Dewatering of Cellulose Nanofibrils using Ultrasound

Udita Ringania

Georgia Institute of Technology

Joseph Harrison

Georgia Institute of Technology

Robert J. Moon

USDA Forest Service Region 8: USDA Forest Service Southern Region

M. Saad Bhamla (✉ saadb@chbe.gatech.edu)

Georgia Institute of Technology <https://orcid.org/0000-0002-9788-9920>

Research Article

Keywords: dewatering, ultrasonic dewatering, cellulose nanofibrils, energy-efficient, continuous, platform

Posted Date: July 29th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-756325/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Dewatering of Cellulose Nanofibrils using Ultrasound

Udita Ringania¹, Joseph Harrison¹, Robert J. Moon² and M. Saad Bhamla^{1*}

1 Chemical and Biomolecular Engineering, Georgia Tech, Atlanta, GA 30332

2 The Forest Products Laboratory, U.S. Forest Service, Madison, WI 53726

Robert J. Moon ORCID iD: 0000-0001-9526-0953

**Corresponding author: saadb@chbe.gatech.edu*

Abstract:

Although cellulose nanomaterials have promising properties and performance in a wide application space, one hinderance to their wide scale industrial application has been associated with their economics of dewatering and drying and the ability to redisperse them back into suspension without introducing agglomerates or lose of yield. The present work investigates the dewatering of aqueous suspensions of cellulose nanofibrils (CNF) using ultrasound as a potentially low-cost, non-thermal, and scalable alternative to traditional heat-based drying methods such as spray drying. Specifically, we use vibrating mesh transducers to develop a direct-contact mode ultrasonic dewatering platform to remove water from CNF suspensions in a continuous manner. We demonstrate that the degree of dewatering is modulated by the number of transducers, their spatial configuration, and the flow rate of the CNF suspension. Water removal of up to 72 wt.% is achieved, corresponding to a final CNF concentration of 11 wt.% in 30 minutes using a two-transducer configuration. To evaluate the redispersibility of the dewatered CNF material, we use a microscopic analysis to quantify the morphology of the redispersed CNF suspension. By developing a custom software pipeline to automate image analysis, we compare the histograms of the dimensions (length, width) of the redispersed dewatered fibrils with the original CNF samples and observe no significant difference, suggesting that no agglomeration is induced due to ultrasonic dewatering. We estimate that this ultrasound dewatering technique is also energy-efficient, consuming up to 36% less energy than the enthalpy of evaporation per kilogram of water. Together with the inexpensive cost of transducers (<\$1), the potential for scaling up in parallel flow configurations, and excellent redispersion of the dewatered CNF, our work offers a proof-of-

30 concept of a sustainable CNF dewatering system, that addresses the shortcomings of existing
31 techniques.

32 Keywords: dewatering, ultrasonic dewatering, cellulose nanofibrils, energy-efficient, continuous
33 platform

34

Introduction:

Cellulose nanofibrils (CNF) are nanosized cellulose particles (20-100 nm in diameter, microns in length) that have a unique combination of characteristics: renewable, sustainable, biodegradable, low toxicity, high aspect ratio, high surface area, high mechanical properties, transparency, self-assembly, industrial scale production, etc, which gives them utility across a wide range of applications in composite materials, coatings, membranes, biomedical, rheology modifiers, etc. (Moon et al. 2011; Rebouillat and Pla 2013; Geng et al. 2018; Patel et al. 2019; Čolić et al. 2020; Nigmatullin et al. 2020). Although CNF presents many promising properties, one hinderance to their wide scale industrial application is associated with the economics of their processing and handling. Due to their hydrophilic nature, CNF are processed in aqueous form with the final suspension of about 90-97 wt% (Rebouillat and Pla 2013), leading to high shipping costs. This cost can be circumvented by shipping a dewatered (i.e., concentrated) or dried form of these materials. However, drying is a cost intensive process (Miller et al. 2015), and can cause permanent changes to CNF nano-dimensional particle morphology, and thus performance. As the water is removed from the system, hydroxyl groups of neighbouring particles get closer resulting in strong hydrogen bonds and triggering agglomeration. Redispersion of these dried particles into stable suspensions that are comparable to that of the original particle dimensions is an ongoing challenge due to the high energy requirements to break these hydrogen bonds(Beck et al. 2012; Missoum et al. 2012; Beuguel et al. 2018). Traditional drying methods such as oven drying, spray drying, freeze drying, suffer from loss of nanoscale dimension due to agglomeration of fibrils associated with inter-particle hydrogen bonding(Peng et al. 2012a). Supercritical drying(Zimmermann et al. 2016) and solvent replacement drying(Hanif et al. 2018) are two methods that has been observed to retain the nanoscale dimensions. However, high cost of operations and complexities associated with scaling up has limited their wide scale application. The advantages and disadvantages of different drying methods used for cellulose nanomaterials (CNMs) is summarized in Table 1.

Drying Method	Advantages	Disadvantages	Ref
Air Drying	<ul style="list-style-type: none">Low costLow energy requirements	<ul style="list-style-type: none">High AgglomerationPoor redispersibility (irreversible)Loss of nanoscale dimensions	(Peng et al. 2012b; Penttila et al. 2012)
Oven Drying	<ul style="list-style-type: none">Economical and scalableEstablished industrial technology	<ul style="list-style-type: none">High AgglomerationPoor redispersibilityLoss of nanoscale dimensions	(Quiévy et al. 2010; Peng et al. 2012a; Zimmermann et al. 2016)
Freeze Drying/ Lyophilization	<ul style="list-style-type: none">Nanoscale dimensions maintained in parts of the fiberRelatively Stable ProcessWell established and scalable process	<ul style="list-style-type: none">Agglomeration to a certain extent can occurSlow processHigh energy requirement	(Peng et al. 2012b, 2013; Han et al. 2013; Zimmermann et al. 2016)
Spray drying	<ul style="list-style-type: none">Scalable and fast processRe-dispersible to a certain extent (in micro-nanosized particle range)Continuous process	<ul style="list-style-type: none">High installation and maintenance costEnergy intensiveSome extent of agglomerationLoss of nanoscale dimension (particle size ranges from nano to micrometers)	(Quiévy et al. 2010; Peng et al. 2012b; Park et al. 2017; Al Zaitone 2019)
Supercritical Extraction *	<ul style="list-style-type: none">Efficient DryingMaintains the nanoscale	<ul style="list-style-type: none">Expensive methodComplex processHard to scale upNot applicable to cellulose nanocrystals (CNC)	(Peng et al. 2012a, 2013; Peng and Gardner 2013; Zimmermann et al. 2016)
Solvent Replacement drying (t-BuOH)	<ul style="list-style-type: none">Accelerated drying rates (~3 time for oven drying and~ 3-20 times for freeze drying) than drying rates for original solutionReduced shrinkage during drying processEasy redispersionMaintains the nanoscale structure	<ul style="list-style-type: none">100% solvent replacement not achievedPresence of t-BuOH on the cellulose particles after drying	(Hanif et al. 2018)

