and the yield efficiency (remaining mass in the system) reduces. The effect of consistency is shown in Figure 4. Increasing fibre consistency results in a smaller reduction in fibre length. This might be due to the formation of fibre networks and increase in fibre-fibre interaction. If we normalize the frequency of the fibres remaining in each sample by their initial frequency and call it F_n, we can observe the reduction on yield efficiency as concentration is increased (Figure 5).

Finally, to ensure that fractionation of the MFC samples had an effect on paper samples, we formed standard handsheets with additions of the MFC which was fractionated using the gel technique (retained fraction), see Figure 6. In both cases tensile index and strain at break of fractionated MFC reinforced papers are enhanced over the papers reinforced with the original MFC. This can be contributed to the increase in the hydrogen bonds, more uniform distribution of stresses due to extended surface area of MFC and increase in fibre-fibre contact area. In fact we report a 25% increase in the tensile index before fractionation and an additional increase of 10% in tensile index and 120% in strain at break after 10% (wt/wt) of the fractionated MFC was added to the samples. The only negative point of addition of MFC in handsheet making is increase in the drainage time[1]. For the case of 15% retained fraction, the drainage time of the tests was too long that we were forced to stop the experiments. (The dewatering time was 5 s, 6 s, 9 s and 11 s for no MFC added, 5 % MFC, 10 % and 15 % MFC added to the system while for the fractionated MFC it increased to 7 s for 5% and 13 s for 10 % and it was too long for 15 % MFC added to the pulp).

[1] The drainage time is the time required for water to pass through the fibre network and leave the handsheet former.

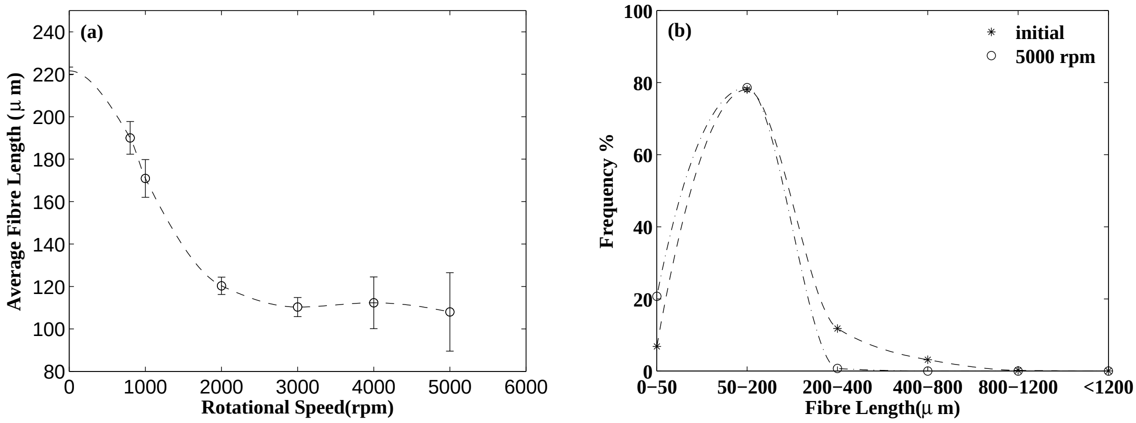


Figure 3-Fractionation of a 0.1% MFC suspension after centrifugation at different rotational rates. In panel (a), the average fibre length is shown as a function of rotational rate. In (b) a representative fibre length distribution is shown initially and after one stage of fractionation at 5000 rpm.

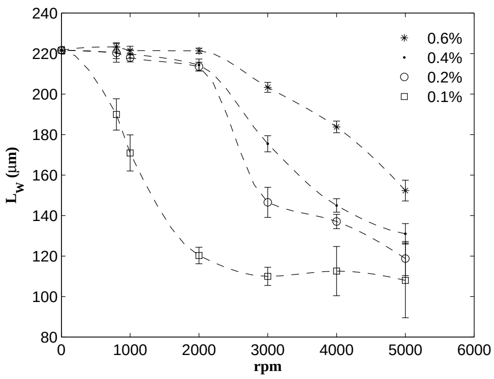


Figure 4-Effect of fibre consistency on length fractionation at different rorational velocities.

