



# Characterization of the filter cake drainage resistance coefficient for cellulose nanofibril suspensions

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**Abstract** The dewatering of cellulose nanofibril (CNF) suspensions is important in several processes such as concentrating the material before transportation, producing dry CNFs for composite structures, and applying the material to the surface of a paper web. Dewatering of a CNF suspension onto paper is one possible way to form a CNF layer on paper to produce excellent grease and oil-resistant packaging materials. Even though this property of the CNF suspension is important, little work has been reported in the literature to model and characterize this process. Refiner produced CNF suspensions were filtered on cellulose acetate membranes, filter paper, and paperboard in a standard Büchner funnel with a known vacuum pressure. The volume filtered as a function of time was recorded and the equation for constant pressure filtration was used to fit the results for

various initial concentrations and different qualities of CNFs. The specific cake resistance coefficient ( $\alpha$ ) was obtained from the results. The results revealed the dewatering of CNFs follows the filtration theory well with an  $R^2$  value greater than 0.98. Increasing the quality or degree of fibrillation of the CNFs gave a small increase in the specific cake resistance. The specific cake resistance obtained was able to predict the dewatering of a CNF suspension on a paperboard in a bench scale sheet former. Significant clogging of the base membrane or paper was found to occur, especially for the cellulose acetate membranes. These results provide insights regarding CNF dewatering and the basis for the design of filtration-based dewatering equipment as well as the operation of wet end processes that deposit CNFs onto the web.

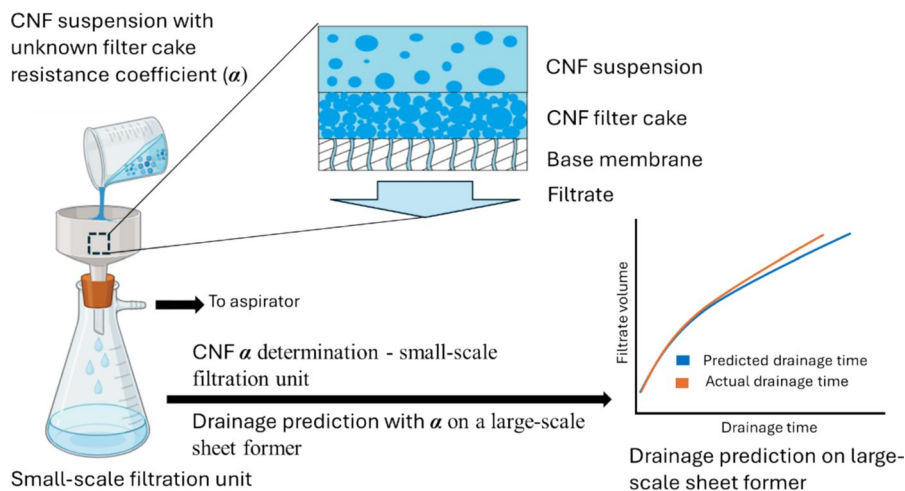
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## Graphical abstract



**Keywords** Cellulose nanofibrils · Nanocellulose · Dewatering · Drainage · Microfibrillated cellulose

## Introduction

Cellulose nanofibrils (CNFs) and related cellulose nanomaterials have attracted an enormous level of attention due to their unique set of properties and they have potential uses as paper coatings, films, adhesives, and reinforcements owing to their excellent grease and oxygen barrier resistance, the inherent outstanding specific strength of the individual fibrils, high aspect ratio, low density, high specific surface area, sustainability, and renewability as reviewed by several groups such as Moon et al. (2016).

CNFs are branched fibril-like cellulosic particles resulting from the mechanical grinding, refining, or homogenizing of wood pulp fibers in aqueous suspensions usually produced at less than 5 wt%. Therefore, for effective transportation and handling of CNFs, an enormous amount of water needs to be removed from the suspension. In addition, water needs to be removed in other various applications including the use of CNFs as reinforcing fillers, especially with hydrophobic matrices requiring the use of dry CNFs. As conventional drying processes are inherently energy-intensive, mechanical removal of the majority

of water present in the CNF suspension is important as it helps reduce drying time, saves energy, and cuts down on production costs as discussed by Amini et al. (2019). It is worth mentioning that different terminologies have also been used to describe CNFs by other researchers. These include microfibrillated cellulose (MFC) (e.g. Lavoine et al. 2012; Dimic-Misic et al. 2013), nanofibrillated cellulose (NFC) (Khalil et al. 2014), and microfibrillar cellulose (Agarwal et al. 2018). Herein, we made use of the name cellulose nanofibrils (CNFs) as suggested by the International Standards Organization as reported in Moon et al. (2016).

The quantity and characteristics of the fines formed during pulp fibrillation for CNF production typically depend on the type and duration of refining and treatment described by Kelly et al. (2021). Herein, fines are defined as fibers with a length of less than 200  $\mu\text{m}$ . The percentage fines of CNFs therefore refer to the percentage of CNFs with fiber length less than 200  $\mu\text{m}$ . Increased percentage of CNF fines typically correlates well with greater fractions of CNFs with highly branched and extended fibrils with higher aspect ratios resulting in an enhanced percolation network within suspensions (Stelte and Sanadi 2009). The enhanced percolation network coupled with the increased hydrogen bonding associated with a higher percentage of CNF fines has been shown to improve

the strength and surface properties of cellulosic non-woven materials such as paper hand sheets. As a typical example, Johnson et al. (2016) reported an increase in the tensile index and reduction in the surface roughness of paper hand sheets which correlated well with the percentage of fines of CNFs used for reinforcement. These enhanced mechanical properties associated with higher percentage fines of CNFs make them excellent candidates for various applications. However, higher percentage fines have been shown to also result in increased drainage times during dewatering of CNF suspensions which limits their use (Ringania et al. 2023).

