Twin-roll forming, a novel method for producing high-consistency microfibrillated cellulosic films

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Abstract Micro-nano fibrillated cellulose (MNFC) films have the potential for applications in, e.g., packaging and printed electronics. However, the production paradigm for these types of products has still not been established. This study uses twin-rollers to form films from high consistency (15% w/w) microfibrillated cellulose furnishes. MFC furnishes were produced at 20% wt dry matter content with enzymatic hydrolysis and PFI refining. We used the twin-roller method to spread the material over a supporting substrate by repeatedly passing between two parallel rollers with decreasing nip. Rheological behavior and physical properties of furnishes were analyzed. We found that only some furnishes with relatively short fiber lengths were formable. Refining improved the formation of the sheets. Roll-formed sheets showed comparable strength and formation to conventional wet-laid hand sheets.

Keywords Roll forming · Formability · Nanocellulose · Rheology · High consistency · Twin rollers · Microfibrillated cellulose · Enzyme treatment · PFI refining

Introduction

One of the most important development targets of the forest-based industries is the large-scale production of “nano boards” and “nano papers” from fine-scale lignocellulosic furnishes (González et al. 2014; Abitbol et al. 2016; Hubbe et al. 2017a; Benítez and Walther 2017; Fang et al. 2019). Micro–nano fibrillated cellulose (MNFC) is defibrillated macroscopic fibers composed of a population of fibril aggregates in a size range of approximately 10–100 nm fiber wide (Bharimalla et al. 2017; Morais et al. 2021). This compares to ordinary pulp furnishes with 20–60 µm width fibers. In the laboratory, MNFC-based products have been shown to have interesting properties and give the potential for developing advanced products with a low environmental impact (Lin and Dufresne 2014; Hoeng et al. 2016; Shanmugam et al. 2019). However, one of the principal limitations for scaling up these products is the difficulty in forming and dewatering MNFC webs (Rantanen et al. 2015; Dimic-Misic et al. 2017; Ahadian et al. 2021). After all, the current paper manufacturing infrastructure is designed to operate with traditional pulp furnishes,
which have completely different rheology and dewatering behavior than MNFC.

The physical and rheological properties of the MNFC furnishes must be considered for designing a proper web forming method. The material behavior of MNFC furnishes, such as viscosity, yield stress, shear-thinning behavior, and viscoelastic properties, depends on the raw material, method of production, and water consistency (Dimic-Misic et al. 2013; Schenker et al. 2019; Jaiswal et al. 2021). Recent developments in MNFC production methods, especially at higher consistency, allow us to look for a new web-forming method. This article examines twin-roll forming as a new method to produce web from high consistency MNFC furnish. Many other industries currently use twin roll forming, but its capability for MNFC furnishes has not yet been extensively investigated.

The physical properties of MNFC depend on the source, such as plants, bacteria, and marine animals, and the extraction and processing methodology (Lin and Dufresne 2014; Nechyporchuk et al. 2016; Dufresne 2019; Dhali et al. 2021). Nanocellulose production includes different approaches to converting large fibers (mm) to smaller units (nm). These methods include chemical fibril extraction by applying, e.g., strong acids or enzymes and/or mechanical defibrillation by grinding or homogenizing (Kim et al. 2015; Nechyporchuk et al. 2016; Phanthong et al. 2018). In addition, bacteria can also produce nanocellulose. In this constructive approach, the molecular unit build-up to a nanoscale unit (Wang et al. 2019). In general, cellulose nanomaterials could be divided into three distinct types, (1) cellulose nanocrystals (CNCs), (2) micro-nano fibrillated cellulose (MNFC), and (3) bacterial cellulose (BC) (Kim et al. 2015). Each type has specific rheological and physical properties which drive from different ranges of particle length, particle width, aspect ratios, fiber swelling, and degree of fibrillation (Hubbe et al. 2017b; Phanthong et al. 2018).

Another critical factor affecting physical properties is the furnish solids content (Hill 2008; Puisto et al. 2012; Lu et al. 2014; Li et al. 2015; Shafiei-Sabet et al. 2016). Because of the high water binding of MNFC, furnishes tend to gelate at relatively low consistency. Thus, to be in a workable viscosity range, many processes for manufacturing and processing MNFC operate at low solids (often less than 3%). Low solids processes tend to be highly energy-intensive—a fact that can severely restrict MNFC implementation.

