Preferred citation: J. Phipps. Invited Perspective: Current and potential use of highly fibrillated cellulose in the paper and board industry. In *Advances in Pulp and Paper Research*, Cambridge 2022, Trans. of the XVIIth Fund. Res. Symp. Cambridge, 2022 (D. Coffin and W. Batchelor, eds), pp 249–268. FRC, Manchester, 2022. DOI: 10.15376/frc.2022.1.249.

CURRENT AND POTENTIAL USE OF HIGHLY FIBRILLATED CELLULOSE IN THE PAPER AND BOARD INDUSTRY

Jonathan Phipps

FiberLean Technologies Ltd., Par Moor Centre, Cornwall, PL24 2SQ, UK

ABSTRACT

The use of microfibrillated cellulose (MFC) in the paper industry has become established following many years of development by both academic and industrial researchers. Commercial installations typically use mechanical disintegration techniques such as refiners and grinders to convert aqueous suspensions of pulp fibres into a material consisting of fibrils and fibre fragments with diameters ranging from the nanometre to the micron scale. MFC suspensions of a few percent solids content show very high viscosity at low shear rates, but also very significant shear thinning behaviour, rapid viscosity recovery after shear and high filtration resistance. MFC added to paper furnishes at up to 5% by weight functions as a strength additive, enabling increases in mineral filler content, improvements in paper properties, reductions in weight and cost savings across a wide range of paper and board grades. As a complementary technology to pulp refining, addition of MFC offers process flexibility as well as improved wet web strength and runnability, reduced air permeability and increased z-direction strength. Although the fine fibrils of MFC do not dewater easily on their own, when added at low levels to paper their effect on machine drainage can be managed without loss of paper machine speed. In recent years, MFC has attracted much interest as a coating material. Layers or films of pure MFC show near-zero air permeability, high resistance to oil and grease and an effective barrier to organic vapours and oxygen. Mixtures of mineral particles and as little as 15% MFC provide an effective surface for water-intensive printing techniques such as flexography and inkjet. Application of MFC suspensions after the wet line of a papermachine has been demonstrated as a practical solution to obtain coatings, exploiting the rheological behaviour of the MFC to achieve excellent holdout onto a poorly-consolidated sheet, and using the vacuum and press sections of the machine to remove excess water. Further development and commercialisation of this technology, together with low cost MFC production and improved product characterisation, should ensure the continued growth of its use in the paper and board industry.

INTRODUCTION

Various forms of highly fibrillated cellulose, known as microfibrillated cellulose (MFC), cellulose nanofibrils (CNF), nanocellulose or cellulose filaments, have been the subject of intense research for more than a decade. All are produced from cellulose fibres, usually from wood, by separating the fibres into their constituent fibrils to various degrees. The first studies are generally attributed to Turbak [1] in the 1980s, who passed softwood sulphite pulp fibres multiple times through a high-pressure homogenizer to yield a viscous gel with suggested uses in food, cosmetics, paints and medical applications. The process was extremely energy intensive and prone to problems with blocking the homogeniser and did not receive much further interest until the early 2000s, when various researchers began experimenting with enzymatic or chemical pre-treatments to reduce the energy required [2,3,4]. Complete separation of fibres into uniform elementary fibrils of a few nanometres in diameter by selective oxidation of the fibril surface to make it highly anionic was demonstrated by Isogai in 2011 [5].

Whilst there are no formal classification systems for fibrillated cellulose, generally those made with a chemical pre-treatment which leads to near-complete separation of the elementary fibrils and formation of transparent gels are called nanocellulose, whereas those using principally mechanical disintegration methods, sometimes aided by enzymatic pre-treatment, are referred to as microfibrillated cellulose (MFC). Commercial mechanical treatments suitable for the paper industry use either modified pulp refiners [6,7] or stirred media mills [8,9] to separate the fibres into clusters of fibrils as well as individual fibrils, giving products covering a wide range of length, diameter and aspect ratio and highly branched structures. As well as being cheaper to produce than nanocellulose, MFC is more suited to paper applications, as it is coarse enough to be easy to retain in the papermaking process, and generally it is only weakly charged and thus does not interfere with wet end additives.

CHARACTERISATION AND PROPERTIES OF MICROFIBRILLATED CELLULOSE

Figure 1 shows some optical microscope images of pulp and MFC at various levels of disintegration using a stirred media mill. The complexity of the materials is evident, with fibrils and fragments covering a large range of lengths and widths and many branched structures present. SEM images (Figure 2) show that fibrils exist at sub-optical resolution down to a few nanometres in diameter. Figure 2 also contrasts the mechanically-produced MFC with a nanocellulose obtained by TEMPO oxidation of pulp prior to disintegration in a homogeniser, where the fibrils are much finer and almost entirely of uniform diameter.

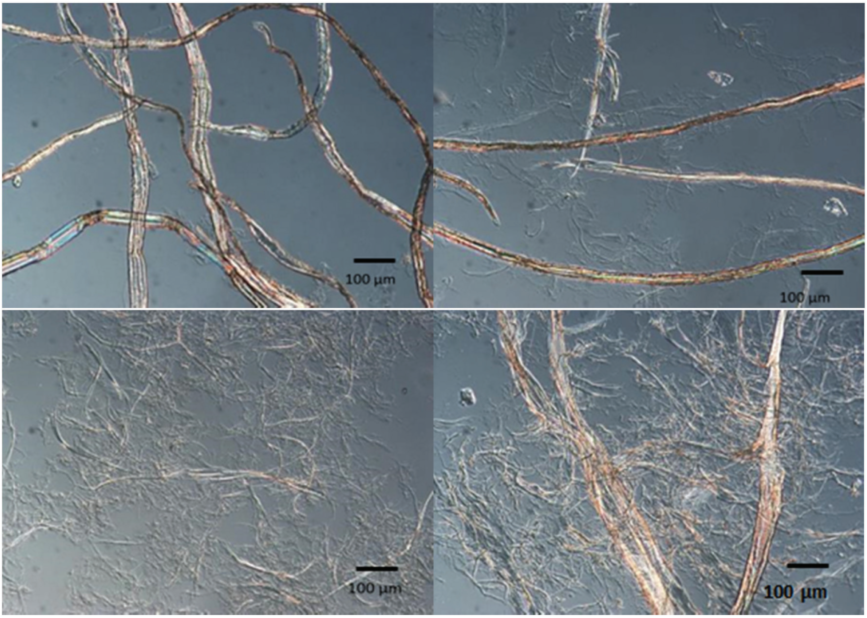

Figure 1. Differential Interference Contrast Microscope Images of Pulp and Microfibrillated Cellulose. Clockwise from top left: Unrefined softwood pulp, Highly-refined softwood pulp (65 CSF), Coarse MFC and Fine MFC.

