Cyclic Pressing as a Viable Approach for Dewatering and Controlling Shrinkage of Micro-Nanofabricated Cellulose Films

Elaheh Sharifi Zamani, a,* Hamidreza Ahadian, b and Thaddeus Maloney a,*

*Corresponding authors: elaheh.sharifizamani@aalto.fi; thaddeus.maloney@aalto.fi

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GRAPHICAL ABSTRACT
Cyclic Pressing as a Viable Approach for Dewatering and Controlling Shrinkage of Micro-Nanofabricated Cellulose Films

Elaheh Sharifi Zamani, a,* Hamidreza Ahadian, b and Thaddeus Maloney, a,*

Cellulose films, predominantly consisting of micro-nanocelluloses, are a new type of product with interesting properties for functional packaging applications. However, the potentially scalable production methodology has not yet been elucidated. Poor dewatering and high web shrinkage are issues that need solutions beyond what is available in conventional paper production. This research investigates a cyclic pressing method that shows potential in cellulose film consolidation. Cyclic pressing allows the MNFC films to be dewatered to about 90% solids while yielding a smooth, flat product. The results show no inherent physical limits for mechanical dewatering these high swelling webs, even at very high solids. Cyclic pressing allows controlled restraint during consolidation, which could be adjusted in an industrial setup to produce even films with desirable product characteristics.

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Keywords: Capillary pressing; Consolidation; Film; Microfibrillated cellulose; Nanopaper; Shrinkage; Wet pressing; Dewatering

Contact information: a: Department of Bioproducts and Biosystems, School of Chemical Engineering, Aalto University, Espoo, Finland; b: VTT Technical Research Centre of Finland Ltd., Espoo, Finland; *Corresponding authors: elaheh.sharifizamani@aalto.fi; thaddeus.maloney@aalto.fi

INTRODUCTION

Cellulose films containing a significant fraction of nanomaterials, commonly including micro- and nanofibrillated cellulose (MNFC), are often called nanopapers. The use of fine-scale materials (sub-100 nm in width) allows for engineering higher functional properties, particularly in producing films and relatively dense webs. There are potentially several cellulose film products of interest, such as substrates for functional printing or bio-based barrier films, to replace less sustainable plastic alternatives (Johansson et al. 2012; Hubbe et al. 2017; Tarrés et al. 2018).

However, the challenging rheology and high-water binding of most MNFC furnishes complicate the large-scale production of nanopapers with current paper machine designs. In a previous study (Sharifi Zamani et al. 2022), the authors explored an unconventional forming approach where a furnish with suitable rheological properties was spread between two hard rollers. Roll forming is commonly employed in food, plastics, and metal industries but is relatively unknown in bio-based industries. While roll-forming appears to be a viable option for MNFC films, the technology to consolidate and dewater the web after forming is still lacking (Kumar and Jaiswal 2022).
Compared to traditional papers, nanopapers encounter specific challenges during the consolidation stage. Firstly, the considerable swelling of the furnish hinders water removal, leading to a reduction in production rates and an increase in overall energy consumption. A second issue is the substantial shrinkage of the web, which directly correlates to the furnish components’ swelling (Wahlström 2004; Manninen et al. 2011; Uesaka 2013; Vishtal and Retulainen 2014; Ketola et al. 2019). While the water retention value (WRV) for macroscopic pulp fibers typically falls within the range of 1 to 3 g water/g solid, (Maloney et al. 1998), MNFC is usually in the range of 3 to 50 g/g (Maloney 2015; Ankerfors et al. 2017). The heightened swelling imposes significant internal stresses during drying, contributing to the dimensional instability of MNFC films (Manninen et al. 2011; Ketola et al. 2019).

In producing paper, machine direction draws, and cross-direction web restrictions control axial shrinkage, resulting in anisotropic tensile properties and planar deviations. While the current technology for managing shrinkage is not perfect, it is mainly adequate for today’s paper and board products (Alzweighi et al. 2021). However, it is a common experience for nanopapers that shrinkage control is challenging, even in lab-scale testing (González et al. 2014; Sinquefield et al. 2020). If the web is allowed to dry freely, nanopaper is cockled, curled, and unusable in most applications. In contrast, fully restraining the nanopaper web can lead to the development of sufficient strain to fracture the substrate. Similarly, fully restrained nanopapers may exhibit a very high modulus, imparting a brittle character. Neither alternative proves satisfactory for full-scale production; shrinkage control should be tailored to meet specific product specifications.