Production of MNFC from fiber sources at high solids content gives a potential route for designing low-energy nanopaper production schemes. Paper and board production from conventional pulps at solids content above 4% has long been a target of manufacturers to realize energy and investment savings. However, limitations on furnish rheology and paper formation have limited the possibilities. MNFC can be produced at solids contents in the range of 4–40% through enzymatic or acid hydrolysis (Tian et al. 2017; Rahikainen et al. 2020; Pere et al. 2020). Thus, it is worth investigating if these types of furnishes can be formed into a web with potentially attractive properties.

Forming and dewatering sheets out of MNFC furnishes is challenging in any consistency range (Brodin et al. 2014). Currently, three of the most common laboratory methods to form MNFC sheets are casting, filtration, and spray deposition. In the casting method, the MNFC suspension forms a film in a mold like a Petri dish after drying in an oven or air. While casting provides high material retention, it is time-consuming because of difficulties in drying and controlling wrinkles. Although casting is currently used in the laboratory to make MNFC films, it has significant limitations in being used as an industrial method (Khan et al. 2010; Siró et al. 2011; Abitbol et al. 2016).

Filtration forming most closely resembles conventional papermaking. In this process, a low consistency suspension is filtered with a vacuum over a wire mesh. If enough fibrils retain on the wire, a web will build up over time (González et al. 2014; Zhu et al. 2014). Both retention and dewatering are much more challenging than conventional papermaking furnishes due to the small size of the MNFC particles. Low permeability wires improve retention but lead to dewatering problems. Conversely, strategies that result in a more open, permeable web may result in poor retention. While, for some furnishes and conditions, low solids production of nanopapers appears to be viable (Zhang et al. 2012; Rantanen et al. 2015; Ahadian et al. 2021), in many cases, dewatering and retention issues are problematic (Sehaqui et al. 2010; Varanasi and Batchelor 2013; Yang et al. 2017).

The spray deposition is a variation of filtration forming that may have some potential as an
industrially scalable method. In this method, the furnish is sprayed on a substrate. The web can be dewatered with vacuum, followed by pressing and thermal energy (Obara and McGinity 1994; Shanmugam et al. 2017, 2018). A number of studies recently investigated the spray deposition method in pilot-scale experiments. Sheets produced by spraying were found to be comparable to vacuum filtered sheets in terms of strength, while they were made at relatively higher consistency (1.5–1.75 wt%) and less time (Beneventi et al. 2015).

The twin-roll forming method (not to be confused with the paper machine twin-wire former) deforms the bulk material into a desirable shape (sheet or specific profiles) between two parallel rollers moving simultaneously. This method is suitable for materials that are isotropic and highly plastic. Industries commonly use roll forming include metal, synthetic polymer, and food, e.g., sheeting the bread dough. Roll forming is a continuous and robust method that fits industrially scale needs for certain materials. Roll forming is not particularly suitable for conventional pulps since these furnishes consist of relatively large fibers that must be organized evenly within the plan of the sheet. However, MNFC furnishes are much more plastic, gelatinous, and isotropic, thus potentially suitable for roll forming operations.

In this article, we investigate twin-roll forming as a new technology to produce nanocellulose (MNFC) films. Here, we are specifically interested in course MNFC materials produced with combinations of enzymatic hydrolysis and refining. This manufacturing approach is potentially scalable to industrial volumes. In this study, a family of MNFC furnishes were produced and formed into sheets at high solids with a laboratory roll forming unit. The roll forming process simultaneously dewaters the sheet through compression and forms a coherent web, thus resembling a combined pressing/forming operation.

Materials and methods

Materials

The pulp used in this study is once dried bleached birch hardwood kraft pulp (BHKP-0) provided by a Finnish mill. The BHKP-0 pulp was soak over night, disintegrated in a valley beater at 1.3% consistency, and adjusted to pH 5.7 with HCl. The solids contents were adjusted to 20% for subsequent treatment using filtration. All solids contents (consistencies) are reported in % as the mass of oven-dried solids/(mass of solids + water).