*Applicable to CNF only

64 Apart from drying, which aims at removing all the water from the suspension, a partial drying
65 approach that concentrates a suspension, referred to as 'dewatering', has also been employed for
66 CNMs. One advantage of dewatering is that it can avoid CNM agglomeration challenges.
67 Examples of a few common dewatering techniques, summarized in Table 2, include centrifugation,
68 filtration, shear-induced dewatering and pressing, with variable success in CNMs. Centrifugation

and filtration are widely used laboratory scale processes; however, scale-up issues inhibit their integration in industries. A main disadvantage of filtration is the long time required for dewatering (1-4hrs/100ml suspension)(Iwamoto et al. 2005; Sehaqui et al. 2010). Electro-assisted filtration for microfibrillated cellulose (MFC) helps tackle this issue (10-30 mins/100 ml of diluted suspension) but only through an increase in the overall energy consumption(Wetterling et al. 2017). Electroosmotic filtration for CNC has been demonstrated to be energy efficient as compared to the thermal methods, however, higher dewatering rates increases these energy requirements and their applicability to CNF remains unknown (Wetterling et al. 2018). Other methods using filtration have employed additives such as salts(Sim et al. 2015) and wood pieces(Amini et al. 2019) to assist in phase separation for CNFs, however their recovery is either not feasible or economically unviable. Application of ultralow shear stress to CNF and MFCs slurry is another approach through which phase separation and dewatering has been achieved(Dimic-Misic et al. 2013, 2017, 2018). However, the approach requires long durational stress application and also results in auto flocculation of the fibers. Finally, subjecting MFCs to high pressure pressing also helps dewater the slurry(Rantanen and Maloney 2015). This is at times assisted with heat(Clayton et al. 2006), or using a plate and frame type pressing mechanism. Thus, with the availability of the present dewatering techniques (Table 2), a low cost, scalable, energy-efficient method is still lacking for dewatering CNMs.

Table 2: Dewatering methods for CNMs

Dewatering Method	Advantages	Disadvantages	Ref
Centrifugation	<ul style="list-style-type: none"> • Easy availability 	<ul style="list-style-type: none"> • Difficult to scale up • High energy requirement • Batch process 	(Astorsdotter 2017; Amini et al. 2019; Zhai et al. 2020)
Filtration*	<ul style="list-style-type: none"> • Easy availability* 	<ul style="list-style-type: none"> • Long dewatering time*,*** 	(Iwamoto et al. 2005; Sehaqui et al. 2010; Wetterling et al. 2017, 2018; Sethi et al. 2018)
Electro-assisted filtration**	<ul style="list-style-type: none"> • Faster dewatering times** 	<ul style="list-style-type: none"> • Difficulty to scale up • Higher energy requirements • Batch process 	
Electro-osmotic filtration***			
Shear Induced dewatering	<ul style="list-style-type: none"> • High dewatering efficiency 	<ul style="list-style-type: none"> • Induces agglomeration • Long dewatering times • Batch process 	(Dimic-Misic et al. 2013, 2017, 2018)
Pressing	<ul style="list-style-type: none"> • High dewatering efficiency • Can be redispersed 	<ul style="list-style-type: none"> • High pressure requirements 	(Clayton et al. 2006; Rantanen and Maloney 2015)

		<ul style="list-style-type: none"> • Applicable mainly for longer fiber dimensions (microfibrillated cellulose) • Batch process
Ultrasonic dewatering	<ul style="list-style-type: none"> • Continuous process • Energy-efficient • Easily Scalable • Fast dewatering times 	<ul style="list-style-type: none"> • Needs modification for This study application to CNC

89

90 The hypothesis of this study is that ultrasonic approaches can be applied to the dewatering of CNFs
91 suspensions. To the authors knowledge ultrasound approaches have not been reported for the
92 dewatering of CNFs suspension, however, ultrasound has been used as a low heat drying method
93 for food, fabric and variety of other solid materials with defined structure (Boucher 1959a; García-
94 Pérez et al. 2015; Musielak et al. 2016; Peng et al. 2017a; Sabarez et al. 2019). The first acoustic
95 drying, as performed by Boucher(Boucher 1959b, a) and Greguss(Greguss 1963), used
96 mechanically generated sound in the sonic and ultrasonic range to dry cotton fiber and a wide
97 variety of other materials. The extremely small 1 to 2 °C increase in temperature during the drying
98 process suggests applicability to drying of heat-sensitive materials and high value-target. However,
99 poor energy efficiency and high sound levels due to the air-jet sirens used decades ago hindered
100 the potential applications of mechanically generated acoustic drying. Nevertheless, increasing
101 availability of modern piezoelectric transducers in the recent decades, producing ultrasound energy
102 at significantly lower power inputs, lower audible sounds and with high energy efficiency,
103 rekindled an interest in ultrasonic acoustic drying. Since then, ultrasonic drying has been
104 successfully demonstrated for a variety of food products with a significant reduction in both drying
105 time and energy used(Gallego-Juárez et al. 2007; García-Pérez et al. 2015; Musielak et al. 2016).
106 Recently, ultrasound has also been implemented in fabric drying showing approximately 10 times
107 reduction in energy use as compared to traditional heat based dryer (Peng et al. 2017a, b).

108 The mode of application of the ultrasound to the sample plays an important role on the final drying
109 efficiency achieved. The two primary modes of application are (i) non-contact mode (or air-borne
110 ultrasound) where the ultrasound is applied to the sample through a medium, usually air, and (ii)
111 direct-contact mode where the ultrasound is directly applied to the sample. Air-borne ultrasound

has been observed to assist the convectional air drying but suffers from significant energy attenuation due to mismatch in impedance between air and solid/liquid media. In contrast, direct-contact mode ultrasound ensures a better transfer of energy from the piezoelectric transducer to the solid/liquid in contact (Gallego-Juárez et al. 1999). For highly shrinkable food products during ultrasonic drying, applying a static pressure to the sample ensured constant contact during the entire process effectively reducing the drying time by 65-70% as compared to heated air drying (Gallego-Juárez et al. 2007). More recently, direct-contact mode has been used for dewatering knitted fabrics (Peng et al. 2017a). The low cost and easy availability of piezoelectric transducers used to produce ultrasound motivates this current work to explore the possibility for designing a low-cost direct-contact mode dewatering platform for CNFs.

