

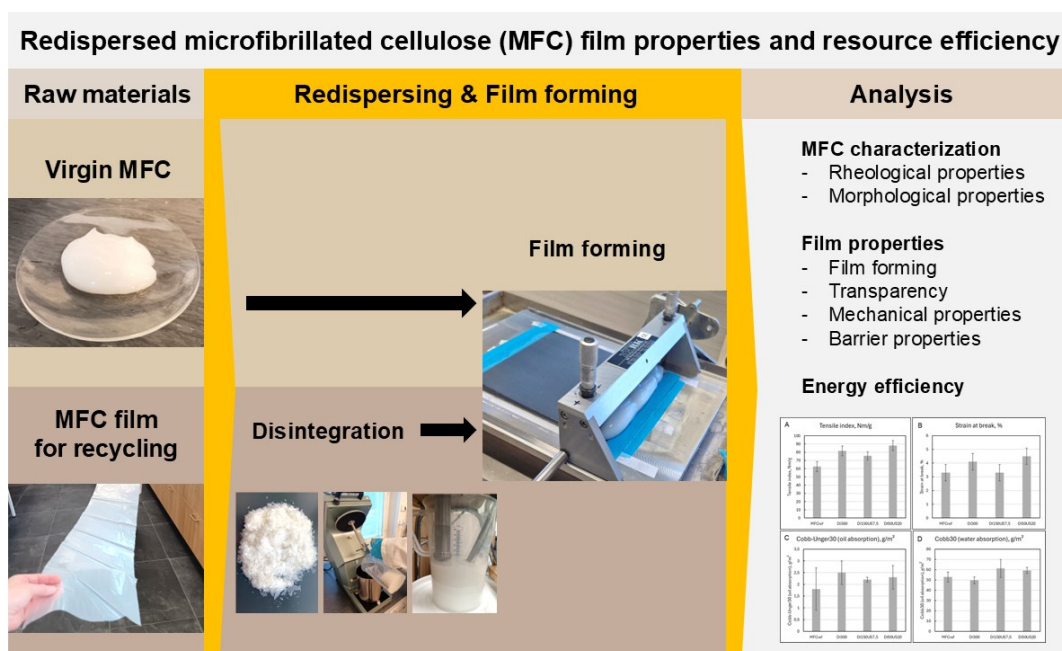
Redispersed Microfibrillated Cellulose Film Properties and Resource Efficiency

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GRAPHICAL ABSTRACT



Redispersed Microfibrillated Cellulose Film Properties And Resource Efficiency

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Microfibrillated cellulose (MFC) is a bio-based material produced by disintegrating cellulose fibers into microscale fibrillar components. Owing to its biodegradability, renewable origin, and favorable oxygen and oil barrier as well as film-forming properties, MFC has potential for sustainable packaging applications. However, its broader utilization is limited by the high energy consumption associated with the fibrillation process. One approach to improving resource efficiency is to collect and reuse any off-specification MFC films, such as products having incorrect grammage, the presence of wrinkles, or excess material related to trimming. In this study, the redispersion of MFC films using an ultrasound-assisted disintegration method was investigated, with focus on film properties and energy efficiency. The results showed that ultrasound-enhanced treatment during recycling improved the UV barrier performance of MFC films, reducing UV transmittance by 24 to 35% compared to unrecycled reference films. Longer ultrasonication time increased the visible light transmittance 14% compared to the reference film with same thickness. In addition, tensile properties were enhanced, with increases of 36% in strain at break and 41% in tensile index. The findings demonstrate that recycling of MFC films does not compromise, and may even improve, mechanical and barrier properties, while enabling notable savings in raw materials, energy, and costs.

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Keywords: Microfibrillated cellulose; Package film; Recycling; Redispersing; Ultrasound; Film formation; Tensile strength; Transmittance; Energy efficiency

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INTRODUCTION

Packaging is the driving force behind today's paper industry, accounting for 81% of current demand, and this trend continues to grow. The shift from plastic packaging towards fiber-based alternatives, known as "paperisation", marks a major change in the industry (Little 2025). Fiber-based packaging plays already an important role in the circular economy. In the EU, its recycling rate exceeds 87%, surpassing all other packaging materials (Eurostat 2025). Research also shows that consumers find fiber-based packaging easier to recycle compared to other types (McKinsey 2025; Two Sides 2025). Replacing plastics with fiber-based materials is especially important in food packaging. However, progress has been slower and costlier than expected, which is largely due to technical and economic challenges. As a result, current research is focusing on developing sustainable barrier technologies and coatings to improve the functionality of fiber-based packaging

without compromising its recyclability (Little 2025).

Today, biopolymers such as cellulose offer promising alternatives to conventional fossil-based plastic packaging materials. The key advantages of cellulose-based packages include biodegradability, renewable resources, and recyclability (Mujtaba *et al.* 2022; Nadeem *et al.* 2022). Cellulose can be further fibrillated into micro or nanoscale to obtain additional properties including enhanced mechanical and barrier properties, as well as film forming properties (Lavoine *et al.* 2012). However, the intense fibrillation method also has downsides, including high costs and energy consumption (Nadeem *et al.* 2020). Carefully engineered recycling of such highly fibrillated cellulose could bring the overall process more resource efficient.

Sourcing MFC Film for Recycling

This study examined the recyclability of clean, homogeneous microfibrillated cellulose (MFC) films. In practical applications, however, recyclable material streams are typically more complex, and uncontaminated MFC streams are not yet widely available for direct recycling. Moreover, MFC materials are presently considered primarily as candidates for future packaging solutions rather than established commercial products.

Nonetheless, recycled MFC could potentially be obtained from selected production-side streams. One option to improve the process efficiency is to efficiently reprocess any MFC films that had not resulted in saleable product. For instance, the initially produced film might have had the wrong mass per unit area (grammage). Alternatively, the initial film might have exhibited unacceptable variability in certain properties, *e.g.* mechanical or barrier properties. Another example is MFC film edge cuttings generated during film manufacturing. VTT (2026) describes a slitter-winder configuration in which substantial amounts of clean MFC film are trimmed from the film edges prior to reeling onto the core. Such edge trim represents a readily recoverable and homogenous material stream suitable for recycling.

The studied approach would allow reusing of MFC films in high value applications, such as renewable packages. Recycling films is an important consideration because repulped MFC films can be reused for film production, which minimizes waste and may reduce energy use and costs (Nadeem *et al.* 2024), especially in post-industrial applications. However, separating MFC from a used product, *e.g.* in post-consumer applications, is expected to increase costs and energy requirements. Previous techno-economic analyses have indicated that the energy required for MFC production is not the dominant cost factor when compared to raw material acquisition and downstream processing (Zambrano *et al.* 2020).

Nevertheless, in post-consumer scenarios, MFC film can be repulped along with the rest of the cellulosic material, thereby contributing to the recycled paper. The MFC fraction could potentially be recovered along with recycled cellulosic fiber fines, which can then be used in standard paper manufacturing. For example, incorporating recovered MFC is likely to reduce the air-permeability of the resulting paper products. Additionally, studies have shown that MFC can be produced from recycled pulp, such as recycled paperboard (Balea *et al.* 2023). Therefore, recycling MFC films may generate a pulp fraction that is both larger in quantity and better in quality for MFC production. In this scenario, the recovered MFC fraction could be further processed according to the methods outlined in this study. However, this route would likely require additional filtration, cleaning, and refibrillation steps to ensure sufficient MFC quality for film formation, which in turn would acquire energy and lower the efficiency.

Furthermore, another consideration would be to form MFC films as a convenient way to transport the material in dry form to another manufacturing facility. It is important to show that such dried films can then be used to manufacture films of high quality, even after they had been dried in the course of the initial forming process. Future research should further address recycling strategies for fiber-containing MFC-coated packaging materials, as well as the design of MFC film-fiber structures that enable efficient separation and recycling.