In wet-pressing of ordinary papers, the compression of the web between the roll and felt generates a hydraulic pressure that will dewater the web, usually up to a solids content of 45 to 50% (Paulapuro 2001). The combination of structural pressure acting on the solid phase and hydraulic pressure causes water to move from the web into the felt. The capillary structure of ordinary press felts is relatively coarse compared to the web, favouring water movement back into the web. This resulting nip and post-nip rewetting lowers the final solids content compared to the maximum solids obtained around mid-nip. If the press felt is replaced with an unsaturated fine capillary substrate, then water will also move from the web to the substrate by capillary transport. Rewetting can be eliminated or reduced, and the web solids can be driven to very high levels if a moisture gradient is maintained between the web and the substrate. In this study, the authors use a dry blotting paper, which is changed between pressing events. The smallest capillaries of this substrate are nanoscale pores within the cell wall, which are opened as the fibres swell when contacted with water. A scale-up solution will obviously entail using fine capillary substrates more suitable for industrial use.

In this paper, the authors explore a sequential pressing approach designed to address the dewatering and shrinkage challenges in the web effectively. This method enables the production of a smooth cellulosic film with minimal reliance on thermal energy, tailored explicitly for nanopaper applications. The films undergo consolidation, transitioning from 40% to 90% solids after forming high solids content. The primary objective of this study is to demonstrate on a laboratory scale the removal of water up to 90% solids with a cyclic, capillary pressing approach. This research builds on the authors’ previous study of roll-formed nanopapers (Sharifi Zamani et al. 2022) and delves further into the high-consistency production of MNFC films.
EXPERIMENTAL

Material

The authors’ previous study (Sharifi Zamani et al. 2022) concluded that the fiber structure should be degraded and fibrillated to achieve good roll-forming rheology. The furnishes in this study followed this general characteristic.

A Finnish mill provided once-dried, bleached Birch hardwood kraft pulp (BHKP-0). The fibers were broken down with enzymatic hydrolysis as follows. After overnight soaking and disintegration, the pulp pH was adjusted to 5.7. The solids contents were adjusted to 20% by vacuum filtration. The pulp (BHKP-0-EH) was hydrolyzed for 2 h at 55 °C, 20% solids, using 1.5 mg/g pulp of endoglucanase enzyme (ECOPULP R, AB Enzymes, Rajamäki, Finland). Hydrolysis was done with constant mixing in a Kenwood chef food processor. After hydrolysis, the material was immediately placed in an ice bath and later kept at +5 °C to deactivate the enzyme. All solids contents (consistencies) are reported in % as the mass of 105 °C - dried solids / (mass of solids + water).

After enzyme treatment, 30 g of BHKP-0-EH was refined in a PFI mill at 15% solids for 5000 or 10000 revolutions to produce BHKP-EH-5K and BHKP-EH-10K, respectively. Thus, a family of MNFC furnishes with differing degrees of fibrillation and swelling was produced.

Methods

The pulp morphological properties were determined with a fiber analyzer (Fiber Image Analyser FS 5; Valmet, Helsinki, Finland). The degree of polymerization (DP) was calculated from the pulp viscosity (SCAN-CM 15:99 (1999)) using the Evans and Wallis equation (SCAN 15:88 (1988)). The water retention value was measured according to SCAN-C 62:00 (2000).

The MNFC films (nanopaper samples) were formed by a rolling machine (Atlas 150; Marcato, Campodarsego (PD), Italy) at 15% consistency using the twin-roll forming method described in (Sharifi Zamani et al. 2022). The films were produced at a constant thickness (375 µm) rather than at a constant grammage. The solids content after forming was in the range of 30 to 40%. A considerable amount of water is expelled from the web in the roll-forming nip.

After the forming stage, the nanopaper samples underwent iterative pressing using an L&W SE 040 hydraulic press (L&W, ABB, Helsinki, Finland). This involved pressing for 1 minute at 253 kPa, sandwiched between two air-dried sheets of blotting paper (250 g/m², Ahlstrom, Helsinki, Finland). The blotting paper had a specific weight of 0.47 g/m², an air permeability of 157 mL/min·cm², and a fine-scale optical surface variation of 2.24 mm. This cycle was repeated, replacing the blotting papers with dry ones each time, until the samples reached a consistency of approximately 90%. The films’ topography was measured for dried sheets using an optical surface roughness device (L&W OptiTopo; ABB, Helsinki, Finland). MATLAB software (Software, The MathWorks, Inc. MATLAB, v.2023, Natick, MA, USA) was employed to generate optical surface variation graphs based on the results obtained from the device mentioned above (L&W OptiTopo). Specific formation and air permeability were measured relatively by use of a Beta formation tester (Ambertec, Espoo, Finland) and Bendtsen air permeability tester (L&W; ABB, Helsinki, Finland).
The samples’ shrinkage and density were assessed through area measurement via image scanning and weighing after each pressing stage. Thickness measurements were conducted in a similar manner using an anvil caliper gauge. To monitor stress development throughout the drying process, the authors positioned the samples in the holders of a universal testing device (MTS 400/M; MTS Systems., Créteil Cedex, France), while allowing them to dry under ambient conditions with 50% relative humidity and at a temperature of 23 °C.