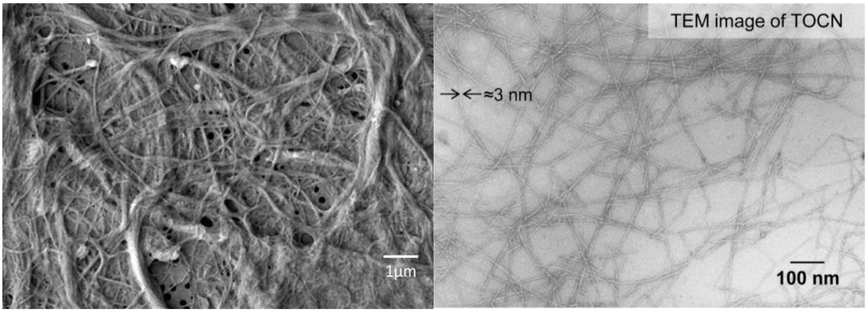

Figure 2. Scanning Electron Micrograph image of Fine MFC from Figure 1 (left). Transmission Electron Micrograph of TEMPO-oxidised nanocellulose (right, [5]).

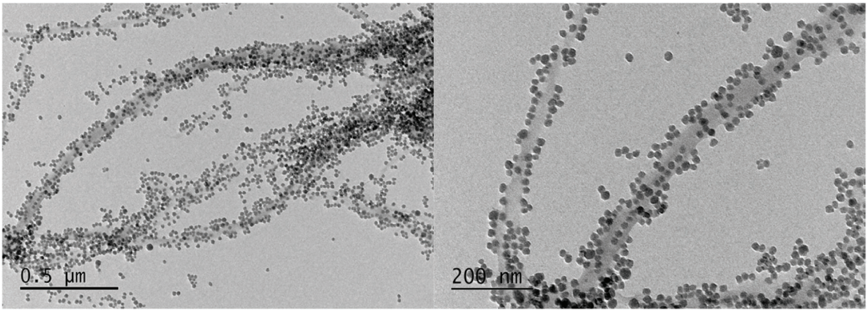

Figure 3. Transmission Electron Micrographs of cationic colloidal silica adsorbed to MFC [11].

As the disintegration process develops and finer fibrils are formed, one obvious change is the increase in the external surface area presented. However, characterisation of wet surface area by adsorption of dyes or polymers is problematic due to the internal porosity of the original fibres, which can register high apparent surface area which does not change substantially upon disintegration into MFC [10]. One way to avoid this problem is to use adsorbing particles that are too large to enter pores in the fibres and yet small enough to cover the external surface area of the fibrils [11]. Adsorption of cationic silica (Figure 3) has been shown to correlate well with the extent of fibrillation and development of other properties.

Despite the complex nature of MFC, conventional instruments for pulp analysis which use optical microscopy and rapid image analysis are frequently used to classify MFC and can be very useful [12]. These give distributions of length and width for the visible fibres and fragments, as well as other parameters such as the proportion of fines, defined as fibres shorter than 200µm, high aspect ratio fines, defined as fibres longer than 200µm but with a diameter of less than 10µm, and fibrillation, which is an estimate of the sharpness of the perimeter of fibres in the microscope images. Degree of conversion of pulp to MFC is often expressed in terms of the percentage of fines [13], but other parameters can correlate more closely with the properties of the material in its intended use.

Figure 4 contrasts the weight-averaged fibre length, the % of fines and high aspect ratio fines of highly refined softwood pulp with MFC processed by two different grinding strategies. Note that whilst both MFC products have similar weight-averaged mean lengths (Lc(w) ISO) and % of fines, they differ greatly in the % of high aspect ratio fines. Figure 5 shows correlations between the high aspect ratio fines and the tensile index of sheets made from 80% filler grade calcium carbonate and 20% MFC, as well as the air permeability of sheets of the pure MFC [14]. Both of these properties are strongly enhanced by a high proportion of long, thin fibrils.

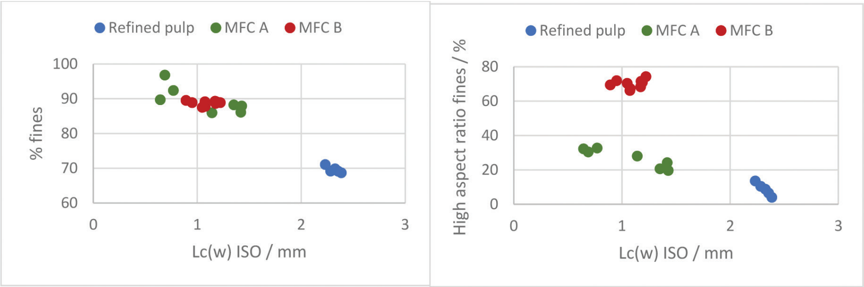

Figure 4. Characterisation of refined softwood (250–65 CSF) pulps and MFC made by different routes by Valmet Fiber Analyser – as % fines (left) and high aspect ratio fines (right).

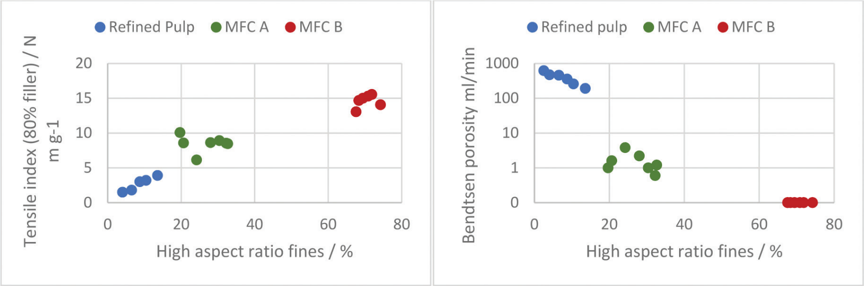

Figure 5. Correlation between % high aspect ratio fines and tensile strength of 80% $CaCO_3$20% MFC sheets (left) and Bendtsen porosity of 10gsm sheets of 100% MFC (right).

Characterisation of the smallest fibrils which are not amendable to analysis by optical microscopy presents further difficulties. Direct analysis of fibril width distributions from SEM images has been carried out by several researchers [15,16,17], but is highly time-consuming and unsuitable for routine analysis. Indirect methods such as measurement of gelation point [15], Water Retention Value [18], and suspension rheology [19] are more commonly used as indicators of the degree of ultrafine fibrillation in MFC.