The BHKP-0 was hydrolyzed with an Endoglucanase enzyme (ECOPULP R, AB Enzymes) which is able to fibrillate and degrade the fibers (Willberg-Kejrilaerin et al. 2019). The hydrolyzed sample (BHKP-0-EH) was produced in a Kenwood chef food processor at 20% solids, 5.7 pH, 55 °C, and 1.5 mg enzyme /g pulp dosage for two hours. The enzyme was then deactivated by putting the material in an ice bath immediately after treatment and stored at +5 °C.

30 g of untreated and enzyme-treated pulp was refined in a PFI mill at 15% solids content for 5000 or 10,000 revolutions with zero gap. These samples are called BHKP-5 K, BHKP-10 K, BHKP-EH-5 K, and BHKP-EH-10 K, respectively.

The pulp morphological properties were determined with a fiber analyzer (METSO Fiber lab SN analyzer). The degree of polymerization (DP) was calculated from the pulp viscosity (SCAN-CM 15:99) using the Evans and Wallis equation (SCAN 15:88).

Rheological characteristics of the pulps were determined with frequency sweep tests using a rheometer (Anton Paar Physica MCR 302) with plate-plate geometry with a 25 mm diameter.

Sheet forming and characterization

MNFC films were formed by a rolling machine (Marcato Atlas 150) equipped with anodized aluminum rollers (Fig. 1). The films were produced to a constant thickness rather than a constant grammage. Solids contents of furnishes were adjusted to 15.5 ± 0.5% before the rolling process. Ten grams of furnish were placed between two 220 GSM blotting papers, which were then passed between the rollers with 1.92 mm nip. This process was repeated with 1.44 mm, 1.26 mm and 0.76 mm nip gaps successively until the films reached 380 ± 20 mm thickness. The roller speed was 6 rpm.

Filtered sheets of BHKP-EH-5 K were made as a reference under one bar pressure on 125 mm filter papers with 12–15 µm particle retention. Hand sheets from BHKP-0 were made according to (Tappi T-205).

Rolled or filtered films were pressed between two blotting papers at 6 MPa for 1 min. The
blotting papers were changed, and the cycle was repeated 13 times. After pressing, the final moisture was removed by drying at 105°C with a 1 kg weight to restrain the sheet.

The films’ topography was measured for wet and dried sheets using an optical surface roughness device (L&W OptiTopo). Dry sheet formation was measured with an Ambertec Beta formation tester (SCAN-C 92:09).

The tensile strength of the dry films was measured using an MTS 400/M. The tensile test procedure was based on Tappi T 494 om-01, except for using the 30 mm gap distance and 5 mm/min strain rate.

**Result and discussion**

Figure 2 shows the visual appearance of furnishes before forming between rollers. These materials differ from each other in enzymatic treatment and levels of refining. The microscopic images of the furnishes are shown in Fig. 3.

Table 1 gives more information about the MNFC properties. The mechanical processing with PFI increases swelling and external fibrillation strongly, and slightly increases the fines content. The combination of enzyme treatment with refining significantly breaks down the fibers. Notable in these series (samples BHKP-0-EH, BHKP-EH-5 K, BHKP-EH-10 K) is that the fines content increases dramatically, and one-third of the average fiber length decreases compared to the unrefined, untreated reference. The WRV...
(Water Retention Value) increase relates to increased fines content and external and internal fibrillation. Increased WRV implies more difficult dewatering and higher bonding in the sheet (Sjöstrand et al. 2019).

Generally speaking, both lab and industrial refiners are not that effective at cutting fibers and producing fibrils. Refiners are designed primarily to preserve the fiber length and fibrillate the surfaces and interior of the cell wall, increasing flexibility and bonding potential. The endoglucanase cocktail used in this study, on the other hand, attacks glucosides bonds at amorphous regions along the fibrils, shortening cellulose chain length and degrading the fiber structure. This type of enzyme is very effective at generating cell wall fragments and fibril aggregates, i.e., micro fibrillated cellulose, but at the expense of cellulose DP.