Pressure filtration where the differential pressure is either supplied by a vacuum (Lee et al. 2020), through pressing (Amini et al. 2017), or by using ultrasonic energy (Ringania et al. 2023), provides an efficient means to dewater CNF suspensions. Ringania et al. (2022) reported a 50% reduction in energy with ultrasonic dewatering of CNF suspensions compared to the conventional spray drying technique. Also, current processing methods to produce cellulose-based composite panels with CNFs as adhesives involve cold press dewatering followed by drying in a hot press (Amini et al. 2017). Tajvidi et al. (2016) were the first to report the “contact dewatering” of CNF slurry when mixed with wood particles. They found that contact of CNF slurry with wood particles turns the majority of the adsorbed water tightly bound to the hydroxy group of CNFs into free water thereby significantly reducing the energy and time required for dewatering during pressing hence providing an energy-efficient technique for composite panel production with CNFs as binder. In addition, CNFs have been shown to be effective grease and oxygen barriers when dried in the form of a film (Aulin et al. 2010; Tayeb et al. 2020a; 2020b). However, the application of CNFs on paperboard by conventional coating techniques is a challenge (Lavoine et al. 2014; Mousavi et al. 2017). Pressure filtration dewatering of CNF suspensions onto a formed web is one of the possible ways to produce paper with a CNF layer. Hence, to optimize the production process, understanding and controlling the drainage rates of CNF slurry is key.

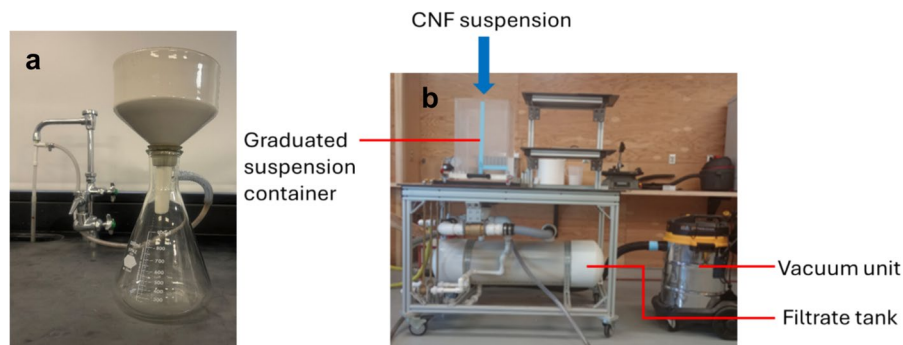
Herein, “dewatering” and “drainage” refer to the removal of liquid from the suspension during a filtration process, and the formed wet solid structure

during dewatering is termed a “filter cake”. To date, there has been some effort to control the drainage rates of cellulose-based suspensions, but little effort on the drainage of CNF suspensions. Paradis et al. (2002) determined the drainage resistance coefficient of different pulp grades of paper-making stocks and showed that the drainage rate changes because the filter cake characteristics change as dewatering progresses. Dimic-Misic et al. (2013) studied the effects of CNF swelling on the drainage behavior of cellulose furnishes and found that CNF suspensions added to the furnish predominantly controlled the dewatering process due to significant clogging of the filter cake with ultrafine fibrils. The influence of salt addition on CNF suspension dewatering behavior has also been studied and it was shown that the drainage rate is affected by the type and concentration of the added salt (Rao et al. 2022). Ringania et al. (2023) assessed the influence of fines content on water retention behavior and dewatering rates of CNF suspensions: high fine percentage displays high water retention capacity resulting in longer dewatering times.

Using compressibility and dynamic parameters obtained from pressure filtration tests, Rainey et al. (2011) developed a dynamic model to predict the dewatering behavior of bagasse pulp. Darcy’s law has also been used to measure the permeability of filter cake during the production of fiberboard (Pettersson et al. 2006). Using the Kozney–Carman theory, Nilsson and Stenström (1997) developed a model to predict the permeability of cellulose filter cake of pulp and paper structures. However, little quantification and modeling of the dewatering of CNF suspensions is reported in the literature.

In this study, the filter cake resistance coefficient ( $\alpha$ ) of CNF suspension was determined using the constant pressure filtration and a standard model for filtration. The influence of the initial slurry concentration and percentage fines of CNF suspension as well as the type of base membrane on dewatering behavior was also investigated. In addition, using the obtained coefficient together with the filtration equation, a model was developed to predict the drainage behavior of CNF suspensions. The results of this study should be of value to designing filtration based dewatering equipment in addition to the design and operation of wet-end processes that deposit CNF onto the web.

**Fig. 1** Small-scale pressure filtration unit **a** and large-scale laboratory-based sheet former **b**



## Materials and methods

A cellulose nanofibril (CNF) suspension of 3 wt% solids that was produced by the mechanical refining of bleached softwood kraft pulp was donated by the Process Development Center (PDC), University of Maine, Orono. CNF samples with 60%, 70%, 80%, and 90% fines were used in the present work. Fines herein are defined as fibrils with a length less than 200  $\mu\text{m}$  measured by a compact fiber analyzer (MorFi, Techpap Inc., Grenoble, France). The 90% fines CNFs have been distributed to a large number of companies and research institutes over the last decade and has been characterized with Transmission Electron Microscope by Mousavi et al. (2017) and Purnington et al. (2019): the refiner produced CNFs had a significant amount of fiber diameters under 100 nm but many of the fibers had diameters that were on the order of 100–200 nm and an average diameter of 125 nm. Other characterizations of the 90% fines CNFs were performed by Desmaisons et al. (2017) and identified as CNF7 in that publication. Dried bleached softwood kraft (BSKP) fibers were soaked overnight in deionized (DI) water and disintegrated at 1.5 wt% consistency for 3 min at 3200 rpm in the production of kraft pulp suspension for comparison.