RESULTS AND DISCUSSION

The untreated pulp and three hydrolyzed MNFC samples (Fig. 1), fibrillated to different extents, are shown in Table 1. The BHKP is a pulp suspension and cannot be roll-formed into sheets because it lacks suitable rheological characteristics, as discussed in (Sharifi Zamani et al. 2022). The other three samples are sufficiently broken down, fibrillated, and swollen that these furnishes have the plasticity required for roll-forming.

![Images](https://bioresources.cnr.ncsu.edu)

**Fig. 1.** (a to c) Light microscopy of BHKP, BHKP-0-EH, and BHKP-EH-10K; (d to f) SEM images of dried sheets cross-section made from BHKP, BHKP-0-EH, and BHKP-EH-10K after drying

The roll-formed sheets from the hydrolyzed samples were pressed at room temperature for one-minute intervals at 253 kPa, each time between two pieces of blotting paper, as detailed in the ‘Methods’ section. This cyclic pressing was repeated until no significant additional water could be removed, indicating that the sheets had reached their maximum dryness. The initial solids content of the sheets, which ranged from 30% to 40%, reflects the output from the roll-forming operation prior to pressing. Figure 2 illustrates a plateau in solids content in the 85% to 90% range, which significantly differs from industrial pressing, typically yielding around 50% solids and even less for slow-draining furnishes.
Table 1. Average Fiber Properties, Degree of Polymerization, and Water Retention Value (Expressed as g Water/g Solids and Solids Content)

<table>
<thead>
<tr>
<th>Materials</th>
<th>Fiber Length (mm)</th>
<th>Fines Content (%)</th>
<th>Fiber Width (μm)</th>
<th>Curl (%)</th>
<th>Fibrillation (%)</th>
<th>WRV² (g/g)</th>
<th>WRV² Solids Content (%)</th>
<th>DP³ (Number)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BHKP</td>
<td>0.860</td>
<td>15</td>
<td>20.0</td>
<td>20.8</td>
<td>0.12</td>
<td>1.25</td>
<td>44.4</td>
<td>3200</td>
</tr>
<tr>
<td>BHKP-0-EH</td>
<td>0.060</td>
<td>70</td>
<td>4.2</td>
<td>19.6</td>
<td>0.67</td>
<td>2.81</td>
<td>26.2</td>
<td>1160</td>
</tr>
<tr>
<td>BHKP-EH-5K</td>
<td>0.041</td>
<td>83</td>
<td>3.7</td>
<td>13.6</td>
<td>0.69</td>
<td>2.93</td>
<td>25.4</td>
<td>965</td>
</tr>
<tr>
<td>BHKP-EH-10K</td>
<td>0.040</td>
<td>85</td>
<td>3.9</td>
<td>12.2</td>
<td>1.13</td>
<td>3.39</td>
<td>22.8</td>
<td>953</td>
</tr>
</tbody>
</table>

¹Length-weight average; ²Water retention values; ³Degree of polymerization

The remaining water must be removed using thermal energy, incurring considerable expenses. For this reason, the limiting solids content in wet pressing is of great interest to paper manufacturers. Previous research has established correlations between various measures of fiber swelling and the solids content after pressing (Maloney et al. 1998; Paulapuro 2001; Hubbe et al. 2020). Wahlström (1990) has concluded that this is due to the flow limitations of water removal from the cell wall pores. Still, others have suggested (Carlsson et al. 1977; Maloney et al. 1997) that water in the larger cell wall pores can be removed by pressing under more extreme conditions, such as those used here, but water trapped in micropores (after about 60% solids) cannot be pressed out. The issue has been examined recently by Kerekes and McDonald (2020).

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Fig. 2. Change of solid contents during the mechanical pressing under 253 kPa pressure. Stage zero shows solids contents before pressing.

