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ORIGINAL RESEARCH

Impact of heat drying on the physical and environmental characteristics of the nanocellulose-based films produced via spray deposition technique

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Abstract Nanocellulose (NC) and NC-based composites have gained considerable attention in recent years due to their biodegradability and recyclability. Spray deposition has emerged as a potential technique to produce NC films due to the rapidity and simplicity of the process. The major hurdle regarding the efficient production of NC-based films is the drying process. The aim of the current study is to significantly reduce the drying time of NC films and consequently examine the impact of drying on their mechanical characteristics, barrier and environmental performance. The NC films produced by spray deposition were dried at 50 °C, 75 °C and 100 °C and their characteristics were compared with the films produced via the same method at ambient temperature (28 ± 2 °C). Heating the films in an oven up to 75 °C had a negligible effect on mechanical characteristics while slightly improving the barrier properties as compared with the ambient dried films. However, the dimensional stability was only achieved when the temperature was below 75 °C. Drying could be accomplished at the faster drying rate and the NC films found to have lower embodied energy in comparison with the conventional packaging materials.

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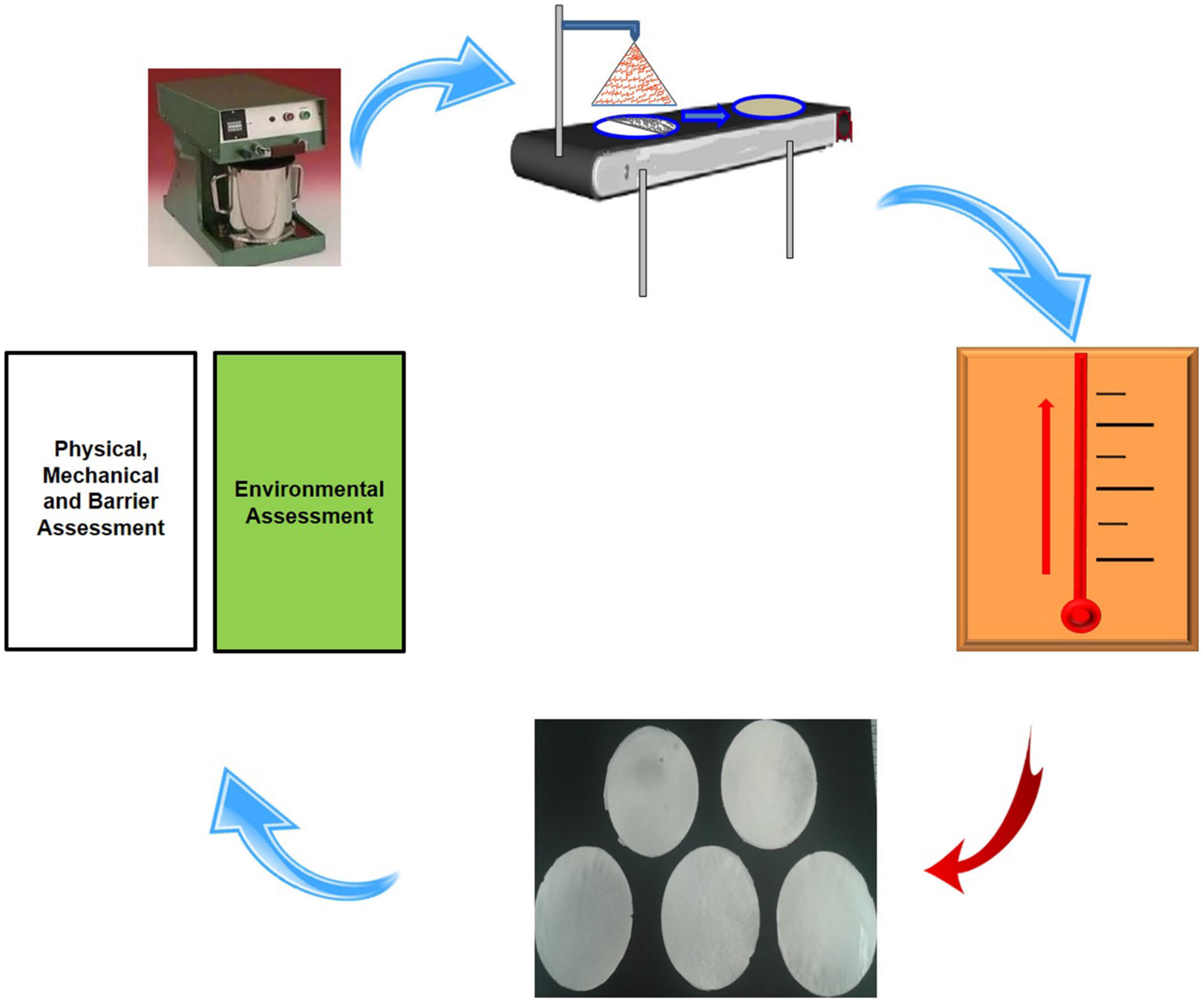
Graphic abstract