Standard paperboard (SP) of basis weight 341  $\text{g}/\text{m}^2$  and thickness 0.437 mm was donated by Sappi North America (Westbrook, Maine). The 125 mm diameter Whatman filter paper (FP) of basis weight 106  $\text{g}/\text{m}^2$  and thickness 0.186 mm was supplied by Sigma Aldrich (Burlington, Massachusetts). SP and FP had a pore size of 2.6 and 3.2  $\mu\text{m}$ , respectively, as determined from mercury porosimetry. The paperboard was used as a membrane because filtration of CNF slurry onto paperboard is one potential method to produce grease-resistant tableware using a layer

of CNFs on the paperboard. Cellulose acetate membranes of 0.8  $\mu\text{m}$  (CA-0.8) and 1.2  $\mu\text{m}$  (CA-1.2) pore size were supplied by Sterlitech (Auburn, Washington). The basis weight and thickness of the membranes were determined to be 44  $\text{g}/\text{m}^2$  and 0.080 mm, respectively.

Vacuum pressure dewatering of the sample suspensions was achieved using a standard 900 mL flask fitted with a 13.5 cm Büchner funnel. Vacuum pressure was supplied using a water aspirator connected to the flask as shown in Fig. 1a. This constituted the bench-scale setup and should be easy for other laboratories to replicate. Prepared suspensions of the samples were immediately poured by hand gently from the beaker into the Büchner funnel lined with a 12.5 cm diameter wetted base membrane including SP, FP, CA-0.8, or CA-1.2. The suspensions were allowed to drain under 0.94 atm vacuum pressure; the pressure was determined with a portable device (model TC-169 A, DEYI, China). The time needed to collect specific volumes of the filtrate was recorded using a stopwatch. The time was started from the initial pouring of the suspension into the Büchner funnel and the time for the level of the filtrate to reach the marked volumes (250–600 mL with an interval of 50 mL) on the Büchner flask as the suspension dewatered was continuously recorded. From the obtained data, the filter cake resistance coefficients were determined using the linearized form of the constant pressure filtration equation.

The filtration of a CNF suspension with a solid concentration of  $C_S$  through a membrane can be modeled as two flow resistances in series, one of which, membrane resistance ( $R_M$ ), defined as membrane thickness ( $L_M$ ) divided by its Darcy's permeability ( $k_M$ ), i.e.  $R_M = L_M/k_M$ , may be assumed

constant whereas the other, the cake resistance ( $R_C$ ) increases with time as the filtercake thickness increases (Mahdi et al. 2019; Iritani et al. 2012). The change in filtrate volume per unit area ( $V_A$ ) becomes a function of time ( $t$ ) at the constant driving pressure drop ( $\Delta P$ ), and water viscosity ( $\mu$ ), giving:

$$\frac{dV_A}{dt} = \frac{\Delta P}{\mu (R_C + R_M)} \quad (1)$$

The resistance to flow through the filter cake ( $R_C$ ) was assumed to be directly proportional to the basis weight of the filter cake (i.e., the mass of cake deposited per unit area) with the proportionality constant termed the filter cake resistance coefficient ( $\alpha$ ), i.e.

$$R_C = \alpha C_S V_A \quad (2)$$

The filter cake resistance is related to the Darcy permeability, the void fraction, and the density of the particles in the filter cake and is discussed in standard texts.

Substituting Eq. (2) into Eq. (1) and integrating with the following boundary conditioning  $t=0$ ,  $V_A=0$  and  $t=t$ ,  $V_A=V_A$ , and rearranging gives:

$$\frac{\Delta P t}{\mu} = \frac{\alpha C_S V_A^2}{2} + R_M V_A \quad (3)$$

Rearranging Eq. (3) to obtain a linear relationship between  $t/V_A$  and  $V_A$  gives:

$$\frac{t}{V_A} = \frac{\mu \alpha C_S}{2 \Delta P} V_A + \frac{\mu R_M}{\Delta P} \quad (4)$$

where  $t$  is the drainage time (s),  $V_A$  is filtrate volume per unit area ( $\text{m}^2$ ) at any time  $t$ ,  $\mu$  is the viscosity of water (Pa.s),  $\alpha$  is the filter cake resistance coefficient ( $\text{m/kg}$ ),  $C_S$  is the suspension concentration ( $\text{kg/m}^3$ ),  $\Delta P$  is the differential pressure (Pa), and  $R_M$  is the base membrane resistance ( $1/\text{m}$ ). This is a form of the standard expression for constant pressure filtration.

A plot of  $t/V_A$  against  $V_A$  should give a linear response. The filter cake resistance coefficient is obtained from the slope of the results using the following values  $\mu=0.001$  Pa s,  $\Delta P=9.5 \times 10^4$  Pa, with an area of base membranes  $A=0.012$   $\text{m}^2$ . From these values unless otherwise stated,  $\alpha$  ( $\text{m/kg}$ )

is calculated from the slope of the obtained linear plots.  $C_S$  changes with solids of the initial suspension and is known.

The base membrane resistance ( $R_M$ ) was determined using the Darcy's equation:

$$R_M = \frac{\Delta P}{\mu v} \quad (5)$$

In determining  $R_M$ , the time  $t$  (s) needed to drain 800 mL of deionized water (DI) through the base membrane was recorded. From the obtained data, the flow rate  $Q$  ( $\text{m}^3/\text{s}$ ) was determined to calculate Darcy's velocity,  $v$  ( $\text{m/s}$ ). All other symbols remain the same as previously defined. From Eq. 5,  $R_M$  ( $1/\text{m}$ ) is calculated.