In this study, ultrasound as a potential low cost dewatering technique for CNFs suspensions is investigated. A direct contact mode approach is undertaken and the effect of system parameters such as suspension flow rate and transducer configuration on the dewatering rate and amount of water removed is studied. The viability of the ultrasonic dewatered material to be redispersed back in suspension is tested along with potential changes in CNF particle morphology. A quick effective and quantitative image analysis approach is developed for visualizing the fibrils in the solution phase using phase contrast microscopy combined with image analysis via a MATLAB code. Histograms generated thereby are compared with original fibrils to identify any changes in morphology or fibril dimensions as a result of dewatering.

Materials and Methods:

Cellulose Nanofibers

Cellulose nanofibers (CNF) at a concentration of 3 wt.% solids in water (Lot# U-103, 90% fines retained) were procured from University of Maine, Orono, ME, USA (UMaine). These CNF were generated through mechanical fibrillation of wood pulp and are often branched or forked with dimensions about 20-50 nm in width and several microns in length. Measurements of the solid content for the CNF suspension by us confirmed a concentration of 3.1 ± 0.4 wt. % solids.

Vibrating Mesh Ultrasonic Transducer

Ultrasound was produced using a piezoelectric vibrating mesh transducer (VMT) (WHDTS, 20 mm diameter, rated power 1.5-2.5 W, rated voltage 70 V(max) and resonance frequency of 113 kHz $\pm$ 3 kHz) which comprises of a metal mesh attached to a piezoelectric ring at the bottom as shown in **Fig 1**. The vertical vibration of the metal mesh on application of voltage across the piezoelectric ring generates an acoustic pressure difference that pushes water through the tapered pores of the metal mesh generating a cold mist ejected from the back surface of the transducer (as seen in **Fig 1d and 1g**).

Fig 1 Vibrating mesh transducer's (VMT) comprises a metal mesh attached to a piezoelectric ceramic ring at the bottom. (a) top view of VMT and (b) back view of VMT. (c) Schematic of the top view of the VMT depicting the metal mesh at the center whereas (d) is the side view schematic of the VMT with CNF placed on the top surface and water removed from the back surface of the metal mesh. (e) SEM image of the metal mesh with pore size of $\sim 18 \mu\text{m}$ on the top surface. The pores have a tapered geometry with 3-5 μm pore diameter on the back surface. (f) Image of CNF placed on a VMT. (g) Image of a water droplet placed over a VMT actively producing mist from the back end

150 **Ultrasonic dewatering**

151 A direct-contact mode ultrasonic dewatering set-up (Fig 2) was designed such that CNF
152 suspensions can flow over multiple transducers allowing for continuous dewatering. The setup
153 comprises of a syringe pump (kd Scientific 78-0100) to control the flow rate, a 10 ml syringe (inner
154 diameter of the outlet: 1.6 mm), an acrylic transducer stand, VMT, a function generator (RIG01
155 DG 1022 Function/Arbitrary Waveform Generator) and a power amplifier (Krohn-Hite Model
156 7602M) to drive the VMT, a scraper blade pulley system operated via external DC motor and finally
157 a collection box to collect the dewatered CNF. The dewatering that occurs as the CNF suspension
158 flows over the transducers results in a higher viscosity CNF gel-like or paste-like material, thus a
159 scraper blade system is incorporated to assist the gravity driven flow of the CNF material over the
160 transducer plates. The velocity of the blade is maintained at 10% higher than the flow rate of the
161 CNF to achieve a consistent flow over the plate (Online Resource 1: SI-Table 1 and SI-Fig 1). The
162 flow rate of the CNF at the syringe outlet and the transducer configuration are varied to assess
163 their effects on the dewatering rate and the amount of water removed. Four flow rates were studied:
164 60 ml/hr, 30 ml/hr, 15 ml/hr and 10 ml/hr; and two transducer configurations are used: single
165 transducer configuration (1T) and double transducer (2T) configuration (Fig 2 (c)). At least 4 runs
166 are conducted for each flow rate at each transducer configuration with the initial sample size of 5
167 ml each (loaded in a 10 ml syringe and initial weight (W_i) recorded using a weighing machine
168 (METTLER TOLEDO ME0204TE/00)). The transducers are operated at 113-117 kHz and 60
169 V_{peak}. The final dewatered samples are collected in the box at the bottom and its final weight is
170 recorded (W_f). Temperature measurements during the continuous flow experiments indicated ~
171 20°C rise in the suspension temperature over the transducer (see Online Resource 1 SI-Fig 2).
172 This level of temperature rise is inconsequential for our system as it is below the thermal
173 degradation temperature of CNF and does not drastically effect the evaporation rates.

Fig 2 (a) Schematic corresponding to the experimental setup for the continuous mode ultrasonic dewatering of CNF using VMT. The setup consists of transducer stand, VMT, power source to operate the transducers, scraper with external DC motor to aid the downward flow of the CNF, syringe pump (for controlled dispensing of CNF), 10ml syringe and a collection box. Inset represents the schematics of the transducer configurations tested. Single transducer configuration (1T) uses a single VMT for dewatering and double transducer configuration (2T) uses two VMT stacked vertically one above the other. (b) Image of the experimental setup with the same components

Characterization

CNF Concentration Calculation

The initial concentration of the CNF in the suspension is ~3wt.% (C_i). This is used to calculate the initial weight of the CNF solids (F_i in mg) in the sample suspension of initial weight W_i (in mg)

$$F_i = \frac{3}{100} \times W_i \quad 1$$

The percent of water removed from the system is calculated as follows:

$$\% \text{ water removed} = \frac{W_i - W_f}{W_i} \times 100 \quad 2$$

Here, W_f is the final weight of the CNF suspension collected after dewatering. The final concentration of the CNF (C_f) is calculated by assuming that CNF solid content in the suspension remains constant and that no CNF is lost during dewatering. Hence final CNF solids weight, $F_f = F_i$

$$C_f \text{ wt. \%} = \frac{F_f}{W_f} \times 100 \quad 3$$

For example, let us start with initial CNF suspension weight of 100 mg with 3 mg of CNF solids. Let us assume that the final weight of the dewatered CNF suspension is 30 mg. Hence, percent water removed = $((100-30)/100) * 100 = 70\%$. The final CNF concentration = $((3/30)*100) = 10 \text{ wt. \%}$.