Redispersion of MFC Film

The aim of the study was to improve the recyclability and resource efficiency of MFC films and to investigate the effects of recycled MFC films on mechanical and barrier properties. In this study, the effect of ultrasound-assisted redispersion was evaluated. Disintegrating dried CNF is challenging due to irreversible or partially irreversible changes caused by drying. When bound water is removed, cellulose fibrils form internal hydrogen bonds and move closer together. This also causes nanoscale interfibrillar pores to collapse and close, making it hard for CNF to reswell when rehydrated, resulting in persistent aggregation and poor redispersibility. Known as hornification, this effect is common during recycled paper pulping (Scallan and Laivins 1993; Ang *et al.* 2021; Sjöstrand *et al.* 2023; von Schreeb *et al.* 2025) and increases energy demands for redispersion. These mechanisms are similar to those found in conventional pulps but are more noticeable in MFC because of its high specific surface area and extensive fibril–fibril contact. Ultrasonic disintegration is currently the primary approach for breaking down hornified CNF (Hoo *et al.* 2022).

Ultrasonic cavitation uses high-frequency sound waves to form bubbles in liquid, and their collapse produces energy that separates particles or molecules. This effect generates localised heat and pressure, aiding nanofiber separation. Disintegration efficiency varies with cavitation intensity, which is controlled by ultrasonic frequency, power, medium consistency, temperature, and duration (Hoo *et al.* 2022). Further studies could provide additional insight into the ultrasound-assisted disintegration stage, including the nature of bonds affected by the applied mechanical energy. These effects may relate to surface bonds formed during early drying and hornification or to other potential delamination sites. In the present study, however, the focus is on the influence of ultrasound-enhanced disintegration on film properties and resource efficiency.

Previous studies have investigated the redispersion of micro and nanocellulose film into recycled films in packaging applications using various disintegration methods. Next is a compilation of results from such studies. It should be noted that depending on the degree of refinement and the resulting fiber dimensions, the material may be referred to by various overlapping terms, including microfibrillated cellulose (MFC), nanofibrillated cellulose (NFC), cellulose microfibrils (CMF), and cellulose nanofibers (CNF). For clarity and consistency, this article refers to the studied material as MFC throughout. When citing previous research, the original terminology used by the authors (such as CNF or NFC) is preserved.

The recyclability of CNF films and the effects of disintegration methods on the properties of regenerated films have been previously investigated (Shanmugam *et al.* 2019; Silva *et al.* 2021; Nadeem *et al.* 2023; Setter *et al.* 2023). Shanmugam *et al.* (2019) and Nadeem *et al.* (2023) investigated the redispersion of nanocellulose films prepared from commercial Daicel CNF using purely mechanical disintegration methods. Their studies showed that the air permeance of the virgin CNF film was low (0.003 $\mu\text{m}/(\text{Pa}\cdot\text{s})$) and

remained low after recycling ($0.0045 \mu\text{m}/(\text{Pa}\cdot\text{s})$), despite a 50% increase in air permeation, indicating a reduction in air-barrier performance.

Water vapor permeability was examined at different disintegration levels by varying the number of revolutions between test points. According to Shanmugam *et al.* (2019), at 75 000 revolutions, the water vapor permeability of recycled films increased by $7.5 \times 10^{-11} \text{ g}/(\text{m}\cdot\text{s}\cdot\text{Pa})$ compared with virgin films. In contrast, Nadeem *et al.* (2023) reported that at 300 000 revolutions, the water vapor permeability of recycled films remained comparable to that of virgin CNF films, approximately $1.5 \times 10^{-10} \text{ g}/(\text{m}\cdot\text{s}\cdot\text{Pa})$. Setter *et al.* (2023) examined the recycling of TEMPO-oxidized CNF films using CTA surfactant addition and ultrasound-assisted disintegration. Their results indicated that redispersion caused a slight increase in water vapor permeability.

Tensile properties have also been widely studied in previous redispersion research. Using simple mechanical disintegration at 75,000 revolutions, Shanmugam *et al.* (2019) observed a 30% reduction in tensile index after recycling. Nadeem *et al.* (2023) showed that increasing the disintegration level to 300,000 revolutions resulted in tensile index values similar to the virgin film ($\sim 50 \text{ Nm/g}$). Their findings were consistent with those of Shanmugam *et al.* (2019), as the tensile index decreased by approximately 20% at 75,000 revolutions and 30% at 150,000 revolutions. Furthermore, Nadeem *et al.* (2023) investigated second-cycle recycling and found that the tensile index decreased an additional 15 to 20% compared with the first recycling cycle.

Silva *et al.* (2021) studied the effect of drying conditions on the tensile properties of recycled films and demonstrated that higher drying temperatures and an increased number of drying/redispersion cycles led to lower tensile strength and elongation due to an increase in aggregate formation. Furthermore, Setter *et al.* (2023) showed that ultrasound-assisted redispersion of TEMPO-oxidized CNF films increased tensile strength by 32% compared to the virgin film. Collectively, these results highlight that different treatments applied to MFC/CNF suspensions noticeably influence the mechanical properties of films produced from redispersed cellulose microfibrers.

Optical properties have also been explored in previous studies. Nadeem *et al.* (2023) reported only a minor decrease in light transmittance after the first recycling cycle. However, during the second cycle, transmittance dropped to approximately half of the original value. Setter *et al.* (2023) observed that spray-dried and recycled films generally exhibited lower transmittance, with the exception of films containing CTAB surfactant, which displayed improved transmittance after recycling. This improvement may be attributed to an increase in the specific surface area of MFC/CNF caused by ultrasonic homogenization, which likely enhanced electrostatic repulsion and resulted in less dense fibrillar clusters.

Overall, previous studies have demonstrated that appropriate redispersion can produce recycled MFC films with mechanical and barrier properties comparable to those of virgin MFC. All studies indicated that for the properties of regenerated films, it is particularly important to achieve high fibrillation and break down MFC agglomerates or aggregates formed during the initial film formation process. Studies show that laboratory refining did not provide further improvement beyond optimal revolutions (Nadeem *et al.* 2023). However, ultrasonication proved to be an effective method for separating fibers (Setter *et al.* 2023).

Based on preliminary investigations (Haatanen *et al.* 2025), ultrasound-enhanced mechanical disintegration was selected as the redispersion method. In this study, two different ultrasonication assisted approaches were implemented and compared with

conventional mechanical disintegration methods with high revolutions. This study introduces the mechanical and barrier properties of both reference and redispersed films. More detailed description of developing the disintegration method applied in this study was previously reported (Haatanen *et al.* 2025).

Ultrasound has already been adopted as a deagglomeration method and is widely employed method for nanocellulose redispersion (Hoo *et al.* 2022). However, previous studies have not yet investigated the effect of ultrasound-assisted mechanical disintegration (without chemical treatment) and the effect of ultrasonication parameters (time) on the mechanical and barrier properties of the recirculated films. The experimental section of this article describes the applied disintegration methods and film forming procedure to produce the test samples. The results and discussion section introduces the properties of the MFC sample used in this study and discusses the effect of recycling on film properties and resource efficiency.

EXPERIMENTAL

Materials

Microfibrillated cellulose (MFC)

MFC was prepared from dried birch-based bleached kraft pulp (Scandinavian pulp mill), which was soaked in reverse osmosis water overnight and pulped at a dry matter content of 5%. MFC was prepared by mechanical two-stage disintegration. The dispersed pulp (1.9% DM) was first refined with a grinder (Supermass Colloider MKZA10-15J, Masuko Sangyo Co., Japan) with MKE10-46 grinders at 1 500 rpm. The process was carried out twice, with the grinding gap set to 0.12–0.16 mm in the first pass and to 0.16–0.18 mm in the second pass. The grinding steps were followed by a treatment with a fluidizer (Microfluidizer MF7125-30, Microfluidics, USA). Microfluidization was performed in three passes using the flow cells APM400-IXC200 and APM400-IXC100 at 1 800 bar. No chemical modification was applied. The original MFC is hereafter referred to as M2F3, according to the method of preparation.