Drainage was also performed on a large-scale setup using a lab-scale sheet former (Fig. 1b) manufactured by 3D Fiber Engineering, LLC (Verona, WI) to confirm the results and validity of the developed model obtained using the small-scale setup. The operation of the sheet former is like that described for the small-scale setup. For the sheet former, a well-graduated rectangular container of 0.093  $\text{m}^2$  drainage area ( $A$ ) is connected to a vacuum pump that supplies a differential pressure ( $\Delta P$ ) of  $9.8 \times 10^4$  Pa. Standard paperboard (SP) with an  $R_M$  of  $1.86 \times 10^{11} \text{ m}^{-1}$  was used as the base membrane. These values, unless otherwise stated, were employed in Eq. (4) for the drainage prediction on the sheet former. The predicted results were then compared to those obtained in an actual experiment using the sheet former.

To better understand the influence of base membranes on CNF dewatering, SP and CA-0.8 membranes were both contacted with CNFs through vacuum dewatering to achieve a coat weight of 40 g/ $\text{m}^2$  each. The coated samples were oven dried and tested for their porosity. The porosity of the fresh and CNF coated SP and CA-0.8 membrane was determined by saturating the samples with silicone oil and measuring the change in weight. To measure the porosity using silicone oil, the samples were first cut into  $2 \times 2 \text{ cm}^2$  square pieces. The mass and thickness of these pieces were then measured followed by soaking in silicone oil for 6 h. The excess oil on the surface was gently removed with wipes and the samples were weighed. The porosity  $P$  of the film was calculated as:

$$P = \frac{W_2 - W_1}{\rho_o AL} \quad (6)$$

where  $W_2$  is the weight of the oil-saturated film,  $W_1$  is the weight of the dry film,  $\rho_o$  is the density of the silicon oil,  $A$  is the surface area of the sample, and  $L$  is the thickness of the sample.

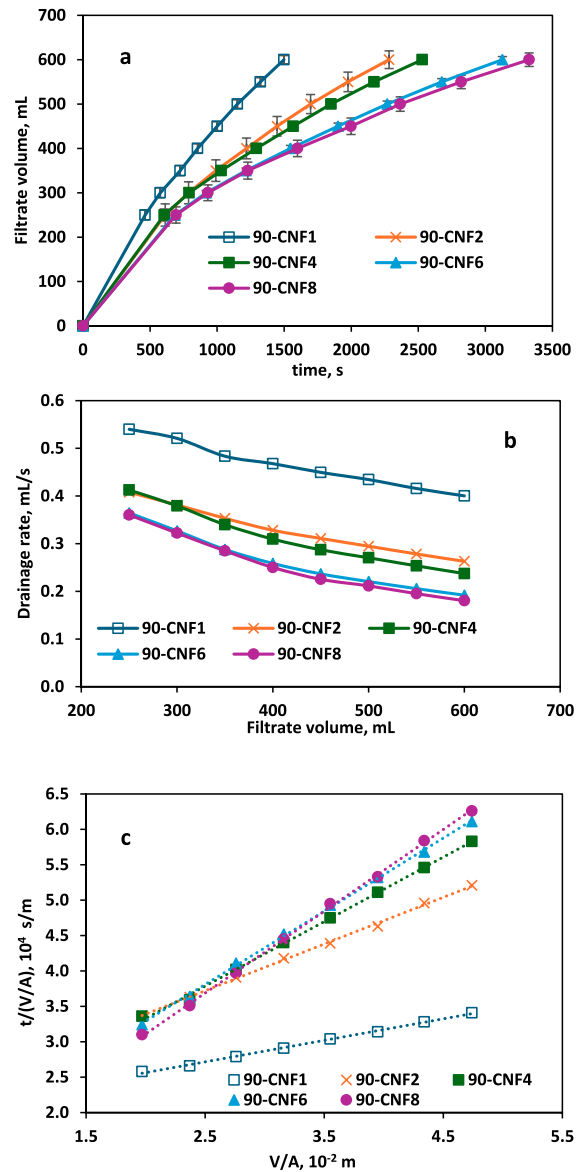
The microstructure and surface morphology of the SP and CA-0.8 membranes were studied using Zeiss Nvision 40 SEM (Oberkochen, Germany) to better understand the influence of the base paper and membrane on dewatering. Cut samples were placed on specimen mounts with double-sided carbon tapes. Conductive silver paint was then applied, and the samples were sputter coated with a 7 nm thick gold/palladium layer using a Cressington 108 auto sputter coater (Ted Pella Inc., Redding, California). The SEM was performed at an accelerating voltage of 3 kV.

Experimental results were analyzed using three repeated measurements and reported as mean values  $\pm$  standard deviation.

## Results and discussion

90% fines CNFs were used to study the influence of concentration on drainage. The concentration of 90% fines CNF suspension used included 1 kg/m<sup>3</sup> (90-CNF1), 2 kg/m<sup>3</sup> (90-CNF2), 4 kg/m<sup>3</sup> (90-CNF4), 6 kg/m<sup>3</sup> (90-CNF6) and 8 kg/m<sup>3</sup> (90-CNF8) with the standard paperboard (SP) as the filtration membrane. The process resulted in a successful separation of CNFs from the suspension as no fibril was visually detected in the filtrate. For the CNF suspensions, there might be significant amounts of fibers of several microns in dimensions that could cover the pores to prevent seepage of the fibrils. In addition, the highly fibrillated and branched network of CNFs is likely to form an entanglement during drainage to prevent movement of the fibrils.