Dewatering Rate

Dewatering rate, in this study, is defined as the amount of water removed from the system in the time taken for processing 5 ml of CNF suspension at a given flow rate. It is calculated using the following equation:

$$\text{Dewatering rate (mg/min)} = \frac{W_i - W_f}{t} \quad 4$$

Here, t is the time required for processing 5 ml of CNF suspension and is equal to 5 min, 10 min, 20 min and 30 min for 60 ml/hr, 30 ml/hr, 15 ml/hr and 10 ml/hr flow rates, respectively.

Redispersion

The dewatered samples are diluted using DI water to 1 wt.%, 0.1 wt.% and 0.01 wt.% of CNF solids and redispersed via a vortex mixer (VWR Analog Vortex Mixer No. 10153-838) at speed 7 for 30s. The original CNF sample with initial 3 wt. % is also diluted using DI water to the above weight percentages and mixed using vortex mixer to form a homogeneous suspension and act as a reference sample.

206 *Microscope Imaging*

207 Phase contrast microscopy images of the fibrils for redispersed dewatered samples and diluted
208 original CNF sample each at 0.01 wt.% are taken using an optical microscope (Nikon ECLIPSE
209 Ti2-U). Phase contrast microscopy is a contrast enhancing optical technique that works by
210 converting the phase shifts in light passing through a specimen into changes in brightness in the
211 image. This technique can be directly applied to CNF suspensions thereby providing a fast and
212 easy characterisation tool for analysing fibril distribution and dimensions. The specimens for
213 imaging are prepared by placing a 15 μ l droplet of the 0.01 wt% sample in a channel (created by
214 placing double sided tape (thickness: 88.9 μ m) $\sim$ 1.5 cm apart) on a glass slide (VWR
215 VistaVision™ Cat.No. 16004-430 (76.2x25.4x1) mm) and covering using a glass cover slip
216 (Thermo Scientific™ Gold Seal™ Cover Glass (25x25) mm No1). Images are taken using the 20x
217 objective lens and the phase 1 contrast filter. At least 3 sets of microscope samples are prepared
218 for each dewatered sample and at least 5 images at various points of each microscope sample are
219 taken.

221 *Image Analysis & Microscale redispersion assessment.*

222 The phase contrast microscopy images are selected for image analysis on account of their desired
223 contrast, clarity, and fibril density. Original CNF samples (reference sample) and dewatered
224 samples are analysed via modified version of a MATLAB code (GTFiber), which is uploaded in
225 GitHub. Changes in fibril lengths and widths induced by ultrasonic dewatering relative to the
226 original sample are assessed. The resulting length and width data are compiled into histograms.

228 **Results and Discussion**

229 **Mechanisms of Ultrasonic Dewatering**

230 For this study vibrating mesh transducer (VMT) is used to generate high frequency pressure waves.
231 The main distinguishing feature of these transducers is the presence of a metal mesh at the center
232 (Fig 1a, c, e) supported by a piezoelectric ceramic ring at the back (Fig 1b), which vibrates at high

frequencies (resonance frequency: 113-117 kHz) on application of voltage. The metal mesh has a tapered geometry with the smaller diameter opening at the back surface (diameter: 18 μm at top surface and 3-5 μm at back surface). When a water droplet is placed on a VMT, the vibration of the metal mesh creates a pressure gradient that pushes the water through the pores resulting in the ejection of water through the back surface as a mist (Fig 1d and 1f). The vibrations also result in surface instabilities generated at the air-water interface of the droplet at the front surface of the transducer, resulting in droplet atomization. Thus, the VMT helps eject water as a combination of mist ejection from the mesh and droplet atomization from air-water interface.

To evaluate if water could be separated from a CNF suspension (instead of a pure water droplet), we place a droplet of CNF suspension (~200 mg) on the VMT and subject it to ultrasonic vibrations. We observe that the liquid from the system is ejected through the back surface of the metal mesh (Fig 1d shows a schematic representation of the water ejected through the mesh) while a secondary liquid removal via atomization is observed from the top surface at the CNF suspension-air interface. This secondary atomization is verified using a 'glass slide test' (Online Resource 1 SI-Fig 3) as the atomization intensity is small compared to that of a pure liquid making it difficult to detect via visual inspection.

The presence of CNF in the water ejected through the mesh is tested by analyzing the solids content of the ejected water (i.e., the water was collected, allowed to dry, and the solids weighed). It is found that the fibrils ejected through the system in the water is insignificant. However, a very small quantity of CNF was seen on the back surface of the transducer mesh stuck to the plate at the end of the runs; their measured weight is insignificant (~0.001 times the initial CNF weight in the starting suspension). Here we hypothesize that, as water is removed from the system, the fibrils come closer together creating a network that prevents the passage of CNF through the metal mesh. Thus, the ultrasonic waves act to remove the unbound excess water in the CNF suspension.

Continuous Ultrasonic Dewatering of CNF Suspensions

Having established the feasibility of ultrasonic dewatering of CNF using VMT, we then design an ultrasonic dewatering platform that can operate as a continuous system (Fig 2). A continuous

platform allows for faster operation times compared to a batch process and potential incorporation into an existing industrial process. The present design allows precise control of the CNF suspension flow rate by using a syringe pump, as well as an easy means of changing the transducer configuration. It must be noted that we control the flow rate of the suspension at the syringe outlet rather than at the transducer surface which can get effected by various parameters such as gravity-effects, scraper blade velocity and changing viscosity of the suspension undergoing dewatering. Dewatering experiments focused on the effect of suspension flow rates and transducer configurations (e.g., 1T and 2T) on the extent of dewatering of CNF suspensions.

Effect of flow rate

For the single transducer configuration water removal from 3 wt.% CNF suspension is 16.3 ± 0.98 wt. %, 23.6 ± 4 wt. %, 36.4 ± 3.4 wt. % and 55.6 ± 11.7 wt. % for 60 ml/hr, 30 ml/hr, 15 ml/hr and 10 ml/hr flow rates, respectively (1T, Fig 3). The percentage of water removal demonstrates an inverse relation with the flow rate of the CNF suspension over the transducer, increasing with lower flow rates. The highest final CNF concentration (at 1T configuration) of 7.1 ± 1.8 wt. % is achieved at the lowest flow rate of 10 ml/hr. These results suggest that lower flow rates ensure higher contact time (residence time) between CNF suspension and the transducer surface, thereby ensuring higher exposure to the ultrasonic waves and higher extent of dewatering.