Keywords Embodied energy · Heat drying · Nanocellulose · Spray deposition

Introduction

Climate change has become a major issue in recent years due to escalating greenhouse gas emissions (Nadeem et al. 2017). Owing to the environmental concerns with the pervasive consumption of non-sustainable polymers in various industries, developing energy-efficient, biodegradable and recyclable barrier materials for packaging is of foremost concern. Following the current trend, the use of packaging materials is expected to grow at approximately 3% by 2022 (Pira 2019). Different materials such as plastics, glasses, aluminium and petroleum-based polymers have been used as packaging materials (Bugnicourt et al. 2013). Biodegradability and re-processability are major constraints in the utilization of these conventional materials. Virgin plastics often have excellent barrier properties, but, when they are recycled, their barrier properties decrease significantly (Hopewell et al. 2009).

Utilization of bio-based raw materials has surged in recent years, owing to their renewability and biodegradability. One class of these materials is nanocellulose (NC) that can be produced via mechanical, chemical or enzymatic processing to break down the cellulosic biomass into nano-fibres (Phanthong et al. 2018). Due to its non-toxic nature, high surface to volume ratio, inherent biodegradability and high compatibility with a number of materials, NC is considered as an ideal candidate for environmentally friendly packaging material (Mondal 2017). The films produced from NC are translucent, strong and have reasonable oxygen barrier properties (Shanmugam et al. 2018). The NC-based films are commonly produced either by casting or vacuum filtration (Shimizu et al. 2014). The casting process is time-consuming as it takes at least 3 days for films to dry, while the vacuum filtration suffers from problems with film handling and separation (Siró and Plackett 2010). Spray coating or spray deposition is a simple and rapid process and has been introduced as a potential alternative to conventional film production methods (Shanmugam et al. 2017, 2018). In brief, a uniform coat of NC is deposited on a substrate and the size of coating depends upon the suspension's concentration and the speed of the conveyor (Shanmugam et al. 2017).

The drying characteristics of NC films are yet to be investigated in detail. There are a limited number of studies available in the literature focusing on the rapid production of NC films. For instance, the casted NC solution was dried in a ventilated oven set at 35 °C to reduce the drying time for the production of films to 2 days (Plappert et al. 2018), however, the resultant films had shown wrinkles. Similarly, Ansari et al. produced the NC films (NC obtained from a never dried commercial pulp followed by enzymatic pre-treatment) via drying the suspension in a vacuum at 95 °C, but found that these films showed quite high moisture absorption rate (Ansari et al. 2014). Additionally, the NC films produced via vacuum filtration were heat-dried in an automatic sheet dryer at 95 °C, that required only 10 min for the films to dry (Varanasi and Batchelor 2013). Unfortunately, the production and drying processes used in existing studies are not viable for commercial scale production. Production of NC films via spray deposition seems to be an ideal candidate for industrial applications, but, currently, the NC films produced by spraying are dried at an ambient temperature taking up to 36 h of drying (Shanmugam et al. 2017). Drying at ambient temperature also requires a reasonable space for storing the films during the drying process (Shanmugam et al. 2017, 2018). Heggset et al. (2017) showed that the NC starts to degrade at 110 °C indicating the NC suspensions need to be dried below this temperature. In another study, the NC films were dried in a ventilation oven at a constant temperature of either 50 °C or 30 °C for 36 h. The results showed that the films dried at 50 °C had shrinkage with more wrinkles and defects (Zhang et al. 2017). The novelty of this research is to recommend an approach that could be attractive in future for scaling up the drying of NC films. To the extent of our knowledge no study has introduced a drying procedure to prepare NC films in a rapid and scalable manner.