Plasticizer

A plasticizer was used as an additive with M2F3 MFC. D-Sorbitol (purity $\geq 98\%$) with a molecular weight of 182.17 g/mol (powder) was acquired from Sigma-Aldrich. Prior to mixing with M2F3 MFC, the D-sorbitol powder was diluted to 50 TS% with deionized water and stirred with a magnetic stirrer for 10 min at room temperature until the solution was clear. Then, 20% w/w of D-sorbitol was added to the M2F3 MFC sample and mixed using a vacuum dissolver (VMA Dispermat VL 10-35-AB, Germany) with a 150 mm diameter dissolver disc for 60 min at 100 mbar(a) and 2 200 rpm. The MFC D-sorbitol mix is hereafter referred to as MFCref. No other chemicals were used in the process.

Methods

Preparing the virgin MFC film with the pilot unit

The virgin MFC film was prepared with a special pilot-scale unit (SUTCO-pilot line, VTT Technical Research Centre of Finland/Bioruukki, Finland). A large pilot-scale piece of equipment was used to prepare the film, yielding enough MFC film for the following disintegration tests. The pilot line is shown in Fig. 1. Prior to casting, air was removed from the MFCref solution using a vacuum dissolver (VMA Dispermat VL 10-35-

AB, Germany). The film-forming pilot line uses manual feeding, following a combination of a blade (set to 2.4 mm height) and a fine rod (set to 2.3 mm height). The MFCref was cast on a plasma-treated polypropylene surface. Based on previous experience, it is observed that plasma treatment enhances the adhesion between the surface and the cast film, preventing drying shrinkage. After casting, the film was allowed to dry under normal ambient conditions for a minimum of 72 h. Minor delamination of the MFC film from the surface during the drying process was observed. This was not critical, since the cast film was to be disintegrated in the following step. The prepared film had a thickness of approximately 30 μm .



Fig. 1. SUTCO-pilot line that was used to prepare the virgin MFC film

Disintegration of the MFC film

The virgin MFC film (Fig. 2A) was then disintegrated for the subsequent recycling tests (South-Eastern Finland University of Applied Sciences/ FiberLaboratory, Finland). Three different disintegration methods were tested. These three methods were selected based on a comprehensive pre-trial (Haatanen *et al.* 2025). At all three test points, the dry film sample was pre-refined in a high-speed kitchen blender (model MMBH6P6B/01, Robert Bosch Hausgeräte GmbH, Germany) in small portions and ground at maximum power, 45 000 rpm, for approximately 10 to 30 seconds. After pre-refining, the powdered MFC film (Fig. 2B) was soaked in deionised water at room temperature for at least 24 h without stirring.

After soaking, all samples were disintegrated at room temperature using a laboratory disintegrator (L&W Pulp Disintegrator model SE 003, USA) (Fig. 2C). The number of revolutions varied between the test points. The rotational speed in the laboratory disintegrator was constant at 3000 rpm.

Further disintegration was performed for the selected test points using an ultrasonic sonicator (Hielscher UIP1000hdT, Germany) combined with a basic homogenizer (IKA T18 Ultra-TURRAX dispersing instrument) to enhance mixing. The setup is shown in Fig.

2D. The ultrasonic sonicator was always operated at full power (1000 W, 20 kHz). The homogenizer was also operated at full power (level 5), corresponding to 25,000 rpm.

Table 1 presents the three test points and their disintegration methods, including the number of revolutions used in the laboratory disintegrator, the duration of ultrasonic treatment, and the target consistency of the sample at each test point.

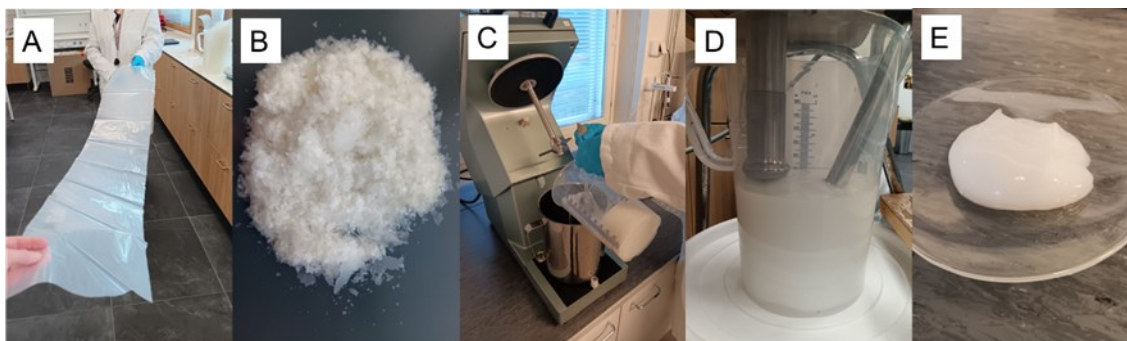


Fig. 2. Disintegration phase of the MFC film: A: a sample of the virgin MFCref film, B: pre-ground MFC film, C: laboratory disintegrator, D: ultrasonic disintegration setup, and E: redispersed MFC sample (DI150US7.5)

Table 1. MFC Disintegration Phase Test Points

Test point	Target consistency* (%)	Disintegration (revolutions)	Ultrasonic treatment (min)
DI300	3	300 000	none
DI150US7.5	2	150 000	7.5
DI50US20	2	50 000	20
*This parameter is based on the amount weighed for the process Sample acronyms: DI= disintegration, US= ultrasonic treatment			

The disintegrated MFC samples and the original MFCref sample were preserved in a refrigerator (7 °C) in sealed containers for approximately 4 to 8 weeks before casting into redispersed MFC films. The sample volume was approximately 2 liters per test point.

Preparing the recycled MFC sheets with the laboratory unit

The disintegrated MFC samples were used to prepare recycled MFC films. The films were prepared with a laboratory unit due to the limited sample amount available for the tests. The laboratory unit is located at the VTT Technical Research Centre of Finland, Jyväskylä unit. To ensure comparability between the test point results, a reference sample, parallel to the virgin MFC film from the pilot line, was also prepared with the laboratory unit.

For the MFC recyclability tests, four batches of laboratory sheets were prepared: one reference point and three different MFC grades processed using different disintegration methods, as described above. In total, 12 sheets per test point were prepared. The reference sheets were cast from virgin M2F3 MFC pulp with 20% w/w D-Sorbitol added. The recycled MFC film sheets were cast from pulp made entirely from disintegrated MFC film. No D-Sorbitol was added to these sheets, as the original film already contained D-Sorbitol, and none was removed during disintegration since the process did not involve any removal steps.

MFC samples were stored in a refrigerator and were not subjected to any thermal treatment prior to processing. Before each casting, the MFC sample was dispersed for 30 min using a high-shear mixer to remove entrapped air and ensure uniform composition. Fig. 3A shows the mixer (ShearMaster, Netzsch, Germany) and the high-shear blade.

MFC sheets were cast using a laboratory coater (K202 Control Coater, RK Print Coat Instruments, U.K.) with micrometer adjustable applicator and casting speed 3.5 m/min. Sheets were cast with a spreading blade onto a coated metal base plate (20 cm × 15 cm) with a casting area approximately 15 × 15 cm. The base plate acted as a carrier for the subsequent drying step. Surface properties affect the adhesion between the surface and the film, similar to the pilot line. The base plate coating was selected based on VTT's previous experiences with this MFC recipe. Prior to casting, the gap between the blade and base plate was adjusted to the desired thickness using a feeler gauge. The targeted sheet thickness was 30 µm. The wet casting thickness (t_w) was determined from the calculated consistency following Eq. 1,

$$t_w (\mu\text{m}) = t_d / C \quad (1)$$

where t_d is targeted dry thickness (µm) and C is consistency (%). The MFC sample was manually placed in front of the blade, after which the device was started and the blade moved across the base plate, spreading the slurry evenly. Figure 3B shows the casting process in progress. Each test point started with an initial casting to verify the correct sheet thickness. The test sheet was dried, and the dry thickness was measured. If necessary, the gap in the laboratory coater was adjusted using a feeler gauge to achieve the target thickness.