The wet surface area to volume ratio and high aspect ratio of MFC leads to entanglements at low volume fractions (<2%), which make MFC suspensions very viscous at low shear rates. Yield stress and low shear viscosity rise very sharply with volume fraction, and suspensions are extremely shear-thinning [20,21]. Figure 6 shows the viscosity of a 1.5wt% suspension of MFC measured with a rotational rheometer and calculated from pressure drop vs flow data in the laminar region in a cylindrical pipe and a rectangular slot. Although the shear-thinning behaviour is often attributed to alignment of fibrils with flow [22], imaging studies have suggested that the fibrils remain in flocs even at high shear, and that shear-thinning is related to the size of the flocs and the interactions between them [23]. Rearrangements of floc structure are also believed to be responsible for the deviations in viscosity/shear rate relationships often observed in the region of 10s^{-1}. Mechanically produced MFC used in the paper industry is

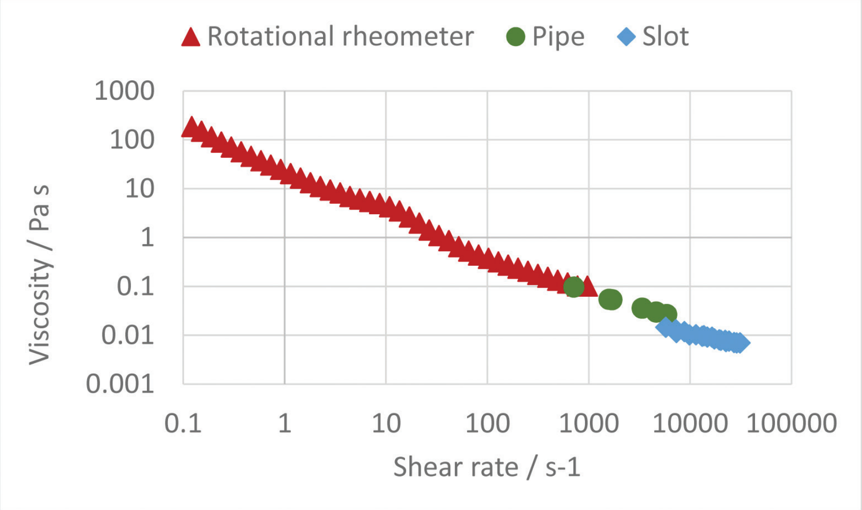

Figure 6. Viscosity of a 1.5% MFC suspension measured with rotational rheometer and calculated from pressure drop with flow in cylindrical pipe and rectangular slot [14].

only weakly charged, and its suspension rheology is thus quite insensitive to salt concentration and pH effects. Because the main source of viscosity is physical entanglements, dispersants also have only a limited effect [22]. MFC suspensions typically fit approximately to power law models of viscosity against shear rate, with flow indices of around 0.25 in the solids range of 0.5–2% [24]. Suspensions that are just sufficiently fluid to be pumpable that may have viscosity around 100 Pa s at 0.1 s^{-1} will approach the viscosity of water at the very high shear rates ($\sim 10^5\ s^{-1}$) typically seen in coating processes [25]. However, viscosity recovery after shear is extremely rapid.

EFFECT OF MFC ON PAPER PROPERTIES

The high surface area and aspect ratio of MFC, together with the flexibility of the fibrils, leads to strong bonding with itself and with pulp fibres when added to paper or formed into sheets of pure MFC [26,27,28]. Studies show that, at low doses of MFC in paper, bonded area increases proportionately to the amount of MFC added. Figure 8 shows the calculated bonded area as a function of MFC addition for handsheets made with bleached Kraft pulp at low and high $CaCO_3$ filler contents [29]. Sheets made entirely from MFC or nanocellulose (often referred to as ‘nanopaper’) show almost zero air permeability and very low internal void space and can have densities approaching that of pure cellulose [30,31]. As a consequence, sheets or layers of MFC show high oil and grease resistance and very low permeability to organic vapours and even oxygen [30,32,33]. However, despite the lack of open pores, MFC sheets have very poor water vapour barrier properties; because the fibrils remain hydrophilic even when dried, sorption of water into the fibril network and transport through it can still occur, especially at relative humidity levels above 60% [33].

Where used commercially, MFC is added in small amounts to the paper furnish, typically at the mixing chest. Its main effects on paper properties are to increase tensile, burst and z-direction strength [34]. Its effect on z-direction strength, also measured as Scott or Internal bond, is the largest, as this is particularly sensitive to bonded area and bond strength [35]. Also significant is the positive effect of MFC on modulus or tensile stiffness index, which helps to maintain sheet bending stiffness despite the loss of bulk and densification of the sheet that occurs as a result of the increased bonded area. Perhaps surprisingly, the increased bonding caused by MFC addition leads to an increase in elongation at break, and this in combination with higher modulus can give rise to significantly higher tensile energy absorption (TEA). At very high levels of MFC addition (beyond those normally achieved in paper), elongation at break may increase several fold.

The addition of MFC to paper also has a dramatic effect on its air permeability, as fibrils fill in the voids between fibres as well as bonding them together. A consequence of this is that surface smoothness and formation may also be improved. However, in the presence of high loadings of mineral fillers, this does not lead to a loss in light scattering, as the filler particles prevent complete collapse of the fibril structure, resulting in a net increase in solid/air interface [29,36]. This is shown in Figure 7, where the scattering coefficient is shown as a function of tensile strength for a range of filler loadings, refining levels and MFC addition. Here, light scattering increases with strength for filler loadings of 20% and above.

The high surface area and flexibility of MFC that leads to strong bonding and low porosity in dry sheets inevitably also leads to high filtration resistance and thus slow drainage compared with conventional refined pulp. The more fibrillated the material, the stronger the effect; drainage measurements can provide a quick evaluation of the relative degree of fibrillation of MFC products and thus can be a useful tool in process control. When MFC is added at low levels (typically <5% by weight) to paper, this is manageable with adjustments to machine settings and an increase in the dose of retention/drainage aids. The formation of a network of fibrils and the entanglement of fines and filler particles can lead to significant

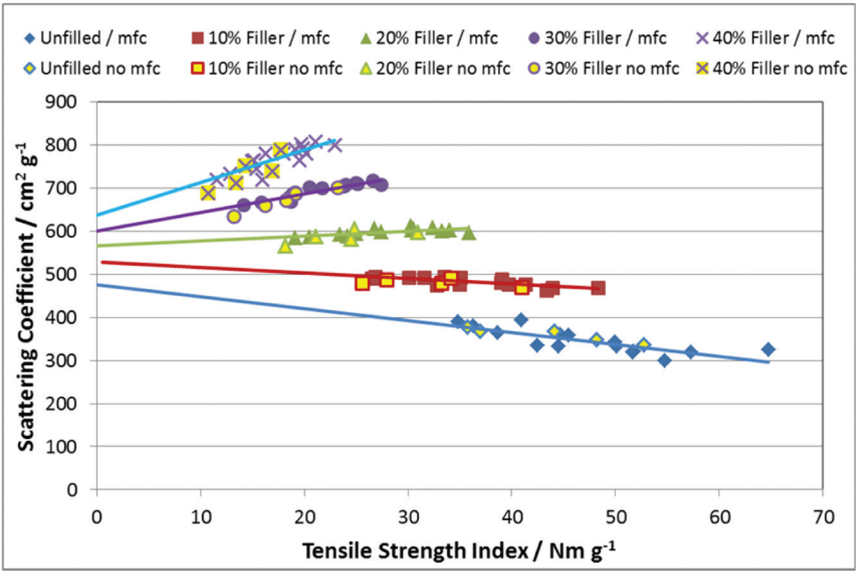

Figure 7. Light scattering vs tensile index for handsheets made with bleached Kraft pulp and $CaCO_3$ filler at different levels of refining and MFC addition [29].