The production of NC itself consumes much energy and is evaluated as one of the major obstacles for its utilization in the future. The minimum embodied energy emissions for the production of NC accounts approximately 1050 MJ kg^{-1} (Li et al. 2013). These limitations make this process neither cost-effective nor time-efficient for large-scale production. The aim of this study is to investigate the suitable conditions for the production NC films via spray deposition with the consideration of their dimensional stability. The NC films were dried at different conditions and their properties including mechanical, barrier and environmental were compared with the ambient dried films. The results here will provide a building block for large- scale production of NC-based films to replace the environmentally unfriendly packaging materials. Moreover, the embodied energies of the NC films dried at either ambient conditions or produced via heat drying were also compared with the conventional packaging materials.

Materials and methods

Film preparation

Fibrous Nanocellulose (KY-100S grade, 25 wt% solid content) used to produce films was purchased from DAICEL Chemical Industries Limited, Japan. The average diameter of NC fibres was approximately 70 nm with the average aspect ratio of 142 ± 28. The NC was added to water at 1.5 wt% solid content and then disintegrated for 15,000 revolutions at 3000 RPM in a 0.5 kW Messmer disintegrator Model MK III C (Netherlands). The suspension was then sprayed using a 0.56 kW Wagner spray system Model 117 (Australia) (Shanmugam et al. 2017, 2018). The sprayed NC was collected onto stainless steel plates carried by a moving conveyor belt (0.75 kW). The diameter of the circular plates used was approximately 160 mm while the orifice of the spray nozzle was 0.38 mm. The speed of the conveyor belt was fixed at 0.65 cm s^{-1}, while the distance of the nozzle tip from the steel plates was 30 cm. Sprayed plates were then finally transferred to an oven (Thermoline Scientific, TD 500 F Australia) for drying at 50 °C, 75 °C and 100 °C for different drying times, while ambient films dried at 28 ± 2 °C were used as the control. The lower temperature limit was chosen because it is slightly higher than the ambient temperature. Additionally, the upper temperature limit was selected based on the fact that the NC films start to degrade at 110 °C (Heggset et al. 2017).

Testing procedures

Basis weight and thickness

The basis weight was determined by weighing a standard area of five films for each sample. For the films prepared at ambient temperature, the basis weight was calculated after 4 h of drying in an oven at 105 °C to remove any traces of moisture. The thickness of five films of each sample was measured using a Thickness Tester Type 21 from Lorentzen & Wettre AB (Sweden) in accordance with Australian/New Zealand standard method 426 (AS/NZS-426 1994).

Moisture content

The moisture content at different drying conditions was determined according to ASTM D2216-10 (2010). The moisture content was calculated as:

$$W = \frac{M_{cws} - M_{cs}}{M_{cs} - M_c} \times 100 \quad (1)$$

where W is the moisture or water content (%), M_{cws} is the mass of the container and wet specimen, M_{cs} is mass of container and oven-dried specimen and M_c is the mass of container. During the drying process, the NC films were removed from the oven and the mass of the container was recorded at standard time intervals.