In the present experiment, essentially all the water in the furnish at the beginning of the pressing sequence was intra-particle water. This can be seen by examining Table 1, which shows the solids content before pressing was well below the equivalent WRV. Above a 70 to 80% solids content for saturated kraft pulps, all water is bound-water, with limited mobility and elevated evaporation enthalpy (Maloney et al. 1998; Caurie 2011). This shows there is not really any physical limitation on the mechanical removal of water.
Rather, product quality and dewatering efficiency impose restrictions on press dewatering. In regular pressing, the capillary structure of the press felt is relatively coarse compared to the web, and capillary water transport out of the web is, therefore, limited. To address this, the authors’ approach involves using a substrate with a fine capillary structure within the cell walls of kraft fibers, thus enhancing capillary forces to facilitate water removal.

One contributing factor to achieving a high solids content in cyclic pressing is eliminating nip rewetting. In dynamic pressing, using conventional press felts, the solids content can peak at approximately 60% during the mid-nip stage (Rantanen and Maloney 2013; McDonald and Kerekes 2017). However, during the subsequent re-expansion phase, water is drawn back from the press felt into the paper web, causing the solids content to decrease to around 45 to 50% typically. This residual water then needs removal in the dryer section.

However, in this cyclic pressing experiment, the very fine capillaries within the cell wall of the blotting paper fibers prevent water from reabsorbing into the web. After all, the “unfilled” capillaries of the substrate are finer than the paper itself, considering the blotting paper is dry (95% solids) at the beginning of the pressing cycle. So the capillaries within the cell wall do not exist in the dry state and are formed as the cellulose is hydrated and swollen. By the last pressing stage, there is no longer a sufficient difference in nanopaper/substrate solids content to drive water transport, and the paper solids content plateaus.

While similar effects have been employed to develop capillary press felts (Wilder 1967; Iliev et al. 2013) with limited success on an industrial scale, there’s potential value in reconsidering this strategy in light of advancements in industrial textile fibers over recent years.

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**Fig. 3.** Comparison of BHKP-EH-5K film drying under full restraint (a), freely dried (b), and the corresponding stress-strain curve created passively during drying, contrasted with a BHKP-0 reference (c). The material exhibits splitting or wrinkling behavior, depending on the applied restraint.
Figure 3 effectively illustrates the challenges associated with controlling shrinkage in nanopapers. Specifically, Fig. 3-a demonstrates that it tends to split when the sheet is fully restrained. This splitting is attributed to the high internal stress that develops, a phenomenon further detailed in Fig. 3-c for both BHKP and BHKP-EH-5K samples. Conversely, considerable planar deviations occur if the sheet is left unrestrained, as depicted in Fig. 3-b. While this issue is not exclusive to nanopaper and can occur in normal paper production, it is markedly intensified in the case of MNFC due to its high swelling and shrinkage potential.

To further the understanding of the consolidation process in the capillary pressing of nanopapers, the authors measured the dimensional changes in the web after each pressing cycle. The apparent sheet thickness is shown in Fig. 4, and the normalized area shrinkage in Fig. 5.

![Figure 4](image1.png)

**Fig. 4.** Evolution of sheet thickness changes with solid content during cyclic capillary pressing, measured at 1-min intervals

![Figure 5](image2.png)

**Fig. 5.** Variations in area shrinkage relative to solids content in cyclic capillary pressing were measured at 1-min intervals (results at 100 solids content are measured after drying in the oven)

In each pressing stage, water is transported from the web into the blotting paper by a combination of hydraulic pressure and capillary suction. In a saturated web, the thickness change is proportional to the water amount exiting the sheet. Thus, the thickness decreases
during the pressing event. For the relatively swollen and pliable MNFC furnish, it is not expected that there will be much elastic strain recovery after the press; i.e. spring-back is minimal. Therefore, as Fig. 4 shows, the thickness decreases monotonically over the pressing cycles.

During a single pressing stage, the web is completely restrained by the pressure exerted through the blotting papers. Thus, no area shrinkage is expected during a single stage. However, internal stress builds up in the sheet as it is dewatered, and this is released after pressing when the sheet is separated from the used blotter and placed between fresh blotting paper. Figure 5 shows there was considerable shrinkage after each pressing stage. Over the entire pressing sequence, the area shrinkage was 15 to 20%, with the more swollen samples in the upper end of this range. The MNFC furnish is distinct from traditional fiber furnish in that they are highly plastic and deformable. Thus, any planar deformations that occur in between each pressing event can be smoothened in the next pressing stage. The net result is a sheet that has shrunk considerably but is still smooth and even. The visual appearance of the sheet is shown in Fig. 6, and the topology is compared to a freely dried sheet in Fig. 7.

