EFFECT OF MODEL AND FRACTIONATED TMP FINES ON SHEET PROPERTIES

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ABSTRACT

Two model fines, representing microfibrils and microgranules, and two fractions of TMP fines were used to demonstrate their effect on tensile strength and optical properties of handsheets formed from TMP and kraft fibers. The model microfibrils behave as a binder between fibers, thus improving tensile strength. The light scattering of kraft handsheets decreases but that of TMP handsheets increases. The model microgranules behave as a pigment by improving light scattering but preventing interfiber bonding. TMP fines, containing both types are capable of increasing tensile strength and light scattering simultaneously. The relation between tensile strength and light scattering depends on the proportion of fibrillated fines and granulated fines. The fractionated TMP fines of high surface area are shown to be very effective in improving the handsheet properties and the relation between tensile strength and light scattering is superior to that achieved by calcium carbonate filler.

INTRODUCTION

The effect of fines on a number of paper properties is well recognized, yet their actual role is not fully understood because of their heterogeneous
nature. The often used definition of fines, being the fraction of pulp passing through a 200 mesh screen, includes particles of different shapes, dimensions, chemical composition, surface properties, etc. This is particularly true in the case of mechanical fines composed of broken fibers, cell wall fragments, middle lamellae fragments, parenchyma, ray cells and bordered pits. Because of the wide range of different types of particles there is no standard classification for them. It has been suggested, however, that in a broad sense the fines can be divided into slime stuff and flour stuff [1]. Alternatively, the two categories can be described as fibrils and flakes [2,3] or ribbons and chunks [4]. The slime stuff consists mainly of swellable and flexible fibrillar particles originating from the cellulose-rich fiber wall. The flour stuff contains flake-like particles, i.e., fiber fragments and pieces of cell wall and middle lamella. Within each category there are particles of different sizes. In general, the slime stuff is considered a good bonding material while the flour stuff has poor bonding ability.

A number of studies dealing with the effect of mechanical fines on paper properties have been reviewed by Luukko [2]. In most of the contributions, the whole fraction of fines was used because of the difficulty to separate fines into distinct categories. The general conclusion is that in mechanical papers, where their content could be very high, fines are very important components dominating the paper properties. It has also been shown that the mechanical and optical properties of paper depend on the type of fines (kraft or TMP) rather than the type of fibers [5]. Introducing kraft fines in paper results in increased tensile strength and decreased light scattering in both chemical and mechanical fiber networks. On the other hand, TMP fines incorporated into a network of either kraft fibers or TMP fibers will increase both the tensile strength and the light scattering. However, when TMP fines are divided into two fractions, one consisting predominantly of fibrillar material and the other of flake-like material, a distinctly different effect on sheet properties is observed [3]. The fibrils are responsible for improving strength while the flakes improve the optical properties. This suggests that fractionated TMP fines could be used as an additive to control specific properties.

The objective of the present work is to investigate the effect of two fractions of TMP fines with different proportions of fibrillated and flake-like material. Two other model fines representing either fibrils or granules are microfibrillated cellulose and microcrystalline cellulose. Their effect on mechanical and optical properties of handsheets formed from either chemical or mechanical fibers is the main focus of this investigation.
EXPERIMENTAL

Materials

Fibers

Thermomechanical and unbeaten kraft fibers without fines were used. Fines were removed using a float-wash fractionator equipped with a 100 mesh screen. The amount of TMP fines passing the 100 mesh screen was 27%.

Fines

Two types of TMP fines fractionated in a small hydrocyclone [6] were used. The fraction of fines of high specific surface (HSF) containing pieces of fiber, fibrillar material and pieces of fiber wall of various sizes down to less than 0.5 μm represents 57% of the total mass of fines. The fraction of fines of low specific surface (LSF) containing mostly ray cells and intact pieces of fiber 10 to 200 μm in length represents 21% of the total fines. The specific surface was determined by turbidimetry [7].

Two model fines representing fibrillar and granular portions were used. Microfibrillated cellulose KY 100S (supplied by Daicel Chemical Industries, Japan) with average length 0.5 mm and diameter 0.01–0.1 μm (MFC), and microcrystalline cellulose (FMC Corporation) with average particle sizes 6 μm (MCC) and 100 μm (MCC-100).

Filler

Precipitated calcium carbonate PCC (Speciality Minerals) with average size of 1.6 μm was used.