The result for the average amount of water removed over time as dewatering progressed for the 90% fines CNFs is shown in Fig. 2a. In Fig. 2b, the drainage rates obtained with accumulated filtrate volumes over time are represented. Figure 2c shows the linearized plots obtained for the different CNF slurry concentrations. From Fig. 2c and Eq. 4, the filter cake resistance coefficients ( $\alpha$ ) for different slurry



**Fig. 2** Amount of water removed over time **a**, dewatering rate **b** with accumulated filtrate volume over time and linear plot **c** for 90% fines samples at the concentrations of 1, 2, 4, 6, and 8 kg/m<sup>3</sup> as dewatering progressed denoted as CNF 1, CNF 2, CNF 4, CNF 6 and CNF 8, respectively

concentrations were determined from the slope of the lines and the results are presented in Table 1. From Fig. 2a, it takes about 11 min (660 s) to drain 250 mL filtrate volume of the 90% fines CNF suspension with an initial slurry concentration of 6 kg/m<sup>3</sup> under the experimental conditions. This result is almost the same as that reported by Rao et al. (2022) using

**Table 1** Filter cake resistance coefficient ( $\alpha$ ) measured for 90% fines CNF slurry of different initial concentrations

| Concentration, kg/m <sup>3</sup> | CNF filter cake resistance coefficient ( $\alpha$ ), $\times 10^{13}$ m/kg |
|----------------------------------|----------------------------------------------------------------------------|
| 1                                | $5.6 \pm 0.2$                                                              |
| 2                                | $5.9 \pm 0.7$                                                              |
| 4                                | $3.9 \pm 0.4$                                                              |
| 6                                | $3.6 \pm 0.4$                                                              |
| 8                                | $3.1 \pm 0.4$                                                              |
| Average                          | $4.42 \pm 1.3$                                                             |

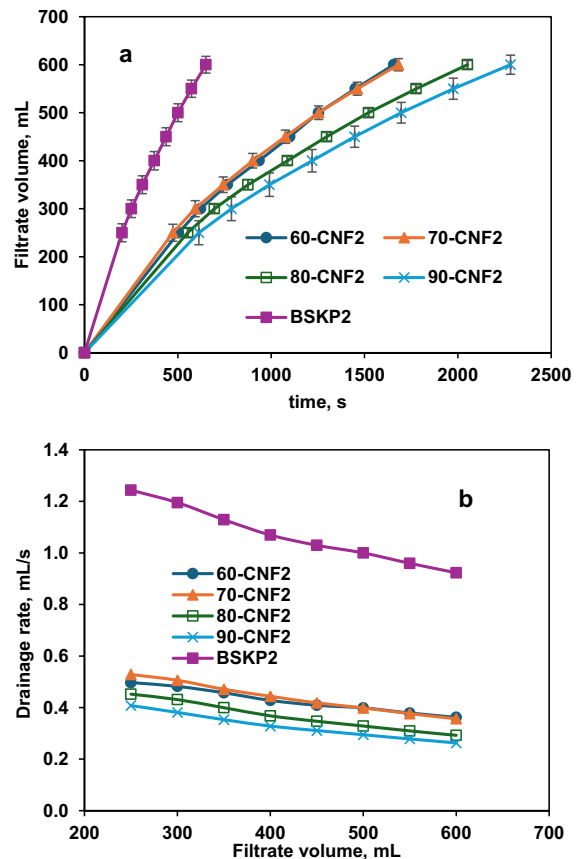
a similar setup and conditions. The group reported a total drainage time of 14 min for 90% fines CNF slurry of initial concentration 6.7 kg/m<sup>3</sup>. Also, the results shown in Fig. 2c demonstrate a strong correlation with an  $R^2$  value greater than 0.98 indicating that the obtained data follows well with the filtration theory (Eq. 4) and therefore they are appropriate for the estimating  $\alpha$ . The  $\alpha$  values obtained for the CNF slurries (Table 1) are similar and not in contrast to typical  $\alpha$  values reported for other types of suspensions. Mahdi et al. (2019) reported  $\alpha$  values of  $5.9 \times 10^{10}$  and  $7.4 \times 10^{11}$  m/kg for calcium carbonate and talc, respectively. From gravimetric water retention (GWR) test, Mousavi et al. (2017) reported water drainage values of about 2500 and 4000 g/m<sup>2</sup> for 3 wt% solids refiner produced CNF suspension (rCNF) for 90 and 180 s dewatering times, respectively. The differential pressure applied was 0.5 bar. Assuming small resistance through the membrane, this gives  $\alpha$  values of  $4.8 \times 10^{13}$  and  $3.8 \times 10^{13}$  m/kg, respectively. These values are close to those reported in Table 1.

The filter cake resistance coefficient  $\alpha$  should be a property only of how the fibers pack together and generate a certain pore space and flow passages through the filter cake;  $\alpha$  should not be a function of the initial concentration of the suspension. The results shown in Table 1 do show that  $\alpha$  tends to decrease at higher initial slurry concentrations. This implies that the drainage resistance is lower with higher solids. There are a few issues that may cause this. The high concentration of fibers may cause flocculation which generates channels through the filter cake. The channels generated improve drainage and hence less  $\alpha$  values at higher CNF concentrations. The change in  $\alpha$  as a function of the initial CNF slurry concentration is not strong and for values that may be of