Mechanical characteristics

Three NC films were cut into five strips (100 mm test span × 15 mm width) and tested for tensile strength using an Instron universal testing machine (Model 5566, USA). The films were conditioned at 23 °C and 50% RH for 40 h before testing following the standard method, ASTM D 882-02 (2002). All the films were tested at a constant elongation rate of 10 mm min^{-1}. The tensile index (TI) was determined using the following equation; where, TI is the tensile index (N m g^{-1}), T′ is the tensile strength (kN m^{-1}) and R is the basis weight (g m^{-2}).

$$TI = \frac{T'}{R}. \quad (2)$$

Water vapour transmission rate and water vapour permeability

ASTM, E96/E96M-05 (2005) was used to determine the moisture barrier performance of the NC films (n = 5). Prior to testing, the films were kept in an oven for at least 4 h at 105 °C to remove any traces of moisture. At first, for each sample, about 40 g of anhydrous calcium chloride ($CaCl_2$) weighed in three non-corroding cups while the two cups were used as blank. The desiccant must be completely dried in an oven prior to the test at 200 °C to remove any moisture content. The films from each sample were then sealed on the open side of the cups forming an airtight assembly. The test was conducted at 23 °C and 50% RH and the assemblies were weighed at regular time intervals. Finally, the water vapour transmission rate (WVTR) was calculated from the slope of the line between the weight and time. The WVTR of NC films were normalized with the thickness of the films and converted into water vapour permeability (WVP). The WVTR (g m^{-2} d^{-1}) can be calculated as follows; where, $\frac{G}{t}$ is the slope of the straight line (g h^{-1}) and A is the test area (m^2).

$$WVTR = \frac{G}{t \times A} \quad (3)$$

The permeance (g Pa^{-1} s^{-1} m^{-2}) for the films can be determined as:

$$Permeance = \frac{WVTR}{S(R1 - R2)} \quad (4)$$

where S is the saturation vapour pressure at a test temperature (mmHg, 1.333×10^2 Pa), while $R1$ and $R2$ are the relative humidities of the source and the vapour sink expressed as a fraction.

The relative humidity and temperature are actually the average values measured during the test. Based on the standard, the relative humidity (R2) values are nominally 0% for the desiccant (completely dried) and 100% for the water (ASTM 2005).

Finally, the average permeability (g Pa^{-1} s^{-1} m^{-1}) of the films can be determined as:

$$Average\ Permeability = Permeance \times Thickness. \tag{5}$$

Morphology

The effects of different drying temperatures on the morphology of NC films were studied using a scanning electron microscope (Nova SEM450, FEI, USA). The NC films were first iridium coated and the images were taken at an accelerating voltage of 5 kV and 500 × and 25,000 × magnifications. The surface topography of the NC films (either ambient or heat dried) at microscale with 5 × magnification was evaluated by optical profilometer (Olympus LEXT OLS5000 Laser Confocal Microscope, Japan).

UV–vis spectroscopy

The effects of different drying temperatures on the optical properties were determined by transmission spectra of the heat and ambient dried NC films using a Agilent Cary 60 UV–Vis Spectrophotometer in the wavelength range of 200–800 nm. Transmittance at the wavelength of 200–700 nm was used as a measure of film transparency.

Embodied energy

Environmental aspects of the optimized films were assessed and compared with the conventional packaging materials to ensure the environmental feasibility of the product. NC is still under rapid development, hence there are many uncertainties regarding its technical system. There are a number of challenges that exist while assessing the relatively new products including unavailability of the data, comparison with existing materials or system and factors effecting the scale-up etc. (Hetherington et al. 2014). The data for the calculation of embodied energy for the production of NC films was acquired from different sources. The main source was the Ecoinvent database (Wernet 2019) (version 3.6), including the inventory data for a number of materials and processes. The other useful source was a publication from Arviddson et al. (2015) while the source of the embodied energy data for PET films was a published report from Cambridge University (Jackson 2006). Cradle-to-gate analysis using Ecoinvent 3.5 (Switzerland) (Wernet 2019) was performed to determine the embodied energy. The assumptions made for this analysis are listed in Table 1.