The MFC sheets were dried with a non-commercial COMBO-dryer (Fig. 3C). The dryer comprises a moving heated carriage and a fixed impingement unit. The cast film sheet, together with its metal plate, was tightened over the curved carriage, and the carriage was automatically moved under the impingement unit, where it performed a reciprocating motion. The carriage temperature, airflow velocity, and temperature of the blowing air could be adjusted. The drying parameters were the same for all sheets: the heated plate temperature was 70 °C, the airflow velocity of the blowing unit was 20 m/s, and the blowing air temperature was 160 °C. Optimal drying parameters are required. Previous studies have shown that the drying temperature affects the trapped air bubbles (Lyytikäinen *et al.* 2021) and sheet strength (Ibrahim *et al.* 2022). Table 2 shows the wet thickness and drying time in each test point. Drying time refers to the time the moving carriage is located below the impingement unit and was determined by visual inspection until the sheet appeared uniformly dry.

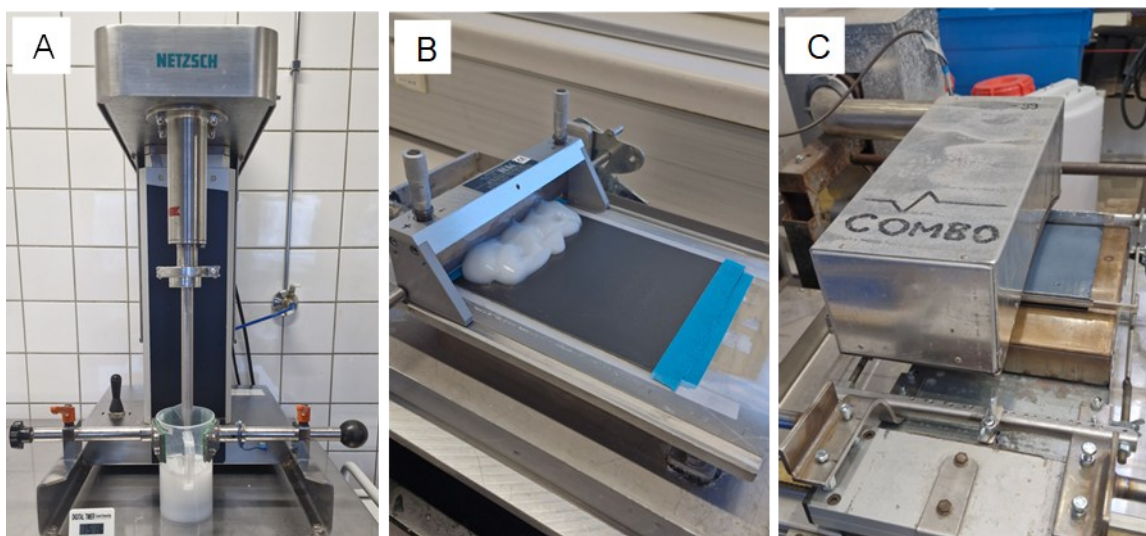


Fig. 3. A: High-shear blade used to mix the MFC samples prior to casting in the laboratory. B: Laboratory coater used to prepare the lab sheets. C: COMBO-dryer used to dry the lab sheets

Table 2. Sample Consistency, Sheet Wet Thickness, and Drying Time in each Test Point

Test point	MFCref	DI300	DI150US7.5	DI50US20
Measured consistency (%)	2.03	2.93	1.98	2.00
Wet thickness (μm)	1 400	900	1 150	1 550
Drying time (s)	240	150	270	280

Analyses and Determinations

MFC sample characterization

The consistency of the MFCref and disintegrated samples was measured according to the principles of standard ISO 638 (2008) with modifications to the temperature and drying time. Consistency was determined by oven-drying at 75 °C for 4 h, following an internal laboratory protocol adapted for heat-sensitive samples. The consistency of the MFCref was determined after the addition of D-Sorbitol.

The viscosity of M2F3 MFC and the disintegrated samples were measured with a high-shear capillary-based viscometer (ACA AX-100, ACA Systems, Finland). All samples were measured in the original consistency with a 1.0×120 mm capillary, low pressure program and shear rate values $25\,000\text{ s}^{-1}$ (1st round) and $1\,000\text{ s}^{-1}$ (2nd to 4th round). In addition, low-shear rotation viscosity was measured with a Brookfield viscometer (DV-II+Pro EXTRA, USA) with spindle number 6 and spindle speed 100 rpm at room temperature.

Nanoscale fiber width of MFC was analyzed using a scanning electron microscopy (SEM) (Hitachi S-4800, Japan) at an accelerating voltage of 1 to 2 kV and the magnification ranging from $\times 25,000$ to $\times 100,000$. Samples were not diluted before sample preparation. MFC samples were coated with palladium-gold layer using a high-resolution sputter coater (Cressington 208HRD, Malaysia) prior to SEM imaging. Microscale fiber properties such as fiber length, fines particles and fibrillation of M2F3 MFC and the disintegrated samples were measured with a fiber image analyzer (Valmet FS5, Finland).

MFC film characterization

The following MFC film properties were tested: thickness, grammage, transparency, mechanical strength, and barrier properties. Barrier properties included oil and water absorption, air permeance, and water droplet contact angle measurement. All samples were conditioned in accordance with ISO 187 (2022) before testing.

The thickness of the test sheets was determined in accordance with the standard ISO 534 (2011) using the single sheet thickness and by averaging multiple measurements. Grammage of the test sheets was determined in accordance with the standard ISO 536 (2019) with deviations regarding the sheet dimensions.

The light transmittance was assessed using a UV-Vis spectrophotometer (Lambda 900, PerkinElmer, USA) following the principle of (ASTM D1746 2023) standard. The tests measured the transmittance percentage (T%) of the sheet over a wavelength range of 200 to 800 nm with a 10 nm interval. The light transmittance was determined against air as a standard. The measurement area was 1 cm × 5 cm, and two parallel samples were analyzed. In addition, transparency was assessed with a visual inspection.

Tensile properties were measured with a material testing machine (ZwickRoell zwickiLine Z2.5, Finland) following the principles of the standard ISO 1924-3 (2005). The test strips were 15 mm wide, the gauge length was 50 mm, and the testing speed was 5 mm/min. Measurements were performed in one direction, corresponding to the casting direction, since the fiber orientation was not expected to be affected by the casting process. Ten parallel measurements were carried out for each sample.

Air permeance was assessed using two standardized measurement methods: the Bendtsen method and the Gurley method. These measurements were conducted in accordance with ISO 5636-3 (2013) and ISO 5636-5 (2013), respectively. The Bendtsen method uses a porosity tester (Messmer Buchel PPS-tester, the Netherlands) and is particularly useful for materials with very low air permeability, applicable to paper and board with air permeability in the range of 0.35 to 15 $\mu\text{m}/(\text{Pa}\cdot\text{s})$. It is not suitable for rough-surfaced materials that cannot be reliably sealed to prevent leakage. The Gurley method uses a densometer (Gurley Precision Instruments GPI, USA) and is applied to papers and boards with air permeability in the range of 0.1 to 100 $\mu\text{m}/(\text{Pa}\cdot\text{s})$ and is also suitable for determining the air resistance of paper and board. Three parallel measurements were performed for each test point.