Methods

Handsheets from mixtures of either TMP or softwood bleached kraft (SBK) long fibers and fines were prepared in a British Sheet Machine according to the standard method (CPPA C.4) and tested for tensile strength and optical properties. Handsheet consisted of 1 g of long fibers and different amounts of fines. No retention aid was used (to prevent the effect of flocculation) and consequently, to achieve high content of fines, considerable amounts were used (up to 0.75 g, 1 g, 5 g of fines, MCC and filler, respectively).

The amounts of retained fines, MCC and MFC were determined from the dried sample assuming that 1 g of fibers was always retained. The amount of retained calcium carbonate was determined by ashing the sample at 500°C.
RESULTS AND DISCUSSION

When dealing with the relation between tensile strength and opacity it is reasonable to consider the role of the surface area of fibers. In a random assembly of fibers, part of a fiber surface is in contact with another fiber at the point where they cross. Within this area hydrogen bonds between fibers can form. The number of interfiber bonds determines (in conjunction with other attractive forces, such as van der Waals) how strongly the fibers stick to each other and consequently the tensile strength increases with bonded area. The fiber surface outside the bonded area is in contact with air and thus presents an interface where scattering of light takes place. In the absence of light absorption (well bleached fibers) the light scattering is directly related to opacity.

The relation between bonded and unbonded area is sufficient to explain the effects caused by beating, filler and fines introduction. The inverse relation between tensile strength and opacity (which for white fibers is a function of light-scattering) is well known for chemical pulp fibers subjected to different levels of beating. Beating results in increased bonded areas due to better conformability and plasticity of the fiber wall and its fibrillation. This means that less of the fiber surface is available for scattering the light and thus the opacity decreases while tensile strength increases.

The effect of pigments incorporated into paper can be also viewed as a relation between bonded and unbonded area. Pigment contributes an additional interface for light scattering and therefore the opacity increases. However, the pigment particles prevent interfiber bonding and consequently tensile strength decreases. The result is again an inverse relation between tensile strength and opacity.

Chemical fines predominantly increase tensile strength. The chemical fines are mostly fibrillar with a high surface area. However, upon drying they collapse on the fiber surface and therefore they do not represent an additional interface for light scattering. Their collapse, though, enables them to form bridges between fiber surfaces, which will happen most likely around the points of fiber crossing where the surfaces are close. The result is then increased interfiber bonding but without a considerable decrease of unbonded fiber surface.

Mechanical fines perform differently due to the presence of both the collapsible fibrils and noncollapsible flakes. They increase both the interfiber bonding and the area for light scattering, thus tensile strength and opacity improve simultaneously. The effect of pigments, chemical fines and mechanical fines introduced into an assembly of fibers is shown schematically in Figure 1.
In order to demonstrate the contribution of fibrils and flakes separately, two model fines were used – microfibrillated cellulose (MFC) and microcrystalline cellulose (MCC). Their performance is observed by introducing them into long kraft and TMP fibers and compared with TMP fines classified into fractions of different specific surface (6). Fractions of high surface (HS) and low surface (LS) area were used. From the micrographs shown in Figure 2 it is apparent that the LS fraction of TMP fines contains mostly ray cells and intact pieces of fiber. The HS fraction contains fiber wall fragments and more of the fibrillated material.

In Figure 3 the tensile strength is shown as a function of fines content in handsheets formed from TMP long fibers and unbeaten long kraft fibers. The results are consistent with the assumption that fibrils contribute to bonding between fibers and consequently tensile strength, as shown in the case of MFC. On the other hand, the granular particles of MCC interfere with interfiber bonding and tensile strength decreases. The effect of HSF and LSF of TMP is between the two extremes. For comparison, the tensile strength of nonfractionated original TMP containing 0.35 g fines per gram of fibers is included as a single point.

In Figure 4 is shown the light scattering behavior of the handsheets. Again,
Figure 2  Micrographs: A – microcrystalline cellulose (MCC), B – high specific surface area fines (HSF), C - low specific surface area fines (LSF), D – microfibrillated cellulose (MFC). The bars are 10 μm.

Figure 3  Tensile strength as a function of different types of fines introduced into TMP fibers (left) and kraft fibers (right).
the trend is obvious. The MCC acts as a pigment by increasing the solid-air interface while the collapsible MFC affects the interface only marginally. The HSF contain small particles with considerable surface area and therefore their contribution to light scattering is high, and when incorporated into kraft fibers it even matches that of MCC. On the other hand, the particles in LSF are large and therefore their low surface contributes little to the total.