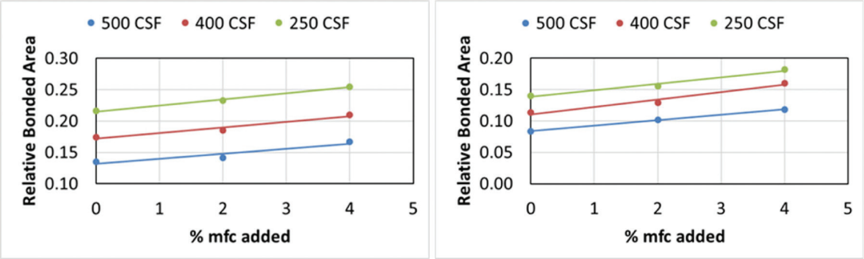

Figure 8. Calculated relative bonded area as a function of MFC addition for filled fine paper. 10% filler (left), 40% filler (right) [29].

improvements in retention. Strength improvements from starch and MFC are generally additive, and indeed the effect of MFC on retention and formation can allow higher doses of starch to be used [37].

One of the most important effects of MFC addition at the papermachine wet end is its positive influence on wet web strength [38]. Given that a paper web contains too much water before entering the dryer section for hydrogen bonding to occur, the mechanism for this is believed to be entanglement friction between the fibres, which MFC increases [39]. Since wet web strength is often the limiting factor for filler content or reductions in basis weight, addition of MFC can enable either of these.

COMPARISON OF MFC ADDITION WITH PULP REFINING

Conventionally, papermakers refine their pulp in order to increase fibre flexibility and to generate fibrils on the external surfaces of the fibres, both of which increase the relative bonded area in the sheet and thus the strength of the paper. In some cases, it may be desirable to shorten the longer fibres in order to increase the uniformity of fibre length within the furnish and improve formation, but generally shortening of fibres is avoided where possible. Addition of MFC can be viewed as an alternative approach – instead of subjecting the whole furnish to limited fibrillation, a small portion of it is converted completely to fibrils and then added back to the furnish. There are a number of potential benefits to this. Firstly, the creation of fibrils is decoupled from increasing fibre flexibility, and thus fibrillation can be pursued separately without the risk of losing fibre length and bulk. In some cases, this can enable tensile strength to be developed with less loss of tear strength that usually occurs with refining [29]. Secondly, studies have shown that MFC addition has a more profound effect on wet web strength than on dry strength, so that, for a given dry tensile strength, adding MFC will lead to a

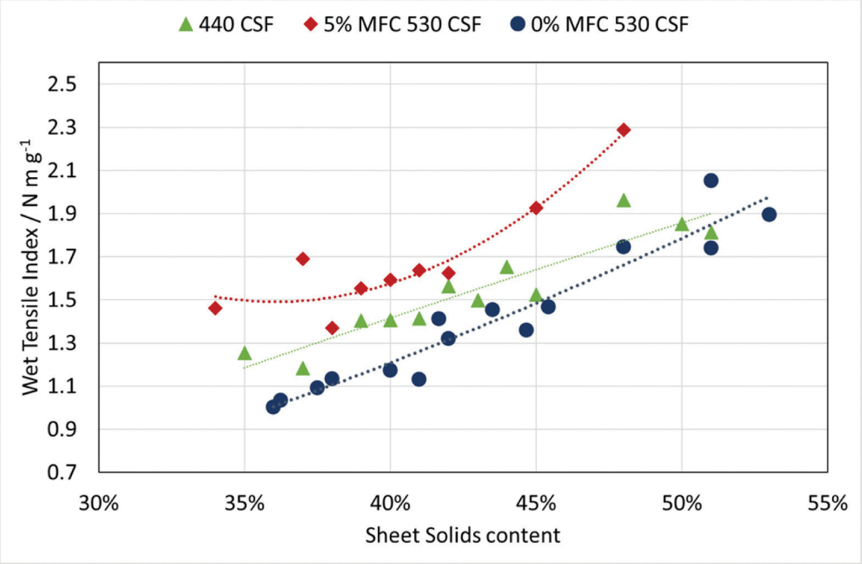

Figure 9. Wet tensile strength after pressing for handsheets made with 20% filler grade calcium carbonate from pulp refined to 530 CSF, from 530 CSF pulp with 5% added MFC, and from pulp refined to 440 CSF. Sheets from 550 CSF pulp with 5% MFC have the same dry tensile strength as sheets from 440 CSF pulp.

stronger web than refining. Figure 9 shows some laboratory data which demonstrates this for handsheets containing 20% filler grade calcium carbonate [38]. MFC has also been shown to have a greater effect on strength after rewetting compared with refining to the equivalent dry strength [40].

In paper machine trials, it has also been observed that the addition of MFC results in less lateral shrinkage of the web and better dimensional stability than refining to equivalent dry tensile index. As shown in Figure 8, in the presence of high filler loadings, the increase in relative bonded area obtained by addition of MFC is comparable to that obtained at lower filler content, whereas the effect of refining on bonded area diminishes with filler content. The mechanism for this is unclear, but SEM images showing how MFC fibrils can wrap around filler particles (Figure 10) suggest that its addition may allow the formation of filler/fibre bonds and thus reduce the loss of bonded area normally associated with fillers [29,41].

MFC can also be made effectively from recycled fibres [42], which often do not respond well to conventional refining, and thus enables strength enhancement of recycled furnishes that would not otherwise be possible. Finally, the ability to adjust MFC addition level rapidly in response to grade changes or variations in pulp feed offers the papermaker increased operational flexibility.

Figure 10. Scanning electron micrograph of co-processed MFC and precipitated calcium carbonate (PCC).

TYPICAL USES OF MFC IN PAPER AND BOARD GRADES

MFC is frequently used to enable an increase in mineral filler content, which may be limited by its effect on either wet web strength (runnability), z-direction strength or occasionally dry tensile or burst strength. In graphic paper grades made to a fixed grammage specification, using MFC to achieve the replacement of pulp with filler can save costs, provided that the cost of the MFC required is not too high [43]. Increasing filler content will lead to improved light scattering and thus opacity; further filler increase can be achieved by switching to a coarser filler which is less efficient at scattering but also less detrimental to strength, whilst still maintaining the original strength and opacity. Alternatively, the increase in strength and runnability generated by the use of MFC can be used to reduce basis weight in cases where this is desirable. The impact of MFC addition on wet end drainage is usually offset by the reduction in fibre content achieved, and with increases in filler content it is common to see press solids increase despite the high water retention capacity of the added MFC. Several researchers have also observed increases in press solids on pilot machines without filler increase [35,44]. However, the reduction in sheet bulk that occurs both from the increased bonding of the MFC and the increase in mineral filler content can be a problem. This can be addressed in several ways – for example by exploiting the effect of MFC on smoothness and formation to allow a reduction in calendering pressure, or by the use of an aggregated, bulking filler such as a coarse grade scalenohedral PCC. Again, any loss in light scattering caused by selection of a coarser filler grade can be compensated by the higher filler loading.