Oil absorption was measured using the Cobb-Unger method in accordance with the standard SCAN-P 37:77 (1976). In this study, a 30-second measurement procedure was used. Three parallel measurements were performed for each test point.

Water permeability was measured using two different methods. Water absorption was determined by the Cobb method in accordance with ISO 535 (2023). In this study, the Cobb 30 value was measured using a 30-second exposure time. Three parallel measurements were performed for each test point. Wettability was assessed by contact angle using a goniometer (PG-3 Pocket Goniometer, Rycobel NV, Belgium) in accordance with standard TAPPI T 458 om-94 (1994) as a framework, with deviations in contact time before the picture was taken. In this study, the contact angle was measured 1 second after the droplet was applied, and the standard's second image at 1 min was not acquired. Two parallel samples with 5 to 9 measurements were performed for each test point.

RESULTS AND DISCUSSION

First, the rheological and morphological properties of the MFC samples used in the present study are presented and discussed. Next to be considered is the effect of disintegration on film properties. The subsequent results on mechanical and barrier properties refer to the produced films. Last to be discussed is the estimation of energy consumption and the overall resource efficiency of recycling.

Properties of MFC Samples Used in the Study

Rheological properties

An aqueous wet MFC sample has a complex and special rheological behavior. In the range of 2 to 3% consistency MFC is shear thinning or pseudoplastic, meaning that the viscosity decreases as the shear rate increases *e.g.* due to mixing, coating or casting (Lavoine *et al.* 2012; Kumar *et al.* 2016). This is an interesting character from a packaging application perspective, where the shear rates in blade, rod, and slot coating can be as high as 10^5 to 10^7 s^{-1} (Kumar *et al.* 2016). Under normal (quiescent) conditions MFC forms a stable and viscous gel-like formulation, but under high-shear stress viscosity decreases substantially, which enables efficient processing and facilitates uniform deposition and homogenous film formation. The measured high-shear dynamic viscosities are given in Fig. 4.

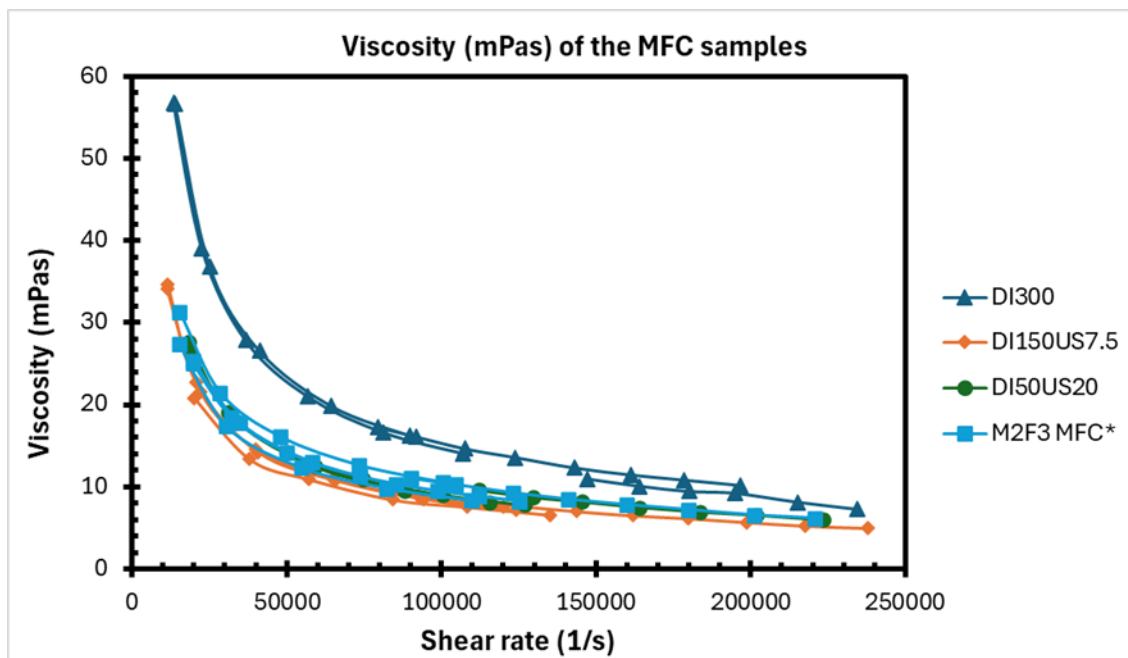


Fig. 4. MFC sample capillary viscosities. Sample consistencies are DI300 2.93%, DI150US7.5 1.98%, DI50US20 2.00% and M2F3 MFC 1.64%. *M2F3 MFC is the reference MFC with no D-Sorbitol added

The results in Fig. 4 reveal clear pseudoplastic behavior in all samples. There was no indication that the redispersion of the MFC would affect this behavior. However, the sample consistency influenced dynamic viscosity. Sample DI300 has higher consistency (~3%) compared to other samples (~2%) and thus higher viscosity. At high-shear stress areas, the difference in viscosity caused by sample consistency decreased and became negligible.

In addition, low-shear Brookfield rotational viscosities were measured. These values were consistent with the previous high-shear measurements and no notable effect from film redispersion was identified. M2F3 MFC, DI150US7.5 and DI50US20 exhibited comparable viscosities of 4 480 mPa·s, 4 450 mPa·s, and 5 050 mPa·s, respectively, whereas DI300, with its higher consistency, showed a viscosity of 9150 mPa·s.

Morphological properties

The characterization of morphological properties of MFC can be challenging due to its heterogenic and especially multi-dimensional nature, from nanometer to micrometer length scales. The specific MFC fiber dimensions vary and depend upon the source of the cellulose and the method of production. However, there are generally accepted dimensions for MFC fibers, which are roughly 5 to 100 nm in width and several micrometers in length. (Lavoine *et al.* 2012; Moon *et al.* 2025). Fiber image analyzer, with detection limit 1 μm , was used in the present study to measure fiber length. SEM images were applied when estimating the nano scale fiber width. The results are given in Table 3 and SEM images with 25 000 $\times$ magnification in Fig. 5.

Generally, MFC owes most of its unique properties to its long, narrow fibers that give it a high surface area. This is expressed as the aspect ratio (a), which can be calculated according to Eq. 2 (Sellman *et al.* 2024),

$$a = L / w \quad (2)$$

where L is the fiber length (μm) and w is the fiber width (μm). The aspect ratio can be used to characterize MFC mechanical properties (Sellman *et al.* 2024). All the MFC samples used in the study had relatively high aspect ratios, above 1000 (Table 3). However, it is important to remember that the aspect ratio uses fiber width which is based on visual image analysis, and thus the result is only indicative. Relatively high aspect ratio of the applied MFC could enhance the film's properties, such as light transmittance and mechanical properties.