**Table 2** Filter cake resistance coefficient ( $\alpha$ ) measured for different percentage fines CNFs and BSKP slurry

| Sample  | Filter cake resistance coefficient ( $\alpha$ ), $\times 10^{13}$ m/kg |
|---------|------------------------------------------------------------------------|
| 60-CNF2 | $3.3 \pm 0.1$                                                          |
| 70-CNF2 | $4.0 \pm 0.2$                                                          |
| 80-CNF2 | $5.4 \pm 0.4$                                                          |
| 90-CNF2 | $5.9 \pm 0.7$                                                          |
| BSKP2   | $1.2 \pm 0.2$                                                          |

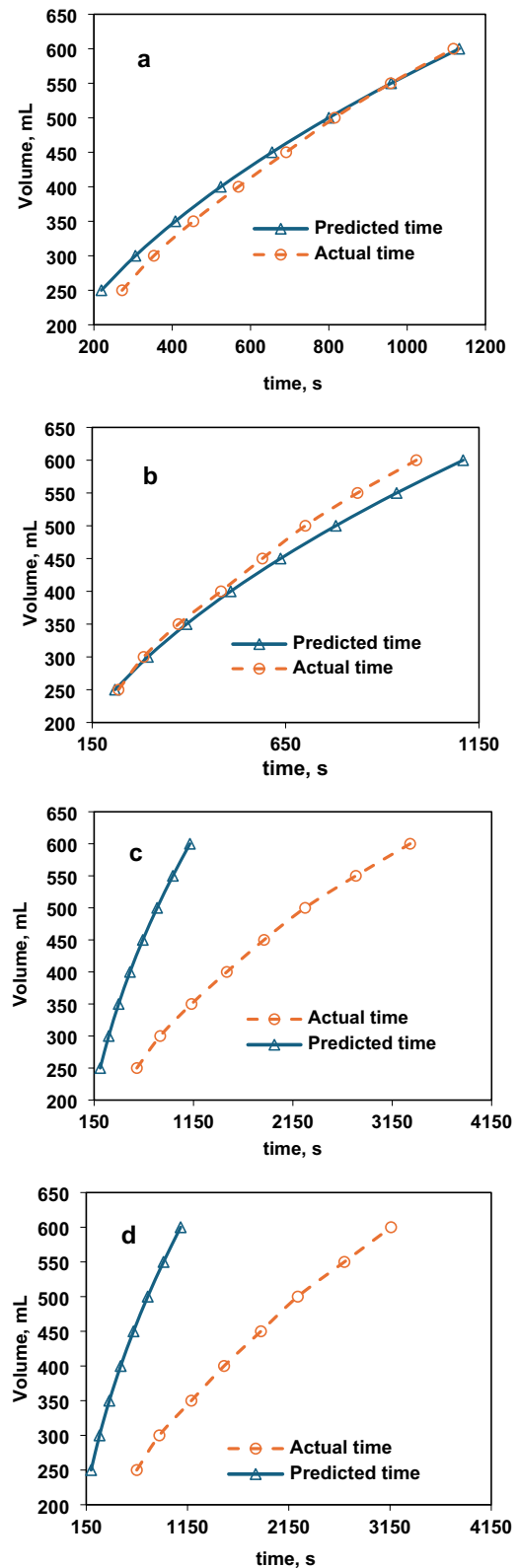
interest, an average is reported. Increasing fines percentage results in an increase in  $\alpha$  value with 90% fines CNFs exhibiting the highest value as reported in Table 2. The  $\alpha$  value of the 90% fines CNFs was five times that of bleached softwood kraft pulp (BSKP).

**Fig. 3** The amount of water removed **a** and dewatering rate **b** with accumulated filtrate volume over time for different percentage fines CNF slurry and BSKP slurry with 2 kg/m<sup>3</sup> initial slurry concentration as dewatering progressed

**Fig. 4** Drainage prediction for 90% fines CNF2 slurry dewatering with SP **a**, FP **b**, CA-0.8 **c** and CA -1.2 **d** as the base membrane

The increase from 60 to 90% fines was a factor of two and this factor correlated well with the rate of water removal as drainage progressed shown in Fig. 3. The increased  $\alpha$  value can be attributed to the tight and dense packing of the finer fibrils of the formed filter cake as dewatering progressed. BSKP recorded the highest rate of water removal at any point in time as dewatering progressed. Not much difference was observed between those of 60 and 70% fines CNFs. Also, it took the 90% fines CNF suspension about 35% longer to drain 600 mL filtrate volume compared to the 60% fines suspension. This observation is similar to that reported by Ringania et al. (2023). The group reported a 20% longer drainage time for 90% fines CNF suspension compared to 60% fines CNF suspension using ultrasonic dewatering. The increase in drainage times for higher fines percentage CNFs can be attributed to the increased sites for hydrogen bonding and tighter and denser packings of fibrils compared with lower fines percentage CNFs and BSKP. For the higher fines percentage CNFs, there is an expected significant ratio of bound water to unbound water compared to lower fines percentage. As bound water is considerably harder to remove compared to unbound water, consequently, longer drainage times are observed at each point for 90% fines CNFs compared to 60% fines CNFs.