For the 2T configuration, a similar inverse trend between flow rate and percent water removal is observed, but the level of water removal is higher. Water removal for 60 ml/hr, 30 ml/hr, 15 ml/hr, and 10 ml/hr flow rates are 19.5 ± 3.5 wt. %, 35.6 ± 2.9 wt. %, 45.4 ± 2.5 wt. %, and 71.9 ± 8.3 wt. %, respectively. The highest water removal of 71.9 ± 8.3 wt. % (Fig 3(a)) (for 2T configuration) occurred at 10ml/hr flow rate, and resulted in a final CNF concentration of 11.2 ± 2.8 wt. % (Fig 3(b)).

However, at lower flow rates (10 ml/hr and lower), a low-pressure induced separation of CNF from the water is observed within the syringe. This leads to the water being pushed out of the syringe while the CNF form a compact network in the syringe making it difficult to pass through the syringe nozzle (see Online Resource 1: SI-Fig 4). An inhomogeneity in the initial CNF

concentration exiting the syringe occurs, resulting in the higher error bars for these experimental sets (at 10 ml/hr, Fig 3). Such phase separation becomes severe for the 5 ml/hr flow rate experiments and hence, flow rates below 10 ml/hr are not feasible for the present setup.

Effect of transducer configuration

Understanding the effect of transducer configuration on dewatering at any given flow rate can help determine the optimum operational conditions and give insight into scale-up potential of this platform. For any given flow rate, the amount of water removed and dewatering rates are higher for the 2T configuration as compared to the 1T configuration (Fig 3). This is attributed to the higher contact area achieved between the CNF suspension and the transducer for the 2T configuration. Interestingly the amount of water removed at the fastest flow rate of 60 ml/hr is similar for both the configurations, suggesting that there exists a critical residence time between the suspension and the transducer surface for the effects of the vibrations to significantly manifest in the system. Thus, below this critical residence time, increasing the contact area (through additional transducers) does not significantly affect the extent of dewatering for the system. Transducers can further be arranged in a sandwiched configuration, with the two transducers facing each other and CNF suspension flowing in between them (Online Resource 1: SI-Fig 5). The increase in the dewatering extent with an increase in the number of transducers suggests the potential for further scale-up of the system as arrays of transducers can be leveraged to increase dewatering amount at faster flow rates.

Fig 3 (a) wt. % of water removed and (b) the final concentration of CNF in wt.%, at various flow rates during the continuous flow ultrasonic drying using VMT. The data were collected for two transducer configurations: single transducer and double transducer. (c) Dewatering rate of the various flow rates and transducer configuration.

312

313 *Dewatering Rate*

314 Dewatering rate for each flow rate at 1T and 2T configuration are calculated using equation 4 and
 315 presented in Fig 3(c). Although higher extent of dewatering is achieved at lower flow rates due to
 316 prolonged contact time with the transducer surface (as seen in Fig 3(a)), dewatering rates follow a
 317 reverse trend. The dewatering rate is directly proportional to the flow rate with highest dewatering
 318 rates ($161 \pm 8.9 \text{ mg/min}$ for 1T and $192 \pm 33.9 \text{ mg/min}$ for 2T) achieved corresponding to 60

ml/hr flow rate. The dewatering rate decreases to $108 \pm 14 \text{ mg/min}$, $82 \pm 6 \text{ mg/min}$ and $80.9 \pm 11 \text{ mg/min}$ for 1T configuration and to $134 \pm 14 \text{ mg/min}$, $107 \pm 9 \text{ mg/min}$ and $102 \pm 10.7 \text{ mg/min}$ for 2T configuration at flow rates 30 ml/hr, 15 ml/hr and 10 ml/hr, respectively. This trend can be explained by the non-linear dewatering behaviour seen for VMT at static dewatering experiments (Online Resource 1: SI-Fig 6). A direct dependence of the dewatering rate on the transducer number is also observed. Higher dewatering rates are observed at 2T configuration as compared to the 1T configuration for all flow rates. Thus, dewatering rate can be leveraged as a design parameter, along with the transducer configuration and flow rates, for future scale-up of our system.

Redispersion of Dewatered CNF

Agglomeration of CNF triggered by the removal of water is a major disadvantage of dried CNFs, as the particles lose their nano-dimensions and high surface to volume ratio. As many CNF applications require a homogeneously dispersed suspension of the fibrils at the nanoscale, successful redispersion of the dewatered samples to attain pre-dewatered dimensions becomes a crucial processing step. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM), although widely used for assessing redispersion quality due to their nanoscale resolution, comes with inherent challenge associated with deconvoluting the effect of the drying needed in TEM and SEM sample preparation, which often lead to agglomeration triggered by droplet evaporation. Thus, to assess the dispersion state and the potential of agglomeration within redispersed CNF suspensions, a suspension-based characterisation technique is needed. One such technique is Dynamic Light Scattering (DLS), which has been used for calculations of hydrodynamic radius of CNMs (Foster et al. 2018). However, the fibril dimensions used in our study exceeded the size range of operation by DLS, thereby rendering this technique unapplicable in this study. Consequently, we use visual inspection and optical microscopy imaging of the suspension to evaluate the level of CNF agglomeration.

The extent of CNF agglomeration is assessed in two modes: macroscopically via visual inspection, and microscopically via phase contrast optical microscopy. By comparing the original never dried CNF suspension (reference sample) to the redispersed dewatered CNF suspensions, it is considered

here that any differences would likely be attributed to the dewatering and redispersion processes. Large agglomeration of the fibrils ($>1\text{ mm}$ in size), identified by the presence of opaque clump like structures in the suspension, are assessed by direct visual inspection of the suspension. This provided a fast and easy initial screening for agglomeration in the redispersed system. Finer scale agglomeration of the fibrils is assessed using phase contrast microscopy imaging to inspect fibril distribution and potential agglomeration in microscale range (between $1\text{ }\mu\text{m}$ to $1000\text{ }\mu\text{m}$). Unfortunately, assessing the fibril features and agglomeration in the nano-scale (below $1\text{ }\mu\text{m}$) is not feasible by optical microscopy, hence, our claims are restricted to the microscale range.

Macroscopic Redispersion

Images of the redispersed samples for various flow rates along with reference sample (original 3 wt.% CNF sample diluted to desired wt.%) at 1 wt. % and 0.1 wt.% for 2T configuration are given in Fig 4 (images for 1T configuration are presented in Online Resource 1: SI-Fig 7). Set A are samples at 1 wt.% and Set B are samples at 0.1 wt.%. Visual inspection of the redispersed samples looked very similar to the reference CNF suspension suggesting that no drastic changes in the CNF fibrils due to agglomeration occur in the redispersed state. Even the highest dewatered CNF concentration (i.e. Fig 4 Set A and B, 2T setup at 10 ml/hr flow rate, corresponding to $\sim 11\text{ wt. \%}$ CNF as seen in Fig 3(b)) resulted into a well redispersed suspension using the vortex mixture.