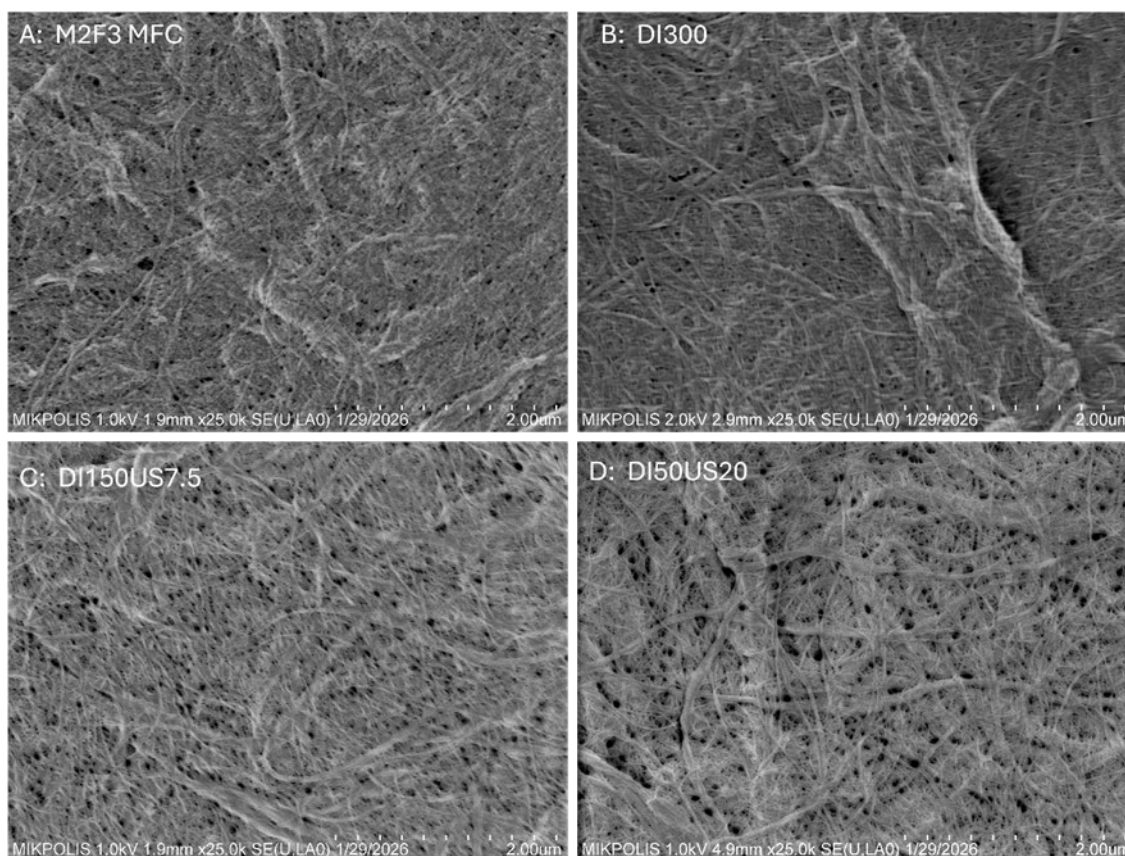


Fig. 5. SEM images of the M2F3 MFC* (A) and redispersed MFC film samples (B-D) with 25 000 times magnification.* no D-Sorbitol added

Table 3. MFC Samples Fiber Analysis Results

	MFCref	DI300	DI150US7.5	DI50US20
Fiber width (nm)	10 to 40	10 to 40	10 to 40	10 to 40
Fiber length (μm)	33	33	38	38
Aspect ratio	1320	1320	1520	1520
Fines B (%)	86.4	84.8	94.6	93.8

Fiber width is estimated from SEM images excluding microscale fibers. Fiber length is the arithmetic mean fiber length. Aspect ratio is calculated using Eq. 2 with average fiber width of 25 nm.

The fiber image analyzer was also used to characterize the residual fiber fragments and fibrillation. Even though the method is not compatible for detecting individual MFC particles in nanoscale due to the detection limit, the larger microfibril bundles and non-fibrillated share are characterized. For instance, the film barrier properties can be detected (Padberg *et al.* 2016). According to the results (Table 3), ultrasonic treatment increased the share of fibrillar fines (fraction Fines B). For the MFCref and DI300 Fines B fraction was 86.4% and 84.8%, respectively. Whereas for the ultrasonically treated samples DI150US7.5 and DI50US20 the Fines B fractions were higher, 94.6% and 93.8%, respectively. Results indicate that the ultrasound-assisted treatment can improve the efficiency of the defibrillation process, which is in line with the previous studies (Hoo *et al.* 2022; Setter *et al.* 2023).

Effect of Disintegration on Film Properties

Film forming

All samples were successfully cast into films without major issues. However, small air bubbles were observed in all films, including the reference film. These bubbles were visible on both the wet and the dry sheets. Figure 6A shows an image of a wet sheet cast from slurry DI150US7.5, revealing air bubbles in the slurry after dispersion. The white spots represent small microbubbles that cannot be eliminated by the applied disperser. The dark spots are larger air bubbles that were broken during casting. Figure 6B shows an image of the same cast after drying, revealing different sizes of air bubbles still present in the dry sheet.

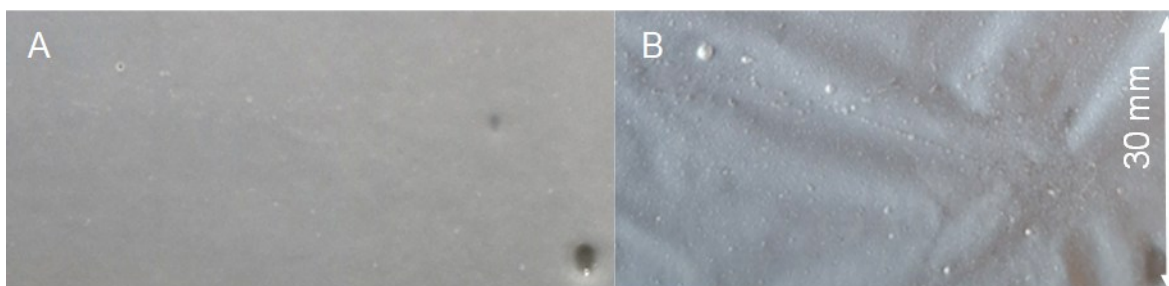


Fig. 6. Visible air bubbles in sample DI150US7.5 A: wet sheet and B: dry sheet

Because of the air bubbles, the sheet surface appeared slightly uneven which can affect the film thickness results. Film thickness and grammages are given in Table 4. The measured thickness was greater than the calculated value. However, the results remained comparable across samples. The increased sheet thickness particularly impacts tensile strength (MPa) and transparency measurements, as these calculations take sheet thickness into account.

Light transmittance and transparency

The films showed a small variation in transmittance across the entire wavelength range, indicating a minor influence of film recycling (Fig. 7). At ultraviolet (UV) wavelengths, a low transmittance value is beneficial, as it indicates effective attenuation of UV radiation by the film. UV-barrier property is beneficial for food packaging applications (Mujtaba *et al.* 2022). Lowest light transmittance in the UV-B region (280 to 315 nm) was observed for DI150US7.5 and DI50US20 ranging from 4 to 6%. In the UV-A region (315 to 400 nm) MFCref and DI300 remained the highest 8 to 12% and DI150US7.5 the lowest 5 to 9.5%, while for DI50US20 light transmittance increased steeply to the level of MFCref and DI300. The results indicate that the ultrasonication stage enhances the UV-barrier, especially in the UV-A region. Cellulose films with transmittance values in this range indicate suitable UV light barrier properties (Li *et al.* 2022).

In addition to UV barrier properties, adequate transparency is also preferred for food packaging films. The sample's light transmittance in the visible light region (400 to 700 nm) varied greatly. Film thickness also influences how transparent the films appear. Thus, the transparency (Tr) of the films was calculated following Eq. 3,

$$Tr (\%) = (\log (T600)) / th \quad (3)$$

where $T600$ is the transmittance in 600 nm (%) and th is the film thickness (mm) (Sothornvit *et al.* 2010; Mascarenhas *et al.* 2022). Table 4 presents the transmittance and transparency of the sample sheets at 600 nm wavelength. The highest transparency was observed for DI300 41.56%, followed by DI50US20 40.04%, MFCref 37.94%, and DI150US7.5 37.70%. The transparency value of the DI50US20 was lower than that of the DI300 film, although the light transmittance was higher. This is because the DI300 was thinner than the other films. Compared to other recycled microfibrillated cellulose films (Fotie *et al.* 2023; Setter *et al.* 2023) values reported here seem modest. This may be due to the larger fiber width and fiber aggregates (Mascarenhas *et al.* 2022). Visual translucency is presented in Fig 8.