The reduction in sheet porosity due to the addition of MFC is generally beneficial for printing properties, and can also result in improved holdout of coatings [45], allowing either a reduction in coat weight or improvements in gloss or other coated paper properties. In principle, improved holdout should also be beneficial in the case of starch applied at the size press for sheet stiffness, though this may sometimes be offset by a reduction in pick-up.

In multilayer packaging grades such as white top linerboard or folding boxboard, MFC can also be used to increase filler content in the white outer layers, which is often limited by z-direction strength or delamination resistance. This in turn can allow a reduction in the grammage of the layer, since its main function is to cover the darker, unbleached layers below [46]. Although outer layers typically have very low basis weight, wet web strength is not a limiting factor because they are supported by the papermachine wire before being brought in contact with the heavier base layers and pressed together, and therefore the effect of MFC on it is of little consequence. However, the positive influence of MFC on filler retention may also be an enabler for increased filler loading. For linerboard, bulk is not a critical property, so any reductions that occur as a result of MFC or increased filler are not limiting. Liner property specifications include burst and short span compression (SCT) strength, both of which are reduced by filler but enhanced by MFC. Tensile stiffness of liners is important for the rigidity of the final corrugated product, and here the effect of increased filler can be offset both by the effect of the MFC and by the increase in base layer grammage where the latter is made from unbleached virgin Kraft pulp or a recycled grade that has higher tensile stiffness than the pulp used for the white layer.

MFC can be used in a similar way in folding boxboard grades [47], although here the effect of reducing outer layer grammage and increasing filler content is more critical, since the bending stiffness of the board is dependent on the I-beam effect of the stiff outer layers separated by bulky middle layers of mechanical pulp. Maintaining constant overall grammage allows the middle layer thickness to be increased, which can compensate for losses in outer layer tensile stiffness. However, in some cases it may be necessary also to use the effect of MFC on retention and formation to allow an increase in outer layer starch dosage in order to maintain the overall bending stiffness of the product.

In unfilled grades, MFC can be used as a complement to refining simply to improve mechanical properties, or to reduce the need for long fibre where it is required for wet web or tear strength. By reducing refining energy and adding MFC to maintain strength or delamination resistance, it may be possible to obtain higher bulk, for example in the middle layers of folding boxboard where CTMP is used and refined as lightly as possible [48].

COATING OF MFC ONTO PAPER AND BOARD

Addition of MFC at the wet end is normally restricted to low doses of 5% or less; at these levels it is possible to manage drainage but beyond this it would become necessary to reduce machine speed which is generally unacceptable. Furthermore, use of higher doses in the whole furnish is likely to lead to excessive costs. However, the properties of sheets made with much higher proportions of MFC, mixtures of MFC and mineral fillers or 100% MFC are attractive for a number of potential applications, and so methods for applying thin layers of MFC onto paper have been explored by several researchers.

Coating of MFC by conventional methods is inherently difficult due to its high water-holding capacity and viscosity even at low solids content. MFC suspensions are not sufficiently fluid to be pumpable above 5 wt% solids, so any coating directly onto dry paper would require orders of magnitude more drying energy than conventional coatings, most of which are applied at above 50 wt%. Despite the high affinity of MFC for water, contact with a porous paper substrate leads to very rapid dewatering and viscosity increase, and so the addition of water-retention agents such as carboxymethyl cellulose is required in order to be able to prevent break-up of the coating. Nevertheless, the coating of MFC by blade [49], rod [50] and slot die [51,52] applicators has been demonstrated at laboratory scale, although the coat weight achievable in a single pass is limited to around 5 gsm. If sufficient coverage can be achieved to form a coherent, defect-free layer, then very low permeability and good barrier properties to oil and grease can be achieved. Mechanical properties, in particular bending stiffness, may also be improved.

One potential solution to coating with MFC is to apply it at the wet end of a papermachine, and to use the vacuum and press sections of the machine to remove excess water [53,54,55]. Here the unique attributes of the material are beneficial; its shear-thinning nature makes it possible to apply it through a slot at high speed without generating excessive pressure, its fine size allows it to be applied at much higher than headbox solids through narrow openings without blocking them, and its rapid viscosity recovery prevents it from penetrating into even a very poorly consolidated base paper. An MFC suspension is applied as a jet or curtain just after the wet line; despite the high filtration resistance of MFC the process has been demon-

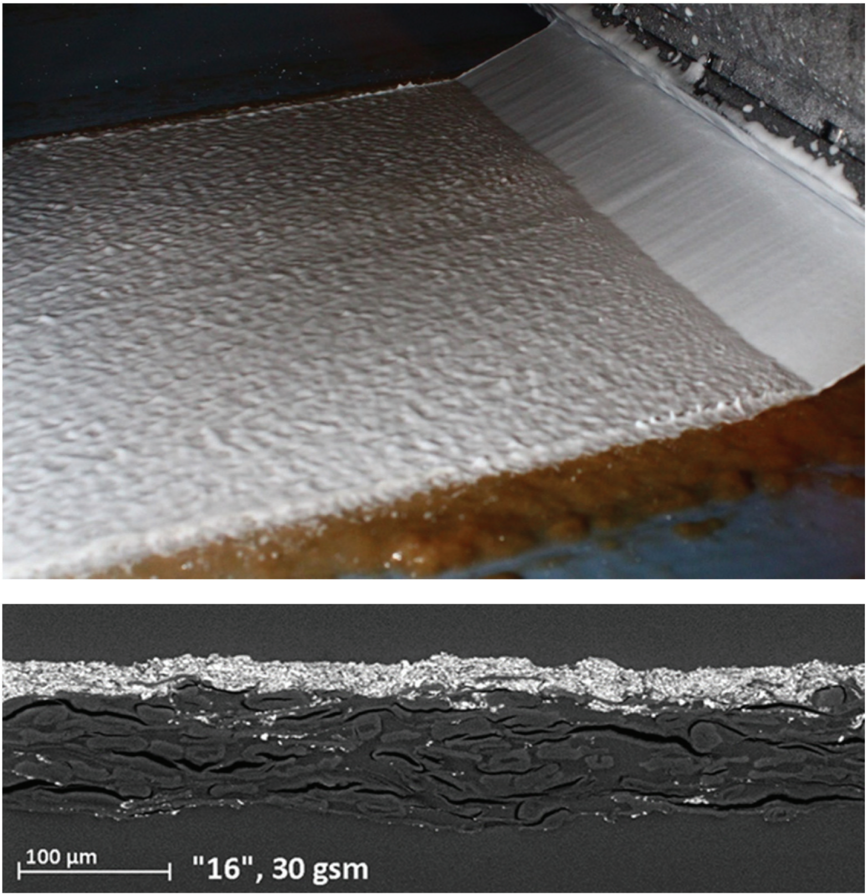

Figure 11. Short exposure photograph of MFC/mineral composite coating at pilot machine wet end at 500 m/min (top). SEM cross section of the coated sheet (bottom)

strated at machine speeds of up to 500 m/min [56,57]. A very simple applicator is required, and with no need to install extra drying equipment, application of MFC at the wet end offers a potentially low cost alternative to conventional coating onto dry paper. As well as providing an effective oil, grease or oxygen barrier, coatings of MFC are also an excellent surface for subsequent application of moisture barrier or release coatings. Mixtures of MFC and mineral fillers may also be applied in this way, potentially enabling the production of grades such as white top liner which conventionally require a machine with multiple forming sections. Here the strong binding properties of MFC allow a layer of up to 85% mineral to be applied with sufficient strength for printing and converting. The high mineral content of such coatings make them excellent surfaces for water-intensive printing methods such as inkjet or flexography [56]. Figure 11 shows an ultra-short exposure photograph of an 80/20 $CaCO_3$MFC coating being applied onto an unbleached base on a pilot machine at 500m/min – a 'contour' coating is visible on the unconsolidated surface. The SEM cross section of the dried paper (Figure 11, bottom) shows only minor penetration of the mineral component into the base sheet, and very even coverage of the surface.