Fiber swelling, fibrillation, fiber length, fines content, and other properties all affect the rheological properties of a furnish. Thus, the combination of mechanical treatment and enzymatic actions can potentially affect the formability of a furnish over a wide range. Comparing furnishes’ viscoelastic properties gives a better understanding of formability.

The furnishes are examples of viscoelastic materials. Viscoelastic materials show a combination of solid and viscous behavior. Oscillatory tests in the linear viscoelastic region (LVE) quantify a material’s viscoelastic properties over a wide range of strain or stress frequencies. The frequency test results give storage modulus, \( G' \), and loss modulus, \( G'' \). The former explains the solid-like behavior of the material. At the same time, the latter defines the viscous regime of the material.

The oscillatory frequency test examines the effect of refining and enzyme hydrolysis on the pulp rheological behavior in Fig. 4. All furnishes display predominantly elastic, solid-like behavior. These results show inter-particle force network stability, typical in
high-solids content nanocellulose furnishes (Naderi et al. 2014).

Both enzymatic treatment and refining decrease the storage modulus. The treatments increase fiber swelling and flexibility and reduce the fiber network’s elasticity. The enzymatic treatment has a more substantial effect on storage modulus than refining because of the relatively high production of fines, which profoundly affects furnish rheology.

At the higher frequencies, the enzyme-treated material experiences a significant drop in $G'$, and the viscous regime of the material becomes more dominant (Fig. 4). This is because shorter fibers form a less stable solid network that yields at higher frequencies (Mendoza et al. 2018). In contrast, refined pulp and reference pulp (BHKP-0, BHKP-10 K) do not show such behavior. This result suggests that for the conditions used in this experiment, the enzymatic treatment produces a furnish that flows and is more formable than the refining.

The formability of furnishes was evaluated visually and using optical topography scanning. The measurements were done on the wet sheets after forming and before pressing and drying. We consider a furnish “formable” if it results in smooth and uniform film, while non-formable furnishes lead to films containing cracks and holes. Figure 5b, c compare the surface of films made of formable and non-formable furnishes, respectively.

Based on our evaluation, BHKP-0-EH, BHKP-EH-5 K, and BHKP-EH-10 K are formable, but BHKP-0, BHKP-5 K, BHKP-10 K are not formable (Fig. 5). This means that the reference pulp became formable only after enzyme treatment and not by refining alone. Surface variation for non-formable materials is three times higher than BHKP-0-EH, which we define as formable (Fig. 5a). The positive influence of enzymatic hydrolysis compared to refining is likely due to the fiber length reduction and fines content increase. 10 K revolutions of PFI refining had a negligible influence on average fiber length for the reference pulp, while the same amount of refining, combined with enzyme treatment, decreased fiber
length from 1.02 to 0.66 mm. While traditional fiber-based papermaking furnishes are not suitable for roll forming, micro and nanocellulose furnishes seem to be. It is notable that the hydrolyzed furnishes that are roll-formable are only partially degraded and have a significant residual fiber fraction.

It is worth noting that BHKP-5 K and BHKP-10 K were more formable than BHKP-0, even though we do not consider them truly formable. The twin rollers were unable to spread the BHKP-0, while the BHKP-5 K and BHKP-10 K had some spreading over the surface but not enough to make a uniform, smooth sheet. The increased fiber swelling seems to play a substantial if insufficient, role in promoting roll formability.

We compared the dry matter content of materials before and after roll-forming to see if they lost water during forming stage (Table 2). Materials’ solids content increases three times due to the roller nip load and the adsorption of water into the blotting paper. The roll forming process examined here has characteristics of wet pressing since the web is formed and dewatered simultaneously. BHKP-0-EH loses more water than BHKP-10 K-EH. The fine-rich, swollen furnish can hold water more effectively, which lubricates fiber surfaces and helps material flow but negatively impacts dewatering and process economics.