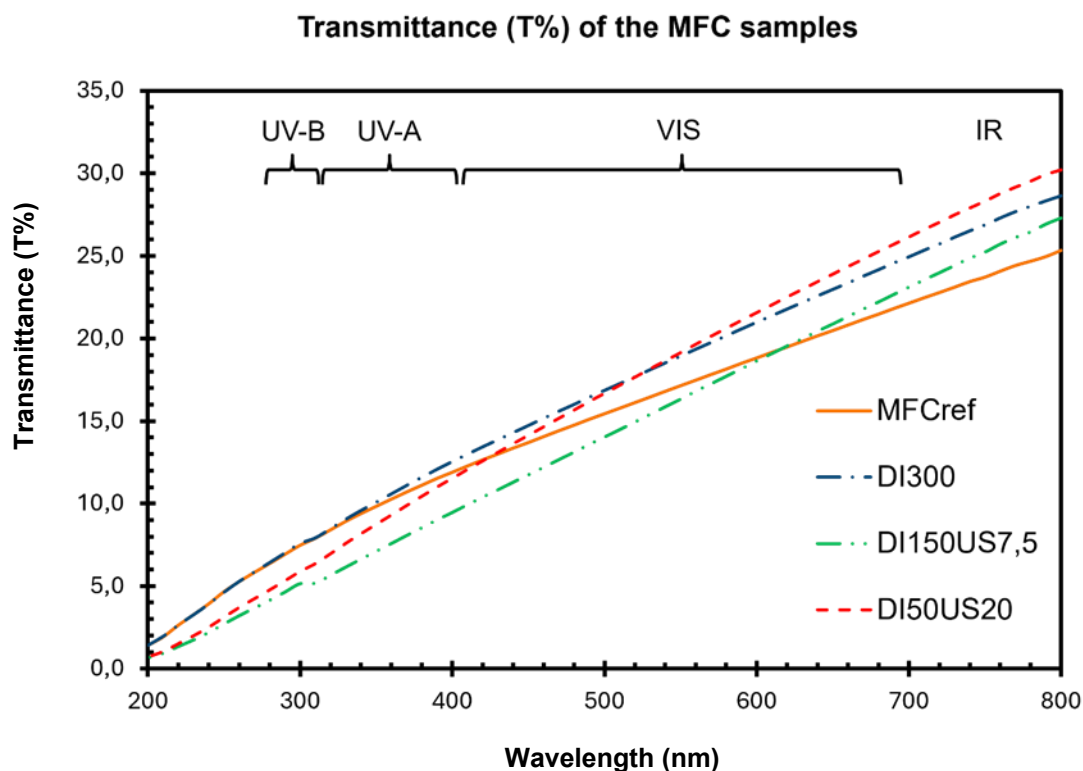


Fig. 7. Average transmittance curves for the test points across the entire measurement range

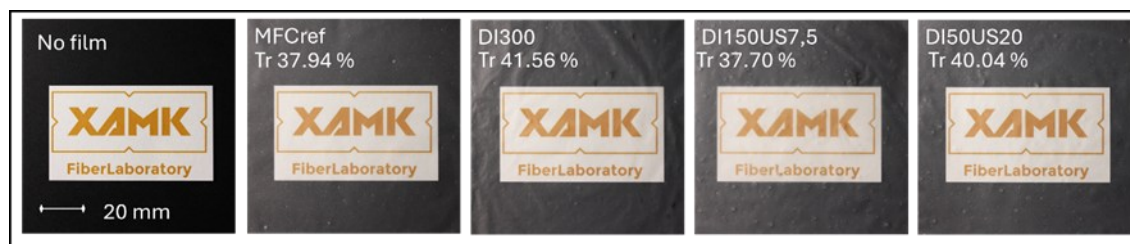


Fig. 8. Visual inspection of the film transparency

In the infrared (IR) region (> 700 nm) the DI50US20 film gave the highest light transmittance results, followed by the DI300 and DI150US7.5 and MFCref with the lowest. However, the IR light adsorption is not a meaningful parameter in normal food packaging applications.

Mechanical properties

Reasonable mechanical properties are essential for most packaging applications. The mechanical performance of the films was assessed by measuring tensile strength and strain at break. Due to the variation in thickness and grammage among the samples, tensile strength was further expressed as a normalized tensile index. The corresponding results are summarized in Table 4. Tensile index and strain at break results are also depicted in Fig. 9A and 9B. It should be highlighted that the sheet grammage and thickness of DI300 was lower compared to other samples.

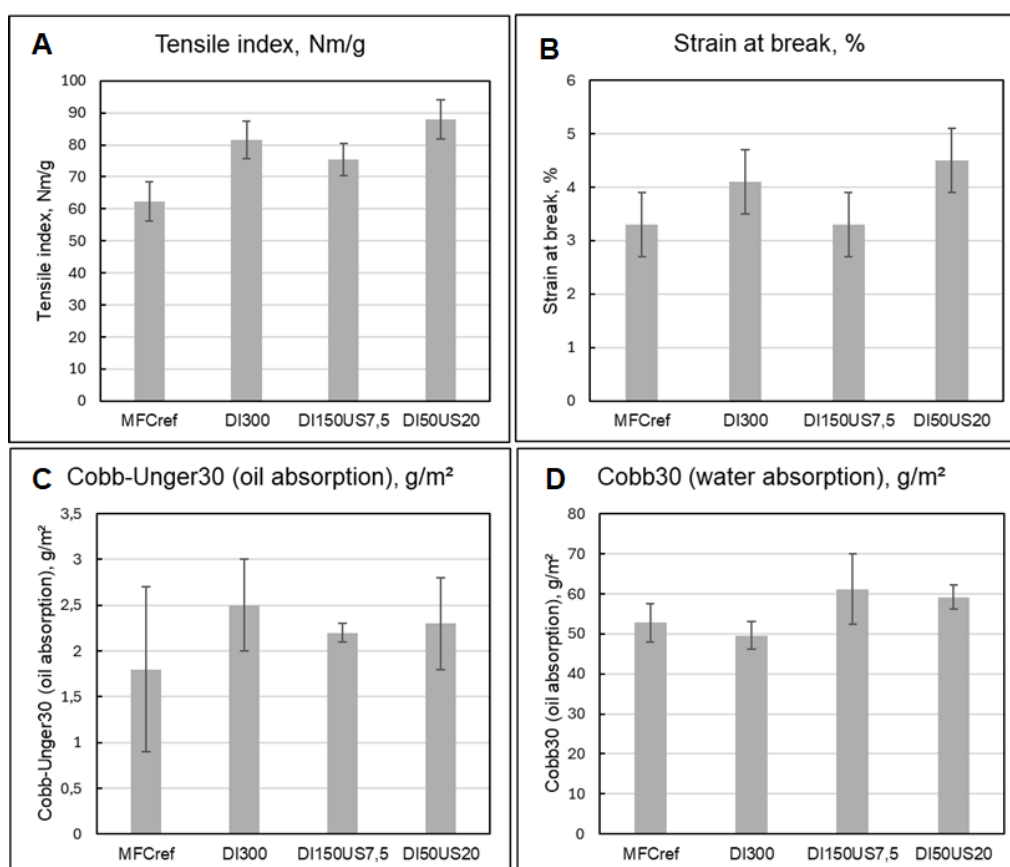


Fig. 9. Tensile index, Strain at break, Cobb-Unger30 and Cobb30 for the MFCref and disintegrated samples

According to the results, disintegration enhanced the mechanical properties of the MFC film. MFCref had the lowest results for tensile properties. High mechanical disintegration in sample DI300 increased the strain at break by 24% and the tensile index by 31%. The result is in line with previous studies, indicating that high revolutions with the mechanical disintegration can yield good results when recycling MFC films for the first time (Nadeem *et al.* 2023). Furthermore, the ultrasound-assisted sample DI50US20 gave the best results, increasing the strain at break by 36% and tensile index by 41%, compared to the reference film. Thus, it seems that ultrasonic treatment reduces the required

revolutions during the mechanical treatment and delivers even better results. Longer mechanical disintegration followed by a shorter ultrasonic treatment resulted in the least increase compared to MFCref.