FUTURE CHALLENGES FOR THE USE OF MFC IN PAPER AND BOARD

The use of MFC in paper and board is now well established, with several paper mills using it in wet end applications from on-site production facilities. Manufacture of MFC is energy-intensive; the main challenge for producers is to minimize energy consumption and costs in order to enable its use to be more widespread. Whilst the use of enzymes and other pre-treatments can reduce the amount of energy required to disintegrate pulp into fibrils, they incur extra process steps and chemical costs, so purely mechanical methods using grinders or refiners remain competitive. On-site production is preferred because it eliminates the need to dewater and transport the material, but in many cases the local demand for MFC is not sufficient to justify a production plant. The unique ability of MFC to hold water and bond strongly and irreversibly to itself when dried makes the production of a dry or high solids content form that can be transported and then easily redispersed extremely difficult. However, if this can be done cost-effectively it will increase the potential for MFC use in the industry substantially.

The coating of MFC onto paper and board has perhaps the most potential to transform the industry by making new, more sustainable products and enabling paper-based packaging to continue to substitute plastics. Wet end application shows promise but is in its infancy, and current approaches are only applicable to Fourdrinier machines. Further developments in equipment for MFC coating will be needed to realise the full potential of this approach. Many of the positive attributes of MFC such as bonding ability, strength and barrier properties are associated with high aspect ratio, fine fibrils, but these also bring high viscosity at low solids content and very slow drainage. Developments in MFC processing, in combination with other additives, in order to enable coating at higher solids content whilst still providing useful properties, would clearly be valuable. Although modern microscopy and image analysis techniques have proven very useful in characterisation of MFC, rapid and quantitative analysis of the sub-optical fraction is still needed in order to optimise both its production and use.

CONCLUSIONS

The use of microfibrillated cellulose in the paper and board industry is now well established, and continues to attract interest from many producers. As a wet end additive, its primary function is to improve paper strength, both during manufacture and in the finished product. Its benefits have been demonstrated in a wide range of applications, but are particularly suited to grades that contain a high level of mineral fillers, allowing filler increase, quality enhancements and cost savings. Addition of MFC to a paper furnish is a complementary technology to pulp refining or the addition of chemical strength agents, offering process flexibility and some specific advantages, such as improved wet web and z-direction strength, reduced air permeability and surface roughness. Developments are under way to establish processes for coating it onto paper, where it shows the potential to provide a low cost alternative to multilayer sheet forming as well as a coating or substrate for packaging applications where barrier properties are needed to meet the increasing demand for biobased and biodegradable alternatives to plastic and petroleum-based additives. These, together with continued improvements in production and characterisation technology, should ensure the continued expansion of MFC use in paper and board production in the years ahead.

REFERENCES

[1] A.F. Turbak, F.W. Snyder and K.R. Sandberg Microfibrillated cellulose, a new cellulose product: properties, uses, and commercial potential. *J Appl Polym Sci Appl Polym Symp*, 37 (9): 815–827, 1983

[2] M. Henriksson, G. Henriksson, L.A. Berglund and T. Lindström. An environmentally friendly method for enzyme-assisted preparation of microfibrillated cellulose (MFC) nanofibers. *European Polymer Journal* 43 (8): 3434–3441, 2007

[3] M. Pääkkö, M. Ankerfors, H. Kosonen, A. Nykänen, S. Ahola, M. Österberg, J. Ruokolainen, J. Laine, P.T. Larsson, O. Ikkala and T. Lindström. Enzymatic hydrolysis combined with mechanical shearing and high-pressure homogenization for nanoscale cellulose fibrils and strong gels. *Biomacromolecules* 8(6): 1934–1941, 2007

[4] L. Wågberg, G. Decher, M. Norgren, T. Lindström, M. Ankerfors and K. Axnäs. The build-up of polyelectrolyte multilayers of microfibrillated cellulose and cationic polyelectrolytes. *Langmuir*, 24(3): 784–795, 2008

[5] A. Isogai, T. Saito and H. Fukuzumi. TEMPO-oxidized cellulose nanofibers. *Nanoscale* 3(1): 71–85, 2011

[6] M. Suzuki and Y. Hattori. *U.S. Patent*. 7,381,294 B2, 2008

[7] K. Schelling, M.A. Bilodeau and M. Gerrer. Installation and start up of a commercial cellulose nanofibril production plant. *TAPPI International Conference on Nanotechnology for Renewable Materials*, Atlanta, GA, 2015

[8] H. Liimatainen, J. Sirviö, A. Haapala, O. Hormi and J. Niinimäki. Characterization of highly accessible cellulose microfibers generated by wet stirred media milling. *Carbohydrate Polymers* 83 (4): 2005–2010, 2011

[9] J. Husband, P. Svending, D. Skuse, T. Motsi, M. Likitalo and A. Coles. *U.S. Patent* 8231764 B2, 2012

[10] H. Ougiya, N. Hioki, K. Watanabe, Y. Morinaga, F. Yoshinaga and M. Samejima. Relationship between the physical properties and surface area of cellulose derived from adsorbates of various molecular sizes. *Bioscience, Biotechnology, and Biochemistry*, 62 (10): 1880–1884, 1998

[11] D. O'Neill. "Using the Light Scattering of Colloidal Silica to Measure the Surface Area of Micro-Fibrillated Cellulose Fibres", University of Bristol undergraduate thesis, 2019

[12] L. Taylor, J. Phipps, S. Blackburn, R. Greenwood and D. Skuse. Using fibre property measurements to predict the tensile index of microfibrillated cellulose nanopaper. *Cellulose* 27 (11): 6149–6162, 2020

[13] D. Cowles. Production of Microfibrillated Cellulose and its use in Specialty Papers, *TAPPI International Conference on Nanotechnology for Renewable Materials*, Montreal, Canada, 2017

[14] J.S. Phipps. The unique properties of Microfibrillated Cellulose and their exploitation in paper and paperboard, *TAPPI International Conference on Nanotechnology for Renewable Materials*, Helsinki, Finland, 2022

[15] S. Varanasi, R. He and W. Batchelor. Estimation of cellulose nanofibre aspect ratio from measurements of fibre suspension gel point. *Cellulose* 20 (4): 1885–1896, 2013