![Fig. 6. Visual representation of finalized dry nanopapers derived from BHKP-EH-5K samples, showcasing the sequential process of their formation at 15% consistency, mechanical pressing to over 90% solids, subsequent oven drying, and equilibration to achieve a stable 95% solids content.](image1)

![Fig. 7. A comparison of surface topography between two sheets made of BHKP-EH-5K material reveals the superior smoothness achieved through the cyclic capillary pressing method (a) in contrast to unrestrained air drying (b).](image2)
Cyclic capillary pressing can be viewed as a process that dewatering the sheet. The area shrinkage for the BHKP-EH-5K unrestrained samples was 37.4% (shrinkage potential). The same sample, after cyclic capillary pressing, shrank 24.5%. Thus, the sample reached 65.5% of its potential level of shrinkage. In an industrial capillary press setup, control of press geometry, draws, residence times, and other design and operating variables could be used to control the shrinkage. This has considerable implications for tensile properties, moisture reactivity, and other properties that depend heavily on the shrinkage and development of internal sheet stresses.

Future research should focus on clarifying the grammage control in roll-forming, considering both the forming and consolidation phases. One complicating factor that needs to be considered is illustrated in Fig. 8. Unlike traditional paper pressing, the grammage notably increases with web solids. This phenomenon occurs due to the MD/CD shrinkage, which increases the mass concentration per unit area.

Fig. 8. Grammage evolution with increasing solids content during the dewatering process using cyclic capillary pressing, measured at 1-min intervals at each stage, emphasizing the significant increase in grammage with web solids during pressing (the final grammage at 100% is measured after putting samples in the oven).

The pore structure greatly influences the properties of the finished sheet. For example, light scattering and gas or liquid transport properties are determined mainly by interparticle spaces. In Fig. 9, the apparent solids density of the sheet is shown. This is the apparent density with water excluded. In Fig. 10, the air volume in the sheet is shown, assuming a specific gravity of water of 1 and dry MNFC of 1.55.

Interestingly, despite their different swelling characteristics, all three samples arrive at about the same sheet density, 1150 to 1200 kg/m³. In Fig. 10, the pathway to arrive at the final density/porosity is different. The lower swelling furnishes allow air imbibition earlier but finally consolidate to a similar density.
Fig. 9. The apparent density of the sheet versus solids content during cyclic capillary pressing was measured at 1-min intervals at each stage.

Fig. 10. Air volume fraction in sheets during consolidation using cyclic capillary pressing, measured at 1-min intervals at each stage. The air volume in the sheet is shown assuming a specific gravity of water of 1 and dry MNFC of 1.55.

CONCLUSIONS

The authors’ investigation into cyclic pressing shows its potential as a consolidation method for micro-nanocellulose film formed under high-consistency conditions. The sequential pressing approach effectively tackles challenges related to consolidating highly swollen and poorly dewatering furnishes.
1. This experiment achieved 90% web solids without using thermal energy. This shows that in addition to the hydraulic pressure used in ordinary industrial wet pressing, capillary movement of water between the web and the substrate may be utilized to increase dewatering. In this study, water in the cell wall pores, including bound water in contact with the cellulose surfaces, was removed by purely mechanical means.

2. Cyclic capillary pressing provides a way to control the shrinkage and consolidation in a different way than conventional pressing/drying. In this study, the roll-formed cellulose films could be dewatered to a smooth and even web. This indicates that this approach may be useful for the industrial production of papers from nanocellulose or other highly swollen materials, even though the scale-up of the cyclic capillary approach requires further investigation of potential capillary pressing felts.

3. The cyclic capillary pressing method represents an innovative blend of restrained and free drying processes. The cellulose films are effectively restrained and smoothed during the pressing stages, contributing to a controlled consolidation. However, between these stages, the films undergo shrinkage. This results in the films reaching 66% of their potential shrinkage under free drying conditions. This finding is pivotal, highlighting the distinctive balance between restraint and natural shrinkage in the cellulose film drying process to ensure smoothness.

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Statement and Declarations
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Competing Interests
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Author Contributions
Elaheh Sharifi Zamani has done the main job of this study. Other authors contributed sufficiently to the concept design, material preparation, data collection, analysis, and writing. All authors read, revised, and approved the final manuscript.

Conflict of Interests
The authors report no conflicts of interest. The authors alone are responsible for the content and writing of the paper.
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