The contributions of small and large particles can be demonstrated by comparing the effect of two MCCs having an average particle size of 6 μm and 100 μm, and shown for TMP handsheets in Figure 4. The calculated specific surface area of cellulose spheres, 6 μm in diameter, is 15 times the area of spheres 100 μm in diameter. This means that the increase of total surface area by large MCC is insignificant in comparison to the smaller ones. The light scattering for original TMP containing 0.35 g fines per gram fiber is also shown. In general, the outstanding performance of HSF, particularly the ability to improve tensile strength and light scattering simultaneously, derives from the presence of both the collapsible fibrils and the granules small enough to possess a considerable surface.

By considering the performance of fines in kraft and TMP handsheets in Figure 4, one may conclude that although similar in trend, there is a difference. Without a more detailed analysis it is difficult to explain why the LSF

**Figure 4** Light scattering coefficient as a function of fines introduced into TMP fibers (left) and kraft fibers (right).
are more effective in kraft handsheets where the difference between LSF and MFC is obvious while in TMP handsheets it is not. Also, why the MFC in kraft handsheets has little effect on light scattering while in TMP handsheets the presence of MFC results in increased light scattering is not entirely clear. One can speculate that this is due to different characteristics of the fibers, particularly their flexibility, stiffness, conformability and other factors that may determine how the fines and their fractions may be accommodated in the fiber network.

The relation between tensile strength and light scattering is of practical significance if both properties are important. This relation allows, for example in filled papers, to select a pigment which for a given improvement in optical properties causes the least drop in tensile strength.

In Figure 5 is shown the tensile strength versus light scattering for handsheets from kraft and TMP fibers. In the kraft handsheets the MFC performs as expected by increasing tensile strength accompanied by a small decrease in light scattering. The MCC acts as a pigment – increases light scattering and decreases tensile strength. However, it is the different performance of HSF and LSF which is of interest. It indicates that the LSF introduced into kraft fibers are detrimental to tensile strength while the improvement in scattering is not great. Their introduction into TMP fibers results in rather small improvements in light scattering and tensile strength. On the other hand, the

![Figure 5](image_url)  
**Figure 5**  Tensile strength versus light scattering coefficient for TMP fibers (left) and kraft fibers (right) containing different fines.
HSF in kraft fibers are effective in light scattering but the tensile strength is not impaired. Their positive effect is even more pronounced in TMP handsheets where both the light scattering and tensile strength improve considerably. This observation naturally leads to the conclusion that it would be beneficial to separate fines from TMP pulp, remove the LSF fraction and then combine the remaining HSF fraction with the long fibers. The relation for original TMP shown as a single point is similar to that of HSF but it should be realized that the handsheet from unfractionated TMP contains 0.35 g fines per gram of fibers, while it is enough to add about 0.15 g HSF to one gram of long TMP fibers to reach similar values.

Because the HSF fraction acts also as filler by increasing the light scattering, it is of interest to compare its performance with that of a conventional filler – precipitated calcium carbonate (PCC). Figure 6 shows the tensile strength of handsheets from kraft and TMP fibers as a function of PCC and HSF. Figure 7 shows the light scattering coefficient for the same handsheets. Figure 8 shows the plot of tensile strength against light scattering which indicates that HSF are superior to PCC because with increasing light scattering there is no loss of tensile strength. However, it has to be realized that for obtaining the same value of light scattering, say 60 m²/kg, which is about the maximum reached with HSF in kraft handsheets it would be necessary to incorporate 0.6 g HSF as opposed to about 0.12 g PCC per gram fiber.

![Figure 6](image-url)  
Figure 6  Tensile strength as a function of HSF and PCC introduced into TMP fibers (left) and kraft fibers (right).
Figure 7  Light scattering coefficient as a function of HSF and PCC introduced into TMP fibers (left) and kraft fibers (right).

Figure 8  Tensile strength versus light scattering coefficient for TMP fibers (left) and kraft fibers (right) containing HSF and PCC.
The superior performance of HSF is even more pronounced in handsheets formed from TMP fibers. Besides the increased light scattering there is a considerable improvement in tensile strength.

Although the same applies as above, i.e., more HSF are required to reach a given value of light scattering, HSF improve tensile strength at the same time. However tempting it is to suggest that HSF can be used instead of mineral pigments, one drawback is rather serious and becomes obvious from considering the opacity as shown in Figure 9 and light scattering shown in Figure 7. Although the HSF are less effective in light scattering, the opacity is similar to that produced by PCC. This is due to the fact that besides scattering the light, the TMP fines also absorb the light. Therefore, the brightness of TMP handsheets containing fines does not improve contrary to handsheets filled with PCC, as shown in Figure 10. In the case of kraft fibers containing TMP fines, the brightness decreases considerably. In principle, this difficulty could be overcome in bleaching the HSF separately, but as a consequence the opacity will suffer.
CONCLUSIONS

Fines in the form of microfibrils are responsible for improving bonding between fibers. Fines in the form of microgranules improve light scattering because they introduce additional solid-air interface. In order to be effective they must be small and thus possess large surface areas.