To test the validity of the determined  $\alpha$ , the average value of the  $\alpha$  for the 90% fines CNF slurries of different concentrations was employed in Eq. (4) and used to predict the time to drain different filtrate volumes of 2 kg/m<sup>3</sup> initial concentration of 90% fines CNF suspension. The predicted results were compared to those obtained in an actual experiment using the small-scale setup. In the prediction, different base membranes including SP, FP, CA-0.8, or CA-1.2 were used to study the influence of the base membrane on  $\alpha$  and drainage behavior. For drainage prediction, using SP or FP as the base membrane, the predicted dewatering times agreed well with the actual experimental times (Fig. 4a, b). The highest recorded relative absolute errors for SP and FP as base membranes for dewatering were 19 and 12%, respectively. However, with CA membranes, there were considerable large deviations shown in Fig. 4c, d. The highest



**Table 3** Base membrane resistance coefficient determination after using for 90-CNF2 slurry dewatering

| Base membrane | Initial drainage time <sup>a</sup> , s | Drainage time <sup>a</sup> after usage, s | $R_M$ before usage, $\times 10^{11}$ 1/m | $R_M$ after usage, $\times 10^{11}$ 1/m | $RI^b$ in $R_M$ |
|---------------|----------------------------------------|-------------------------------------------|------------------------------------------|-----------------------------------------|-----------------|
| SP            | $132 \pm 3$                            | $930 \pm 182$                             | 1.9                                      | 14.0                                    | 6               |
| FP            | $91 \pm 2$                             | $633 \pm 38$                              | 1.4                                      | 9.6                                     | 6               |
| CA – 0.8      | $98 \pm 5$                             | $1268 \pm 150$                            | 1.5                                      | 19.0                                    | 12              |
| CA – 1.2      | $55 \pm 4$                             | $1223 \pm 198$                            | 0.8                                      | 18.0                                    | 20              |

<sup>a</sup> Time required to drain 800 mL of DI water<sup>b</sup> Relative increase in base membrane resistance coefficient after using it for CNF slurry dewatering

recorded relative absolute errors were 67 and 69% for CA-0.8 and CA-1.2, respectively. It took about 3300 s (55 min) and 3200 s (53 min) to drain 600 mL filtrate volume of water as opposed to the predicted time of about 1100 s (18 min) for the CA-0.8 and CA-1.2 membranes respectively (Fig. 4c, d).

To better understand the reason for the failed prediction for the CA membranes, an equal volume of filtrate of 2 kg/m<sup>3</sup> initial concentration of 90% fines CNF slurry was drained using SP, FP, CA-0.8, and CA-1.2. The formed filter cake on the base membranes was gently wiped off and the  $R_M$  of the already used base membranes was determined and compared

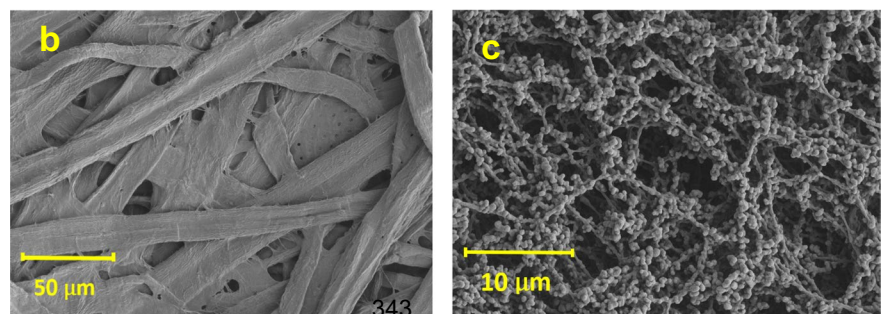
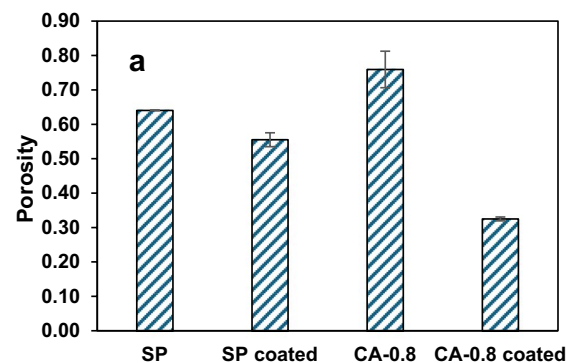
to that of the fresh base membranes. The relative increase ( $RI$ ) in  $R_M$  was calculated from the equation below:

$$RI = \frac{R_{M-used} - R_{M-fresh}}{R_{M-fresh}} \quad (7)$$

where  $R_{M-used}$  is the  $R_M$  determined for the base membrane already used for the CNF suspension filtration and  $R_{M-fresh}$  is the  $R_M$  determined for the fresh base membrane unused for CNF suspension filtration.

As shown in Table 3, the difference in drainage time between the used and fresh CA membranes was

**Fig. 5** Porosity measurement of 90-CNF coated and uncoated SP and CA-0.8 base membrane **a**. Scanning electron microscopy (SEM) micrographs of SP **b** and CA-0.8 **c** base membrane



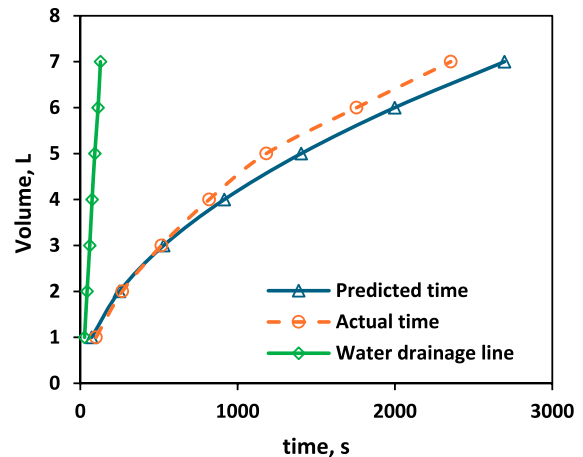
**Table 4** Filter cake resistance coefficient determination with different base membrane types

| Drainage system       | Filter cake resistance coefficient ( $\alpha$ ), $\times 10^{13}$ m/kg | Drainage time <sup>a</sup> , min |
|-----------------------|------------------------------------------------------------------------|----------------------------------|
| 90-CNF2 with SP       | $5.9 \pm 0.7$                                                          | $19 \pm 0$                       |
| 90-CNF2 with CA – 0.8 | $12 \pm 1.4$                                                           | $56 \pm 15$                      |
| 90-CNF2 with CA – 1.2 | $12 \pm 2.2$                                                           | $53 \pm 8$                       |
| BSKP2 with SP         | $1.2 \pm 0.2$                                                          | $11 \pm 2$                       |
| BSKP2 with CA – 0.8   | $1.8 \pm 0.0$                                                          | $8 \pm 0$                        |