The formation, roughness, waviness, and visual appearance of the roll-formed sheets are compared to filtrated sheets in Table 2 and Fig. 6. In this experiment, the roll-formed sheets are compared to sheets formed at low solids content (0.5%), which represents a state of very good formation. In addition to other factors, fiber flocculation is dependent on solids content, with lower solids content leading to better formation. In roll forming, the mechanism of distributing mass in the plane of the sheet is obviously quite different than in filtration. Even though the roll-formed sheets are formed above 15% solids, these achieve a quite good formation, particularly in

| Material       | Sheet forming method | Specific formation $\sqrt{g/m}$ | Optical surface variation (finescale) [mm] | Optical surface variation (waviness) [mm] | Initial forming solids content [%] | Post-forming solids content [%] |
|----------------|----------------------|---------------------------------|-------------------------------------------|------------------------------------------|----------------------------------|-------------------------------|
| BHKP-0         | Filtration           | 0.46                            | 1.65                                       | 5.63                                     | 0.5                              | N/A                           |
| BHKP-0-EH      | Roll-forming         | 1.11                            | 0.49                                       | 7.35                                     | 15.2                             | 41.5                          |
| BHKP-EH-5 K    | Roll-forming         | 0.80                            | 0.40                                       | 3.22                                     | 16.0                             | 39.7                          |
| BHKP-EH-5 K    | Filtration           | 0.70                            | 0.42                                       | 4.02                                     | 0.5                              | N/A                           |
| BHKP-EH-10 K   | Roll-Forming         | 0.48                            | 0.55                                       | 2.95                                     | 16.0                             | 32.2                          |

Fig. 6 Comparing the visual appearance of a twin roll-formed sheet and filtered one from BHKP-EH-10 K; (a) roll-formed sheet; (b) filtered sheet
the case of the BHKP-10-EH sample, where the fibers are sufficiently degraded to reach the good formation, low roughness, and low waviness. These early results suggest that roll-formed sheets may be produced with excellent structural properties if the furnish characteristics are suitable.

The machine direction (MD) and cross direction (CD) anisotropy were evaluated by examining the sheet tensile properties (Table 3). Interestingly, the rollers orient fibrils in the machine direction, giving a higher elastic modulus than the cross direction. Oriented fibrils also increase the failure stress in MD compared to CD. Anisotropy ratio increases with refining due to more available fibrils in the sheet becoming oriented in the same direction.

We also compared sheets’ tensile properties of vacuum filtrated and twin-rolled methods Table 3. The roll-formed sheets have comparable strength to the filtered sheet in the rolling direction but fail at lower stress in CD.

### Conclusion

Film-forming of nanocellulosic material using twin-rollers at about 15% solid content was investigated. Six MNFC furnishes were produced by combinations of enzymatic treatment and mechanical beating. We were able to form topographically uniform sheets with three out of the six furnishes. Fiber degradation with enzyme treatment was necessary to make our MNFC furnish formable. This result shows that fines content and fiber length play a central role in formability. The rheological assessment shows that fines and shorter fiber length decrease the material’s loss and storage modulus and make it easier to deform. In addition, refining the material after enzymatic treatment improved the formability, formation, and strength properties. The roll forming process orient fibrils in the rolling direction and causes an anisotropic structure. The roll-formed sheet has comparable tensile properties and formation to sheets formed with filtration. Twin-roll forming seems a promising industrially scalable technique to produce nanocellulosic products.

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### Author contributions

E.S.Z. has done the main job of this study. Other authors had sufficient contribution to the concept design, material preparation, data collection, data analysis and writing. All authors read, revised, and approved the final manuscript.

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### Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

### Declarations

#### Competing interests

The authors declare no competing interests.

#### Consent for publication

I, Elaheh Sharifi Zamani, give my consent for the publication of identifiable details, which include photographs and details within the text (“Material”) to be published in the “Cellulose” Journal and Article.

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| Material     | Sheet forming method | MD/CD Elastic modulus [Pa/Pa] | Failure stress (MD) [MPa] | Failure Stress (CD) [MPa] |
|--------------|----------------------|-------------------------------|--------------------------|--------------------------|
| BHKP-0       | Filtration           | N/A                           | 11*                      | N/A                      |
| BHKP-0-EH    | Roll-forming         | 1.26                          | 45                       | 37                       |
| BHKP-EH-5 K  | Roll-forming         | 1.36                          | 60                       | 40                       |
| BHKP-EH-5 K  | Filtration           | N/A                           | 60*                      | N/A                      |
| BHKP-EH-10 K | Roll-forming         | 1.63                          | 59                       | 43                       |

*Isotropic sheets
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