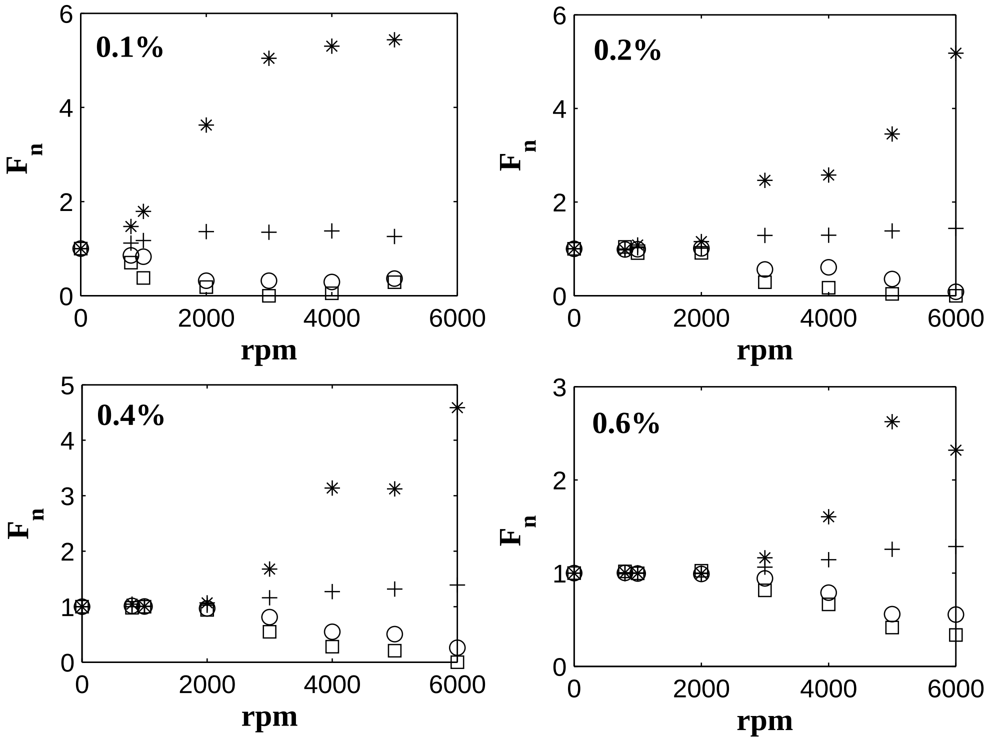


Figure 5-Variation of normalyzes fibre frequency F_n at different rotational velocities and fibre cocentrations for retained sample. '*', '+', 'circle' and 'square' stand for 0-50 µm, 50-200 µm, 200-400 µm and 400-longer.

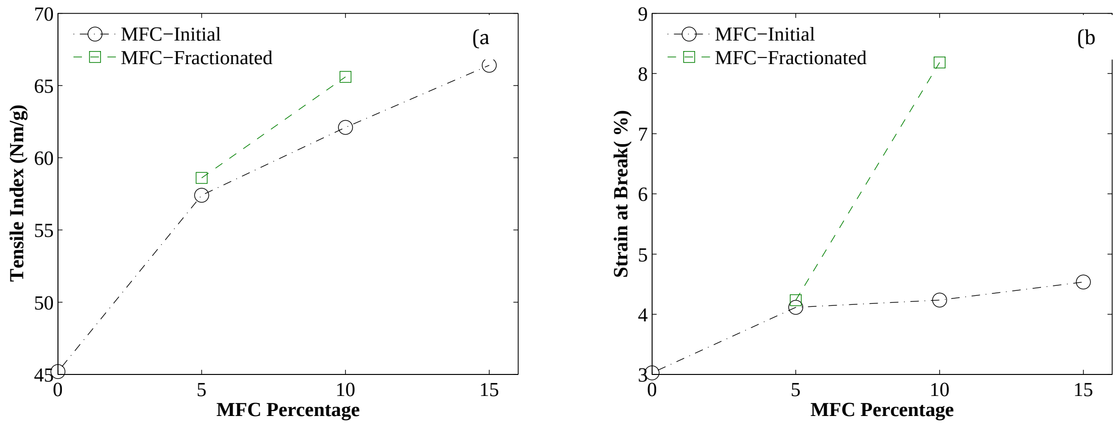


Figure 6-Changes in physical properties of MFC reinforced handsheets. In (a) tensile index increase and in (b) strain at break increase is shown before and after fractionated MFC is added to the handsheets.

SUMMARY AND CONCLUSIONS

Three different fractionation techniques were tested to remove relatively long fibres from two different MFC samples. The results showed that all the three methods are capable of removing longer fractions of MFC. The gel fractionation technique shows a larger reduction in fibre length in comparison to either the hydrocyclone and pressure screen for the conditions tested. As shown in Figure 7 only one stage of fractionation using gel technique is more efficient than multiple stages of fractionation using screen and hydrocyclone, for the conditions tested. Results of strength tests showed improvement in the tensile index and strain at break of MFC-reinforced handsheets.

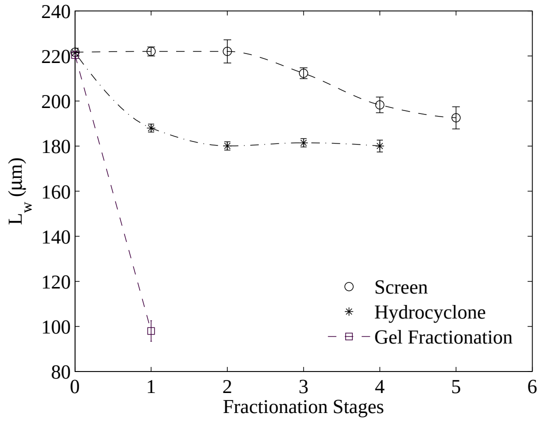


Figure 7-Comparison of 3 different fractionation techniques.

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