Fig 4 Redispersed suspensions of ultrasonic dewatered CNFs at various flow rates along with the diluted reference CNF sample for 2T configuration. Set A: 1 wt.% samples. Set B: 0.1 wt.% samples.

368 *Microscopic Redispersion*

369 The micro-scale dispersion state and the level of CNF agglomeration is assessed by phase contrast
370 optical microscopy. To assess if dewatering altered the level of CNF agglomeration, the
371 redispersed suspensions were compared to the reference CNF suspensions. Images obtained for
372 the redispersed samples using the phase contrast microscope are presented in Fig 5 (and in Online
373 Resource 1: SI-Fig 8 and 9). For each sample, the images were taken at multiple different locations
374 to rule out user bias (~15 images per sample). A comparison with the reference sample revealed
375 that the images for different dewatered samples are visually indistinguishable from each other. At
376 1 wt.% and 0.1 wt.% concentration, the fibril density was quite high forming intertwined networks.
377 Although the fibrils looked well dispersed, at these high densities it was difficult to judge if any
378 agglomeration is taking place (see Online Resource 1: SI-Fig 8). Hence, suspensions with higher
379 dilution (i.e. 0.01 wt.%) are used to obtain sparsely packed fibril images thereby allowing us to
380 analyse the fibril arrangement and potential agglomeration using the phase contrast mode for
381 imaging.

Fig 5 Phase contrast optical microscopic images using 20x magnification lens for the fibrils in suspension for (a) original CNF sample diluted to 0.01 wt% and (b) CNF sample dewatered using 2T configuration at 10 ml/hr, redispersed to 0.01 wt%.

382
383 Optical microscopy images at 0.01 wt% for a diluted original CNF sample along with a redispersed
384 dewatered sample are shown in Fig 5(a) and (b) respectively (and in Online resource 1: SI-Fig 9).

Long branched fibrils as well as short individual fibrils are seen throughout the suspension in all the samples. The images for the ultrasonic dewatered samples are visually indistinguishable from the original sample. Hence, a more statistical approach is undertaken to assess the potential agglomeration on redispersion of the dewatered samples through analysing the changes in fibril width and length for these samples.

Manual calculation of the length and width of individual fibril is tedious time-consuming work and leads to user bias and manual errors. Hence a computational approach is adapted in this work where all fibrils in each image are analysed using a modified MATLAB code originally designed to analyse fibre dimensions in high contrast imaging techniques such as TEM and SEM (Online Resource 2). The phase contrast optical microscopy imaging technique used in this study successfully generated high contrast images that are compatible to be used with the code. Thus, phase contrast microscopy in combination with the MATLAB code provided a fast and easy fibril characterisation technique for CNF suspensions. At least 10 images (>600 fibrils) for each sample are analysed. The length and width data for each flow rate and the reference sample were then compiled into histogram graphs as presented in Fig 6. The length and width histograms have similar distributions for both the redispersed dewatered CNF suspensions and the reference CNF suspension, suggesting minimal microscopic agglomeration effects. The general trend displayed by this analysis for all the flowrates and transducer configurations (Fig 6 and Online Resource 1: SI-Fig 10) exhibits that fibril lengths and widths are widely conserved during the ultrasonic dewatering process. This dimensional conservation suggests that the ultrasonic dewatering technique presented in this study successfully removes water from CNF suspensions without inducing much if any permanent agglomeration.

Fig 6 Histogram data for length and width of the redispersed samples subjected to ultrasonic dewatering at 2T configuration and various flow rates. At least 10 images (>600 fibrils) for each sample are analyzed. (a) Fibril length for 10ml/hr. flow rate samples plotted along with the reference sample. Fibril length data for samples dewatered at (b) 15 ml/hr. (c) 30 ml/hr. and (d) 60 ml/hr. flow rates. (e) Fibril width for 10 ml/hr. flow rate samples plotted along with the reference sample. Fibril width data for samples dewatered at (b) 15 ml/hr. (c) 30 ml/hr. and (d) 60 ml/hr.

Energy Consumption to Dewater:

The existing dewatering and drying methods for CNFs are highly energy intensive, thereby making it important to develop energy-efficient and economically sustainable dewatering/drying methods. Ultrasonic dewatering system for CNFs is developed with these goals in mind and estimation of its energy consumption is conducted. Based on the power rating for a single VMT used in this study i.e. 2.4 J/s, the energy consumed by the ultrasonic dewatering platform is estimated for per kilogram of water removed. For this calculation, we assume that the system is operating at 1T configuration and 10 ml/hr flow rate of the CNF, which takes 30 mins to dewater 5 grams of a 3 wt.% CNF suspension. The dewatering time is extrapolated for removal of 1 kg of water from CNF suspension and the energy consumed by the system is estimated to be ~1542.8 kJ/kg of water removed (summarized in Table 3). A comparison of the 2T system with the 1T system is also presented in Table 3.

Energy consumption data for CNM dewatering processes such as centrifugation, filtration, shear induced dewatering or pressing is not available in literature to the best of our knowledge. While energy consumption comparison with these methods would have been ideal, we evaluate our

system based on thermal drying systems to offer a rough estimate. The enthalpy of evaporation of water at 25°C is 2441.7 kJ/kg(Engineering Toolbox 2010), which is the minimum energy that must be supplied for any thermal drying system. The average energy consumption value for an industrial spray drier, the most widely used drying technique for CNMs, is ~5469.3 kJ/kg of water removed(Baker and McKenzie 2005) which is significantly higher than the estimated energy consumption for the ultrasonic dewatering system. It is also worthwhile mentioning that the transducers used in this study are inexpensive (<\$1 each) and are easily available for bulk purchase. Thus, the low energy requirements of these transducers along with their low capital cost makes ultrasonic dewatering a promising energy efficient and economically viable alternative for dewatering of CNFs.