TMP fines are a mixture of fibrillated and granular particles and thus contribute to improvement of both the tensile strength and the scattering of light. However, for optimum performance it is beneficial to separate the TMP fines into a fraction of high surface area, which contains mostly the microfibrils and small granules. This HSF fraction is superior to calcium carbonate filler because it can increase opacity to similar values, and simultaneously also improve tensile strength, however at the cost of reducing brightness in kraft papers.

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REFERENCES


Transcription of Discussion

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This paper nicely presents the great potential that the fines have in improving paper properties and also some of the dangers. If there are quality variations in the fines of mechanical pulp, can also cause them serious variations in paper properties. Unfortunately the fines fraction of mechanical pulp is the most expensive fraction because there is a lot of energy needed to produce it. Have you considered how to produce fines and separate fines in the mill scale.

Theo van de Ven

The answer is no. We’ve just taken fines and separated them from the whole TMP pulp, and used those fractions in our experiments. I fully agree that if you want to use fines as an additive, you need economic ways of producing them.

John Roberts  Department of Paper Science, UMIST

Did you use any retention aids in your handsheet preparations and if not, would you care to speculate, particularly what the effect of cationic retention aids might be on the state of aggregation of the fibrillar fines, as you refer to them and what impact they may have on bonding and strength as a result?

Theo van de Ven

Well that is a very good question. The only retention aids I talked about here was when we introduced the fillers into the sheets. With the fines, we have not used retention aids in this study, and so we had to determine what the fraction was in the handsheets because when we made them, you don’t have 100%
fines retention. Obviously retention aids will affect how much fines are retained in the first pass retention but also other effects such as the state of flocculation of the fines. My guess is that this will not affect the correlation between the optical properties and the surface area of the larger pores, but it might have a different effect on the strengths because if you have fines in aggregates, they might bind in a different way with fibres than as individual fines. It would be very good additional research topic to look into.

**Hannu Paulapuro** Helsinki University of Technology

How did you actually fractionate these two fines into these two categories and how did you check the success of the fractionation?

**Theo van de Ven**

This was done in Paprican in Pointe Claire. They use a float washer I believe to separate the fines and we just took them as separated by my colleagues in Pointe Claire. We did look at them by electron micrograph to see what the material looked like.

**Hannu Paulapuro**

But you didn’t check how much fibrillar type of fines did you had in one fraction or how much chunky type of fines you had in the other type of fraction – quantitatively?

**Theo van de Ven**

I don’t understand your question. We just measured the total amount of fines and from optical evidence, we know roughly how much of it is chunky and how much is fibrillar, but that is very difficult to quantify, so we have not done that.

**Hannu Paulapuro**

There are methods to quantify, for example, if you look at Kari Luuko’s thesis.

**Kari Ebeling** UPM-Kymmene Corporation

The problem in the real papermaking is that with tight water circulation the
fines circulate a long time and collect all kinds of materials on the surfaces. For handsheets made in laboratory conditions you have a certain co-operation between the strength contribution and optical properties contribution of the fines, but when fines collect the colloidal material, the resins and all other dirt from the white water, they contribute a little bit differently on the relationship between strength and opacity.

But could you speculate on an idea if it were possible to fix, with chemical reactions, fibrils on the surface of the parent fibre so that you would not have fines at all, but you would have these fibrils that won’t collapse due to the surface tension forces, and thus take care of the optical properties and they you could have the chemical pulp like hydrophilic fibrils contribute to the strength. Is that something that the chemists can do because then we would not have any retention problem?

Theo van de Ven

What you could do is, using the microfibrillated cellulose, which is commercially available, as an additive and you know you only have fibrils which are reasonably well defined, but that is a very expensive proposition for papermaking because that material is quite expensive. I don’t know of a simple way of only getting the fibrillar fines and leaving out the other material that has more negative properties. The study we did was more to show how important the various fractions are rather than trying to optimise the strength properties or the optical properties by even further fractionating the fines.

Kari Ebeling

Just a comment, from the papermakers viewpoint, you should attach the fines to the parent fibre, don’t make separate fine – you generate fines that follow the parent fibre.

Theo van de Ven

I agree.