Table 4. Sheet Mechanical and Barrier Properties

Test point	MFCref	DI300	DI150US7.5	DI50US20
Dry thickness (μm)	33.6 ± 1.1	31.8 ± 1.3	33.7 ± 1.8	33.3 ± 1.0
Sheet grammage (g/m^2)	31.3 ± 1.3	25.0 ± 1.0	31.5 ± 1.2	31.7 ± 0.8
Transmittance 600 nm (%)	18.82	20.97	18.64	21.55
Transparency 600 nm (%)	37.94	41.56	37.70	40.04
Strain at break (%)	3.3 ± 0.6	4.1 ± 0.6	3.3 ± 0.6	4.5 ± 0.6
Tensile strength (MPa)	57.0 ± 5.6	64.4 ± 4.6	68.3 ± 4.5	82.7 ± 5.8
Tensile index (Nm/g)	62.44 ± 6.14	81.59 ± 5.88	75.53 ± 5.02	88.08 ± 6.13
Bendtsen (air permeance) (ml/min)	2.16 ± 0.02	2.16 ± 0.02	2.12 ± 0.03	2.09 ± 0.04
Gurley (air permeance) ($\mu\text{m/Pas}$)	below the detection limit	below the detection limit	below the detection limit	below the detection limit
Cobb-Unger30 (oil absorption) (g/m^2)	1.8 ± 0.9	2.5 ± 0.5	2.2 ± 0.1	2.3 ± 0.5
Cobb30 (water absorption) (g/m^2)	52.8 ± 4.8	49.6 ± 3.4	61.2 ± 8.8	59.2 ± 3.0
Contact angle, $^\circ$	< 45	< 45	< 45	< 45

Values are given as average ($\pm$ standard deviation).

Barrier properties

Depending on the packaging application, the film most often requires one or multiple barrier properties, such as resistance to air, moisture, or grease. Air permeability of the films was determined using the Bendtsen and Gurley methods. In addition, water and oil absorption were measured using the Cobb and Cobb-Unger methods, and the water contact angle was measured to evaluate the films' wettability. The corresponding results are summarized in Table 4. Cobb30 and Cobb-Unger30 results are depicted in Fig. 9C and 9D.

MFC is recognized for its excellent air barrier properties, especially in low humidity (Kumar 2018). This was also supported by the present results. All measured air permeance results were at or below the detection limits of the employed apparatus ($0.1 \mu\text{m/Pa}\cdot\text{s}$), indicating that redispersion of the MFC films does not compromise their inherent air barrier performance.

In contrast, MFC is recognized for having poor moisture resistance. The MFCref and DI300 samples had the lowest Cobb30 water absorption results, namely 53 g/m^2 and 50 g/m^2 , respectively, indicating better water barrier compared to the ultrasonically treated samples DI150US7.5 and DI50US20, which had water absorption results of 61 g/m^2 and 59 g/m^2 , respectively. Cobb60 values below 100 g/m^2 can be considered relatively good water resistance for certain packaging applications. However, it should be remembered that the measurement time in the present study was the shortest possible, 30 s, and thus the results are not directly comparable. In addition, all contact angle results were below 90° , which indicated that the films had no hydrophobic properties. Values for all samples were less than 45° , and thus all samples had high surface wettability. However, the recycling stage showed only a minor reduction, indicating that the MFC can be recycled without notably reducing the water barrier or wettability.

Oil absorption results indicated only a minor reduction due to the disintegration stage. MFCref exhibited an oil absorption value of 1.8 g/m², which is considered highly suitable for most packaging applications (Bru *et al.* 2024). Ultrasonically treated samples DI150US7.5 and DI50US20 demonstrated the next best performance, with values of 2.2 g/m² and 2.3 g/m², respectively, followed closely by DI300 at 2.5 g/m².

Recycled MFC Energy Saving Potential

The use of CNF or MFC is limited by its energy-intensive production process and high cost. Recycling is considered a promising approach to improve the resource efficiency of MFC film. However, the energy efficiency of the applied redispersion method plays a crucial role (Nadeem *et al.* 2022). The following calculation provides an approximate estimate based on clean post-industrial MFC film. In contrast, more complex post-consumer scenarios require consideration of the additional energy requirements and costs associated with sourcing MFC for recycling or redispersion.

Production of CNF fibers is associated with high energy consumption. Depending on the pre-treatment and production methods applied, the energy requirement can be up to the range 25,000 to 75,000 kWh/tonne (Nadeem *et al.* 2022, 2024). It is to be noted that production of MFC can be done with considerably lower energy consumption compared to CNF due to less energy-intensive fluidization needed for MFC production.

Test point DI50US20 was selected for the energy consumption measurements because it had the longest sonication time and was thus assumed to have the highest energy consumption of the tested redispersion methods. The overall energy consumption of DI50US20 was 4,500 kWh/tonne, including pre-refining (with the share of 5% of the overall energy consumption), mechanical disintegration (28%), sonication and simultaneous homogenization (68%). Energy requirement to produce the virgin MFC M2F3 (two times Masuko refiner and three times fluidization) was 18,000 kWh/tonne. This indicates a possibility of 13,500 kWh/tonne energy saving when recycled MFC would be used instead of virgin MFC in MFC film production. With electricity price 50 €/MWh this means a notable 675 €/tonne energy saving.

A notable energy and cost saving of raw materials is achievable when recycled MFC is used as raw material instead of virgin MFC in MFC film production. Thus, recycling can be considered a noteworthy way to increase the resource efficiency of MFC production. As indicated by previous studies, in order to truly benefit from recycling of MFC film, the number of revolutions to disintegrate the fibers should be optimized while remaining the original mechanical and barrier properties (Nadeem *et al.* 2024). The present study showed that ultrasound-assisted disintegration can be one potential option, since it can help to reduce the number of revolutions in the disintegration phase and resulted in similar or even better mechanical and barrier properties.

CONCLUSIONS

1. Considering the results from the present study, recycling the MFC film from bleached kraft pulp by redispersing the film using ultrasound-assisted mechanical disintegration does not noticeably reduce the mechanical and barrier properties and can even enhance the properties. Thus, recycling can be considered a noteworthy way to increase the resource efficiency of MFC production.

2. Ultrasound-assisted treatment enhanced UV light barrier properties of MFC films. DI150US7.5 treatment yielded the best performance, with UV light transmittance (280 to 400 nm) ranging from 4 to 9%, which is 24 to 35% lower compared to the reference film.
3. Ultrasound-assisted treatment also affected the transparency of MFC films. Longer ultrasonication time (DI50US20) resulted in the best transmittance 600 nm value of 21.55% which is 14% higher compared to the reference film with similar film thickness.
4. Ultrasound-enhanced treatment resulted in better tensile properties. DI50US20 gave the best results, increasing the strain at break by 36% and tensile index by 41%, compared to the reference film.
5. Redispersion of the MFC films can maintain their intrinsic air barrier properties, with all reported air permeance values demonstrating excellent performance.
6. All MFC film samples showed moderate resistance to moisture. Nonetheless, redispersion had no impact on their water barrier properties or wettability. Oil absorption results indicated only a minor reduction due to the disintegration stage.
7. Theoretically, notable energy and cost saving of raw materials is achievable when recycled MFC is used as raw material instead of virgin MFC in MFC film production. However, the energy and costs related to sourcing the MFC for recycling or redispersion must be taken into consideration.

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Conflict of Interest

Authors have no conflict of interest.

Use of Generative AI

The authors used the AI tool Microsoft 365 Copilot GPT-5 to assist with the paper's language, including text translation from Finnish to English and editing. The data is original. The authors have reviewed and edited the content generated by the AI tool and take full responsibility for the accuracy of the presented information.

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