Table 3: Overview of operational and economical parameters for the ultrasonic dewatering platform

System	Initial weight	Dewatered water %	Time taken	Final CNF concentration	Energy consumed
1T	5g of 3wt.% CNF	~56 wt. %	30 mins	~7 wt. %	~ 1542.8kJ/kg of water removed
2T	5g of 3wt.% CNF	~72 wt. %	30 mins	~11 wt. %	~2400kJ/kg of water removed

Conclusion

A new dewatering technique that uses ultrasonic transducers to remove water from the CNF suspension is introduced in the present work. The custom build ultrasonic dewatering platform is shown to efficiently remove water from the system without triggering agglomeration of the fibrils. The flow rate and the spatial transducer configurations are varied to study their effect on the dewatering performance. Decreasing the flow rate of the CNF suspension resulted in an increase in the amount of water removal for the system. This is due to the increased residence time of the suspension over the transducer surface. However, the trade-off for increased dewatering is longer dewatering durations (due to lower flow rates) along with the higher inconsistency in the amount of water removed (higher error bars) due to the low-pressure induced phase separation within the syringe at lower flow rates. On the other hand, increasing the number of transducers increases the amount of water removed for all flow rate through an increased working contact area between the transducer and the suspension. The dewatering rate for our system is found to be directly dependent on the flow rate as well as the transducer number as a result of the non-linear dewatering behaviour

of the transducer. Up to 72 % of water removal is achieved, corresponding to a final CNF concentration of ~11 wt.%.

The redispersion of the dewatered samples achieved via vortex mixing resulted in homogeneous suspensions with no additional agglomeration observed on visual inspection. Further analysis using phase contrast microscopy provided a fast and easy technique for microscale visualisation of these fibrils. Dimensional analysis of the fibrils revealed a similar histogram distribution for the redispersed samples when compared to the original sample. Finally, energy estimations revealed that the proof-of-concept system operates at reduced energy requirements when compared to thermal drying systems thus presenting a promising prospect for an energy-efficient and economically sustainable alternative for CNF dewatering.

Acknowledgment

The authors would like to acknowledge the support from Renewable Bioproducts Institute at Georgia Tech, Atlanta, USA.

Declarations

Conflicts of interests

The authors declare no conflict of interest.

Availability of data and material

All MATLAB code and data for this article are accessible here: https://github.com/bhamla-lab/Ultrasound_CNf_Dewatering2021

Consent for publication

The authors consent for publication by the journal.

References

Al Zaitone B (2019) Drying kinetics of cellulose nanofibers suspensions. In: 21st International Drying Symposium.

475 pp 11–14
 476 Amini E (Nima), Tajvidi M, Bousfield DW, et al (2019) Dewatering Behavior of a Wood-Cellulose Nanofibril
 477 Particulate System. *Sci Rep* 9:1–10. <https://doi.org/10.1038/s41598-019-51177-x>
 478 Astorsdotter J (2017) Dewatering Cellulose Nanofibril Suspensions through Centrifugation
 479 Baker CGJ, McKenzie KA (2005) Energy Consumption of Industrial Spray Dryers. *Dry Technol* 23:365–386.
 480 <https://doi.org/10.1081/DRT-200047665>
 481 Beck S, Bouchard J, Berry R (2012) Dispersibility in Water of Dried Nanocrystalline Cellulose. *Biomacromolecules*
 482 13:1486–1494
 483 Beuguel Q, Tavares JR, Carreau PJ, Heuzey M (2018) Ultrasonication of spray- and freeze-dried cellulose
 484 nanocrystals in water. *J Colloid Interface Sci* 516:23–33. <https://doi.org/10.1016/j.jcis.2018.01.035>
 485 Boucher RMG (1959a) Ultrasonic boosts heatless drying.pdf. *Chem Eng* 66:151–154
 486 Boucher RMG (1959b) Drying by airborne Ultrasonics. *Ultrason news* 3:8–9
 487 Clayton SA, Scholes ON, Hoadley AFA, et al (2006) Dewatering of biomaterials by mechanical thermal expression.
 488 *Dry Technol* 24:819–834. <https://doi.org/10.1080/07373930600733093>
 489 Čolić M, Tomić S, Bekić M (2020) Immunological aspects of nanocellulose. *Immunol Lett* 222:80–89.
 490 <https://doi.org/10.1016/j.imlet.2020.04.004>
 491 Dimic-Misic K, Maloney T, Gane P (2018) Effect of fibril length, aspect ratio and surface charge on ultralow shear-
 492 induced structuring in micro and nanofibrillated cellulose aqueous suspensions. *Cellulose* 25:117–136.
 493 <https://doi.org/10.1007/s10570-017-1584-3>
 494 Dimic-Misic K, Maloney T, Liu G, Gane P (2017) Micro nanofibrillated cellulose (MNFC) gel dewatering induced
 495 at ultralow-shear in presence of added colloiddally-unstable particles. *Cellulose* 24:1463–1481.
 496 <https://doi.org/10.1007/s10570-016-1181-x>
 497 Dimic-Misic K, Puisto A, Paltakari J, et al (2013) The influence of shear on the dewatering of high consistency
 498 nanofibrillated cellulose furnishes. *Cellulose* 20:1853–1864. <https://doi.org/10.1007/s10570-013-9964-9>
 499 Engineering, Toolbox (2010) Water - Heat of Vaporization. [https://www.engineeringtoolbox.com/water-properties-](https://www.engineeringtoolbox.com/water-properties-d_1573.html)
 500 [d_1573.html](https://www.engineeringtoolbox.com/water-properties-d_1573.html). Accessed 4 Jun 2021
 501 Foster EJ, Moon RJ, Heux L, et al (2018) Current characterization methods for cellulose nanomaterials. *Chem Soc*
 502 *Rev* 47:2609–2679. <https://doi.org/10.1039/c6cs00895j>
 503 Gallego-Juárez JA, Riera E, de la Fuente Blanco S, et al (2007) Application of high-power ultrasound for
 504 dehydration of vegetables: Processes and devices. *Dry Technol* 25:1893–1901.
 505 <https://doi.org/10.1080/07373930701677371>
 506 Gallego-Juárez JA, Rodríguez-Corral G, Moraleta JCG, Yang TS (1999) A new high-intensity ultrasonic
 507 technology for food dehydration. *Dry Technol* 17:597–608. <https://doi.org/10.1080/07373939908917555>
 508 García-Pérez JV, Carcel JA, Mulet A, et al (2015) Ultrasonic drying for food preservation. In: *Power Ultrasonics*.
 509 Elsevier, pp 875–910
 510 Geng L, Mittal N, Zhan C, et al (2018) Understanding the Mechanistic Behavior of Highly Charged Cellulose
 511 Nanofibers in Aqueous Systems. *Macromolecules* 51:1498–1506.

<https://doi.org/10.1021/acs.macromol.7b02642>

Greguss P (1963) The mechanism and possible applications of drying by ultrasonic irradiation. *Ultrasonics* 1:83–86.