[16] S. Ang, V. Haritos and W. Batchelor. Effect of refining and homogenization on nanocellulose fiber development, sheet strength and energy consumption. *Cellulose* 26 (8): 4767–4786, 2019

[17] D. Hewson, J. Phipps, J.A. Shatkin and D.Skuse. Commercialising MFC products: Compliance to Ethical Standards and Legislation, *TAPPI International Conference on Nanotechnology for Renewable Materials*, online, 2021

[18] F. Gu, W. Wang, Z. Cai, F. Xue, Y. Jin and J.Y. Zhu. Water retention value for characterizing fibrillation degree of cellulosic fibers at micro and nanometer scales. *Cellulose* 25 (5): 2861–2871, 2018

[19] G. Cinar Ciftci, P.A. Larsson, A.V. Riazanova, H.H. Øvrebø, L. Wågberg and L.A. Berglund. Tailoring of rheological properties and structural polydispersity effects in microfibrillated cellulose suspensions. *Cellulose* 27 (16): 9227–9241, 2020

[20] M. Iotti, Ø.W. Gregersen, S. Moe and M. Lenes. Rheological studies of microfibrillar cellulose water dispersions. *Journal of Polymers and the Environment* 19 (1): 137–145, 2011

[21] M. Schenker, J. Schoelkopf, P. Gane and P. Mangin. Rheology of microfibrillated cellulose (MFC) suspensions: influence of the degree of fibrillation and residual fibre content on flow and viscoelastic properties. *Cellulose* 26 (2): 845–860, 2019

[22] M. Schenker, J. Schoelkopf, P. Gane and P. Mangin. Rheological investigation of complex micro and nanofibrillated cellulose (MNFC) suspensions: Discussion of flow curves and gel stability. *Tappi Journal* 15 (6): 405–416, 2016

[23] E.G. Facchine, R.J. Spontak, O.J. Rojas and S.A. Khan. Shear-dependent structures of flocculated micro/nanofibrillated cellulose (MNFC) in aqueous suspensions. *Biomacromolecules* 21 (9): 3561–3570, 2020

[24] T. Turpeinen, A. Jäsberg, S. Haavisto, J. Liukkonen, J. Salmela and A.I. Koponen. Pipe rheology of microfibrillated cellulose suspensions. *Cellulose* 27 (1): 141–156, 2020

[25] V. Kumar, B. Nazari, D. Bousfield and M. Toivakka. Rheology of microfibrillated cellulose suspensions in pressure-driven flow. *Applied Rheology* 26 (4): 24–34, 2016

[26] Ø. Eriksen, K. Syverud and Ø. Gregersen. The use of microfibrillated cellulose produced from kraft pulp as strength enhancer in TMP paper. *Nordic Pulp & Paper Research Journal* 23 (3): 299–304, 2008

[27] F.W. Brodin, Ø.W. Gregersen and K. Syverud. Cellulose nanofibrils: Challenges and possibilities as a paper additive or coating material–A review. *Nordic Pulp & Paper Research Journal* 29 (1): 156–166, 2014

[28] T.B. Jele, P. Lekha and B. Sithole. Role of cellulose nanofibrils in improving the strength properties of paper: a review. *Cellulose* 29 (1): 55–81, 2022

[29] J. Phipps, T. Larson, D. Ingle and, H. Eaton. The Effect of Microfibrillated Cellulose on the Strength and Light Scattering of Highly Filled Papers. In *Advances in Pulp and Paper Research, Transactions of the 16th Fundamental Research Symposium*, 231–254, 2017

[30] C. Aulin, M. Gällstedt and T. Lindström. Oxygen and oil barrier properties of microfibrillated cellulose films and coatings. *Cellulose* 17 (3): 559–574, 2010

[31] M. Henriksson, L.A. Berglund, P. Isaksson, T. Lindström and T. Nishino. Cellulose nanopaper structures of high toughness. *Biomacromolecules* 9 (6): 1579–1585, 2008

[32] K. Syverud and P. Stenius. Strength and barrier properties of MFC films. *Cellulose* 16 (1): 75–85, 2009

[33] J. Wang, D.J. Gardner, N.M. Stark, D.W. Bousfield, M. Tajvidi and Z. Cai. Moisture and oxygen barrier properties of cellulose nanomaterial-based films. *ACS Sustainable Chemistry & Engineering* 6 (1): 49–70, 2018

[34] P. Svending and J. Phipps. Microfibrillated Cellulose – Mineral Composites for Paper and Paperboard Applications, *TAPPI PaperCon Conference*, Atlanta, GA, 2015

[35] D.A. Johnson, M.A. Paradis, M. Bilodeau, B. Crossley, M. Foulger and P. Gelinas. Effects of cellulosic nanofibrils on papermaking properties of fine papers. *Tappi Journal* 15 (6): 395–402, 2016

[36] R. Bown. The relationship between strength and light scattering coefficient for filled papers. In *Papermaking Raw Materials, Trans. 8th Fund. Res. Symp.* (ed. V. Punton), 543–576, 1985

[37] M. Ankerfors, T. Lindström and D. Söderberg. The use of microfibrillated cellulose in fine paper manufacturing – Results from a pilot scale papermaking trial. *Nordic Pulp & Paper Research Journal* 29 (3): 476–483, 2014

[38] J. Phipps, T. Larson, M. Paradis and D. Tanase. The effect of microfibrillated cellulose on the wet-web strength of paper. *Tappi Journal* 20 (1): 61–68, 2021

[39] M.H. de Oliveira, M. Maric and T.G. van de Ven. The role of fiber entanglement in the strength of wet papers. *Nordic Pulp & Paper Research Journal* 23 (4): 426–431, 2008

[40] J. Su, W.K. Mosse, S. Sharman, W.J. Batchelor and G. Garnier. Effect of tethered and free microfibrillated cellulose (MFC) on the properties of paper composites. *Cellulose* 20 (4): 1925–1935, 2013

[41] C. Hii, Ø.W. Gregersen, G. Chinga-Carrasco and Ø. Eriksen. The effect of MFC on the pressability and paper properties of TMP and GCC based sheets. *Nordic Pulp & Paper Research Journal* 27 (2), 388–396, 2012

[42] D. Skuse, J. Phipps and T. Larson. Co-ground mineral/microfibrillated cellulose composite materials: Recycled fibers, engineered minerals, and new product forms. *Tappi Journal* 20 (1): 49–58, 2021

[43] P. Svending, J. Phipps, R. Lai and A. Martoni. Substituting pulp for filler is increasingly attractive for papermakers *TAPPI PaperCon Conference*, Indianapolis, IN, 2019

[44] I. Kajanto and M. Kosonen. The potential use of micro- and nanofibrillated cellulose as a reinforcing element in paper. *Journal of Science & Technology for Forest Products and Processes* 2 (6): 42–48, 2012

[45] D.A. Johnson and M.A. Paradis. Enhancing Coating Holdout with Cellulosic Micro-Fibrils. *TAPPI International Conference on Nanotechnology for Renewable Materials*, Madison, WI, 2018