<sup>a</sup>The time required to drain 600 mL filtrate volume of  $2 \text{ kg/m}^3$  initial concentration of the slurry

much higher than those of the used and fresh SP and FP membranes resulting in a large increase (*RI*) in  $R_M$  for the used CA membranes. The *RI* in resistance for the used CA membranes (CA-0.8 and CA-1.2) were 12 and 20 times higher than that of the fresh CA-0.8 and CA-1.2, respectively (Table 3). Those of SP and FP were both only six times higher. This suggests that there is considerable clogging of the pores of the CA membranes during CNF slurry dewatering resulting in higher  $R_M$  values. The results agreed well with the porosity measurement tests for the fresh and CNF coated base membranes given in Fig. 5a. The porosity of SP barely reduced after coating with CNFs whilst that of CA-0.8 reduced significantly from 0.8 to 0.3 indicating significant clogging of the pores of the CA-0.8 membrane as dewatering progressed. The SEM micrograph (Fig. 5b) depicts broad and lateral fibers with hidden pores for SP which are less susceptible to clogging by finer CNF fibrils whilst that of CA-0.8 (Fig. 5c) depicts widely open and exposed pores and hence susceptible to clogging by finer CNF fibrils.

The clogging was also evident when CA membranes were used for  $\alpha$  determination for 90% fines CNFs. As shown in Table 4, the  $\alpha$  values were higher than expected when CA membranes were employed, and therefore higher deviations were observed for the drainage prediction (Fig. 4c, d). However, those of BSKP remained constant irrespective of the base membrane used. The clogging of CA membranes with ultrafine fibrils results in a modified permeability and hence high  $R_M$  value, which seems to control the dewatering behavior. A similar observation was made by Dimic-Misic et al. (2013) for cellulose furnishes with CNFs addition. Therefore, clogging of the

**Fig. 6** Drainage prediction for 90% fines CNF slurry dewatering with SP as the base membrane with water drainage line

pores and the resulting modified permeability resulted in high  $R_M$  and erroneous  $\alpha$  values of the 90% fines CNFs and therefore the observed poor drainage prediction. These observations were not made for BSKP suspension with CA membranes, probably due to a lower number of ultrafine fibrils. Also, such observations were not made for CNF with SP and FP probably due to their hidden and tortuous pores hindering significant clogging.

To further verify the validity of the determined  $\alpha$  and its potential use for the design of filtration-based dewatering equipment, the determined  $\alpha$  for the 90% fines CNFs using the Büchner funnel and aspirator system was employed in Eq. (4) to predict its drainage on a lab scale sheet former with  $0.093 \text{ m}^2$  drainage area. As shown in Fig. 6, with SP as the base membrane, the determined  $\alpha$  value using the aspirator resulted in predicted drainage times that agreed well with the actual experimental values obtained using the sheet former. Also, the straight water drainage line obtained shows that the base membrane resistance does not change much with time from the drainage of just water. It further shows a faster drainage rate for DI water compared to CNF suspension whose drainage rate decreases over time due to the buildup of filter cake.

While the filtration model assumes that the drainage resistance of the media does not change during filtration, good predictions are still obtained when the resistance coefficient is obtained using the membrane or paper of interest. Even when the membrane is

clogging, the same dependence of time is obtained as in Eq. (3). Therefore, it is suggested to use the membrane or paper of interest to determine the resistance coefficient that pertains to the situation of use.

## Conclusions

This study examined the dewatering behavior of different fines percentage CNFs and characterized their filter cake resistance coefficient ( $\alpha$ ) using the constant pressure filtration equation to understand and hence predict the dewatering of CNF slurry. The dewatering of CNF suspensions was found to follow well the filtration theory with an  $R^2$  value greater than 0.98. Increasing the quality of CNFs from 60 to 90% fines resulted in an increase in  $\alpha$  values by around a factor of two indicating increased difficulty in dewatering. The increased  $\alpha$  values with increasing fines percentages can be attributed to the tight packing of the finer fibrils of the formed filter cake as dewatering progressed. The value of  $\alpha$  did depend on the initial slurry concentration of CNFs decreasing by around 50% as slurry concentration increased from 1 to 8 kg/m<sup>3</sup>. This result is likely caused by channels generated through the filter cake as a result of flocculation at higher concentrations during dewatering. These channels created enhance drainage and hence lower  $\alpha$  values at higher concentrations. The determined  $\alpha$  value for the 90% fines CNFs predicted well the dewatering rate of CNF slurry and gave good predictions on a lab scale sheet former. It was also shown that clogging of the pores of cellulose acetate membranes by CNF suspensions prolongs dewatering time due to associated higher base membrane resistance.

**Author contributions** BA carried out the experiments, undertook the data analysis, and wrote the first drafts of the manuscript. JW reviewed the data analysis, helped supervise the work, and reviewed the manuscript. MT helped set up the experiments using the sheet former and reviewed the manuscript. DB supervised the work, reviewed all drafts of the manuscript, suggested the modeling approach, and coordinated the funding.

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**Data availability** Data is provided within the manuscript.

## Declarations

**Conflict of interest** No authors have relevant financial or non-financial interests to disclose.

**Ethical approval** Not applicable.

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