[https://doi.org/10.1016/0041-624X\(63\)90059-3](https://doi.org/10.1016/0041-624X(63)90059-3)

Han J, Zhou C, Wu Y, et al (2013) Self-assembling behavior of cellulose nanoparticles during freeze-drying: Effect of suspension concentration, particle size, crystal structure, and surface charge. *Biomacromolecules* 14:1529–1540. <https://doi.org/10.1021/bm4001734>

Hanif Z, Jeon H, Tran TH, et al (2018) Butanol-mediated oven-drying of nanocellulose with enhanced dehydration rate and aqueous re-dispersion. *J Polym Res* 25:191

Iwamoto S, Nakagaito AN, Yano H, Nogi M (2005) Optically transparent composites reinforced with plant fiber-based nanofibers. *Appl Phys A* 1112:1109–1112. <https://doi.org/10.1007/s00339-005-3316-z>

Miller T, Kramer C, Fisher A (2015) Bandwidth Study on Energy Use and Potential Energy Saving Opportunities in U.S. Pulp and Paper Manufacturing. US Dep Energy

Missoum K, Bras J, Belgacem MN (2012) Water Redispersible Dried Nanofibrillated Cellulose by Adding Sodium Chloride. *Biomacromolecules* 13:4118–4125

Moon RJ, Martini A, Nairn J, et al (2011) Cellulose nanomaterials review: structure, properties and nanocomposites. *Chem Soc Rev* 40:3941. <https://doi.org/10.1039/c0cs00108b>

Musielak G, Mierzwa D, Kroehnke J (2016) Food drying enhancement by ultrasound – A review. *Trends Food Sci Technol* 56:126–141. <https://doi.org/10.1016/j.tifs.2016.08.003>

Nigmatullin R, Johns MA, Muñoz-García JC, et al (2020) Hydrophobization of Cellulose Nanocrystals for Aqueous Colloidal Suspensions and Gels. *Biomacromolecules* 21:1812–1823.

<https://doi.org/10.1021/acs.biomac.9b01721>

Park C-W, Han S-Y, Namgung H-W, et al (2017) Effect of Spray-drying Condition and Surfactant Addition on Morphological Characteristics of Spray-dried Nanocellulose. *J For Environ Sci* 33:33–38.

<https://doi.org/10.7747/jfes.2017.33.1.33>

Patel DK, Dutta SD, Lim KT (2019) Nanocellulose-based polymer hybrids and their emerging applications in biomedical engineering and water purification. *RSC Adv* 9:19143–19162. <https://doi.org/10.1039/c9ra03261d>

Peng C, Momen AM, Moghaddam S (2017a) An energy-efficient method for direct-contact ultrasonic cloth drying. *Energy* 138:133–138. <https://doi.org/10.1016/j.energy.2017.07.025>

Peng C, Ravi S, Patel VK, et al (2017b) Physics of direct-contact ultrasonic cloth drying process *. *Energy* 125:498–508. <https://doi.org/10.1016/j.energy.2017.02.138>

Peng Y, Gardner DJ (2013) Influence of drying method on the material properties of nanocellulose I : thermostability and crystallinity. *Cellulose* 20:2379–2392

Peng Y, Gardner DJ, Han Y (2012a) Drying cellulose nanofibrils: In search of a suitable method. *Cellulose* 19:91–102. <https://doi.org/10.1007/s10570-011-9630-z>

Peng Y, Gardner DJ, Han Y, et al (2013) Influence of drying method on the surface energy of cellulose nanofibrils determined by inverse gas chromatography. *J Colloid Interface Sci* 405:85–95.

<https://doi.org/10.1016/j.jcis.2013.05.033>

- Peng Y, Gardner DJ, Han Y (2012b) Drying Cellulose Nanofibrils : Morphology Characterization. In: 55th International Convention of Society of Wood Science and Technology. pp 1–9
- Penttilä PA, Ra P, Maunu SL, et al (2012) The effect of drying method on the properties and nanoscale structure of cellulose whiskers. 901–912. <https://doi.org/10.1007/s10570-012-9695-3>
- Quiévy N, Jacquet N, Selavons M, et al (2010) Influence of homogenization and drying on the thermal stability of microfibrillated cellulose. *Polym Degrad Stab* 95:306–314
- Rantanen J, Maloney TC (2015) Consolidation and dewatering of a microfibrillated cellulose fiber composite paper in wet pressing. *Eur Polym J* 68:585–591. <https://doi.org/10.1016/j.eurpolymj.2015.03.045>
- Rebouillat S, Pla F (2013) State of the Art Manufacturing and Engineering of Nanocellulose: A Review of Available Data and Industrial Applications. *J Biomater Nanobiotechnol* 04:165–188. <https://doi.org/10.4236/jbnt.2013.42022>
- Sabarez H, Swiergon P, Knoerzer K (2019) Drying process and apparatus
- Sehaqui H, Liu A, Zhou Q, Berglund LA (2010) Fast Preparation Procedure for Large , Flat Cellulose and Cellulose / Inorganic Nanopaper Structures. 2195–2198
- Sethi J, Oksman K, Illikainen M, Sirviö JA (2018) Sonication-assisted surface modification method to expedite the water removal from cellulose nanofibers for use in nanopapers and paper making. *Carbohydr Polym* 197:92–99. <https://doi.org/10.1016/j.carbpol.2018.05.072>
- Sim K, Lee J, Lee H, Youn HJ (2015) Flocculation behavior of cellulose nanofibrils under different salt conditions and its impact on network strength and dewatering ability. *Cellulose* 22:3689–3700. <https://doi.org/10.1007/s10570-015-0784-y>
- UMaine The Process development Center. <https://umaine.edu/pdc/nanocellulose/>. Accessed 7 Jul 2021
- Wetterling J, Jonsson S, Mattsson T, Theliander H (2017) The Influence of Ionic Strength on the Electroassisted Filtration of Microcrystalline Cellulose. *Ind Eng Chem Res* 56:12789–12798. <https://doi.org/10.1021/acs.iecr.7b03575>
- Wetterling J, Sahlin K, Mattsson T, et al (2018) Electroosmotic dewatering of cellulose nanocrystals. *Cellulose* 25:2321–2329. <https://doi.org/10.1007/s10570-018-1733-3>
- Zhai L, Kim HC, Kim JW, Kim J (2020) Simple centrifugal fractionation to reduce the size distribution of cellulose nanofibers. *Sci Rep* 1–8. <https://doi.org/10.1038/s41598-020-68642-7>
- Zimmermann MVG, Borsoi C, Lavoratti A, et al (2016) Drying techniques applied to cellulose nanofibers. *J Reinf Plast Compos* 35:682–697. <https://doi.org/10.1177/0731684415626286>

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [FinalOnlineResource1.pdf](#)