[46] J. Phipps, P. Svending, T. Selina, J. Kritzinger, T. Larson, D. Skuse and S. Ireland. Applications of Co-Processed Microfibrillated Cellulose and Mineral in Packaging. *TAPPI PaperCon Conference,* Minneapolis, MN, 2017

[47] J. Phipps and R. Hill. Benefits of microfibrillated cellulose in Paperboard *TAPPI Conference on Nanotechnology for Renewable Materials*, Chiba, Japan, 2019

[48] T. Larson. Developments in use of MFC for End Market Applications. *Specialty Papers US Conference*, Appleton, WI, USA, 2018

[49] S.M.M. Mousavi, E. Afra, M. Tajvidi, D.W. Bousfield and M. Dehghani-Firouzabadi. Application of cellulose nanofibril (CNF) as coating on paperboard at moderate solids content and high coating speed using blade coater. *Progress in Organic Coatings* 122: 207–218, 2018

[50] N. Lavoine, I. Desloges, B. Khelifi and J. Bras. Impact of different coating processes of microfibrillated cellulose on the mechanical and barrier properties of paper. *Journal of Materials Science* 49 (7): 2879–2893, 2014

[51] V. Ottesen, V. Kumar, M. Toivakka, G. Chinga Carrasco, K. Syverud and Ø.W. Gregersen. Viability and properties of roll-to-roll coating of cellulose nanofibrils on recycled paperboard. *Nordic Pulp & Paper Research Journal* 32 (2): 179–188, 2017

[52] R. Koppolu, T. Abitbol, V. Kumar, A.K. Jaiswal, A. Swerin and M. Toivakka. Continuous roll-to-roll coating of cellulose nanocrystals onto paperboard. *Cellulose,* 25 (10): 6055–6069, 2018

[53] J. Phipps, P. Svending, T. Selina, T. Larson and D. Skuse. Application of a coating of co-processed microfibrillated cellulose and mineral at the wet end. *TAPPI Advanced Coating Symposium*, Stockholm, Sweden, 2016

[54] D.A. Johnson and M.A. Paradis. Surface application of cellulose nanofibrils to fine paper using different base sheet freeness levels. *TAPPI PaperCon Conference*, Minneapolis, MN, 2017

[55] D. Bousfield, M. Paradis, D. Johnson and M. Bilodeau. Table drainage and press dewatering when cellulose nanofibers are applied on the wet end. *TAPPI PaperCon Conference*, Minneapolis, MN, 2017

[56] P. Svending. Corrugated Board surfaces suitable for water intensive printing. *TAPPI PaperCon Conference*, Charlotte, NC, 2018

[57] D. Cowles. Surface Application of Microfibillated Cellulose *TAPPI International Conference on Nanotechnology for Renewable Materials*, Madison, WI, 2018

CURRENT AND POTENTIAL USE OF HIGHLY FIBRILLATED CELLULOSE IN THE PAPER AND BOARD INDUSTRY

Jonathan Phipps

FiberLean Technologies Ltd, Par Moor Centre, Cornwall, PL24 2SQ, UK

Imanuel Kriesten Mercer Fibre Centre

I am speaking about refining, speaking about energy intake into a furnish, I find it hard to not stating anything about energy used to do it. So, you showed that you use almost 2,500 kilowatts per tonne of energy for making MFC, so adding 5% means adding quite a lot of energy into the stock. So, I would be interested if you compare the refined pulp with the same energy amount applied to fibres with your MFC.

Jon Phipps

I made the comparison between the addition of MFC and the refining of pulp in the early part of my presentation to address this question and to highlight the differences between them. Our experience is that almost no paper mill that we have approached has said that they could achieve the same results with refining that we see with MFC addition. This may be because they don't want to change their refining conditions for several reasons, and sometimes also because they have limited refining capacity. There are also many more considerations than the comparative energy consumed in making a few percent of MFC as an alternative to refining the whole pulp. I did show some published values for the amount of energy used in MFC production, but these don't necessarily reflect current industrial practice. They are illustrative primarily of studies that have been done in universities. However, minimising energy consumption is one of the challenges in making MFC, as I hope I highlighted in considering the future challenges and research directions for the industry. Clearly our aim is to use less energy to make the MFC than would be needed to refine the whole pulp to the same strength.

Alexander Bismarck University of Vienna

Just wondering, you put in a massive amount of filler and you calculate a grammage based on the amount of cellulose in your paper. You reduce the amount of cellulose but you add additional weight, and especially for packaging, should I not consider the overall density of my package material?

Jon Phipps

The grammage of the sheet includes all of the components. So, for example, an 80 gsm sheet at 20% filler has 16 gsm of filler and 64 gsm of fibre. The main reason why paper mills would like to increase filler content is because they define their grades by grammage. If they make and sell an 80 gsm product, then if they can increase the filler, which let us say costs $100/tonne, and reduce the fibre, which might cost closer to $700/tonne, then they will make a substantial saving overall.

Elias Retulainen Fiber and Fibril

In principle we would like to usually have a very homogeneous material when we make microfibrillated cellulose for different purposes. Like today we have heard about barrier properties. I think the bigger particles, which we can call ‘residual fibres’ may be a problem. So have you any idea of how to get rid of them, or fractionate out and overcome the problem?

Jon Phipps

Not surprisingly, we are working on eliminating the larger fibre fragments for many applications. In the optical micrographs that I showed, it’s clear that many of the fibrils remain attached to larger fibre fragments, which is probably useful in order to retain them in papermaking, for example. It’s much less useful if we consider coating with them, for example to make barrier layers. So, for those applications, reducing the larger particles is something that we aim to do. Fractionation, however, is much more difficult. So, the approach that we, and probably everybody else who makes MFC industrially, prefer to take is to produce the narrow size distribution that we want rather than make a wider distribution with components that we don’t want and then try to remove them.

Bill Sampson University of Manchester

Can you just clarify, you talked about refined pulp, but there are lots of different kinds of refiners and lots of different kinds of pulp, so what are those data for?

Jon Phipps

The pictures and data that I showed are for bleached softwood Kraft pulp refined in a Valley beater.

Bill Sampson

Okay, that is why you have not lost very much fibre length.

Jon Phipps

Yes, quite possibly. We would always like to be able to do our lab refining as realistically as possible, but the choices we have are limited – we could use a PFI mill, which is perhaps more representative of commercial refining than the Valley beater, but it only produces a few grams of material at a time. Alternatively, we could purchase a pilot refiner, but they are much larger than we need and are very expensive to buy and operate.

Bill Sampson

I think we have one of the last surviving Medway beaters in Manchester. It works like a Valley beater but at about 10 times the speed and processes a 120 g batch; it leaves almost no fibres after 20 minutes. You should come and visit us with a bucket.

Martin Humval SCA

Have you looked into the effect of hygroexpansion between adding MFC and traditional refining and beating during drying, for example?

Jon Phipps

We haven’t investigated the effects of MFC on hygroexpansion. However, one of the things that we observed during trials is that lateral shrinkage of the paper web is lower when MFC is used to improve strength compared with increased refining. But I do not have any quantitative data on this, just observations reported from trials.