Statistical analysis

The results were statistically analyzed by Minitab 17 software (Minitab Inc., State College, USA) using Student's *t* test and the data were expressed in terms of 95% confidence intervals. In all evaluations, $P < 0.05$ was considered as significant.

Results and discussion

Effect of temperature on moisture content

There are a few studies available on the production of NC films via spray deposition, however, it required at least 24 h for the films to dry at ambient conditions (Beneventi et al. 2014; Shanmugam et al. 2017, 2019). The drying of films at ambient conditions is regarded as a huge constraint in the commercialization of the process. The use of spray deposition has reduced the time of operation to a significant extent; however, the research conducted to date has not investigated a rapid drying procedure. Therefore, we decided to reduce the production time of the films while drying them at a faster rate in an oven. Measuring the moisture content is critical in optimizing the drying time and temperature in the heat drying process. If the films are heat dried for too long, wrinkling and shrinkage might happen, while the extended drying increases the energy consumption. The moisture content for all films was determined following the standard method ASTM D2216-18, and the results are displayed in Fig. 1. As this Figure illustrates, the moisture removal

Table 1 Assumptions in determining embodied energy

Process	Assumptions
Materials	1. The pulp used to produce NC fibres has an embodied energy of 50–70 MJ kg^{-1} (Arvidsson et al. 2015)
	2. The CNF fibres for the spray deposited films were assumed to be produced from cotton pulp via no pre-treatment route
	3. The total embodied energy was 92 MJ kg^{-1} for PET films, while 70 MJ kg^{-1} for recycled PET films respectively (Jackson 2006)
Production	1. Heat losses are negligible
	2. All films (NC, recycled NC and PET) weighs 2 g (16 × 16 cm)
	3. Approximately 300 films were produced via spray depositing in 1 h
	4. Approximately 12 films could be produced with 1 L of suspension
	5. Approximately 200 films could be heat dried in a 2 kWh oven in one batch
	6. Around 100 NC films could be dried at ambient conditions in a 780 mm × 390 mm ventilated block. The block was constructed using green tiles (Nadeem et al. 2017)
Other	1. Transportation emissions of NC fibres were considered as negligible due to two reasons i.e. firstly due to emerging nature of NC and secondly by assuming that NC fibres were shipped in bulk
	2. The source of electricity is assumed to be from biogas requiring 1.62 MJ to produce 1 kW (Wernet 2019)

from all films generally followed a declining drying rate over time for all drying temperatures. The drying of the films occurred in three stages. During the first stage, surface evaporation dominates (Warson and Finch 2001), while during the second stage, the rate of evaporation significantly decreased. In the final stage, water escapes by vapour diffusion through the coalesced films. The dry layer was first formed at the surfaces and continued to grow thicker unless the film was fully dried (Heldman 2012).

The total drying time for an ambient dried film was in days, whilst for heat dried films, an increased drying rate was observed. Approximately, 95% of the moisture has been removed in about 1.5 h at a temperature of 100 °C, while it took 2 and 4 h for drying at 75 °C and 50 °C, respectively. This finding when compared with ambient dried films, where drying took more than 24 h, indicates that the heat drying can speed-up the production of the NC-based films. For ambient dried NC films, the higher humidity of the air (55%) was the main contributor for the longer drying times. However, for the heat-dried films, a dehydrating oven with an air exhaust control was used. When the air exhausts remained fully opened during the drying of the films, the humidity inside the oven was quite negligible. For example, at 100 °C the relative humidity inside the oven was close to 0%, while at 75 °C, it was almost negligible (approximately 1–2%). However, the relative humidity inside the oven at 50 °C was in the range of 8–10%. The image of the top view of the oven is provided in the supplementary information. Additionally, the evaporation rate of water is strongly dependent on the surface i.e. the materials having porous and higher specific surface area can cause the water to evaporate rapidly from the surface (Yu et al. 2015). This study suggests that the spray deposited NC films could be heat-dried for a shorter duration as compared with the ambient dried films, however, impact of heat drying on the films' properties is really important and are discussed in the subsequent sections.

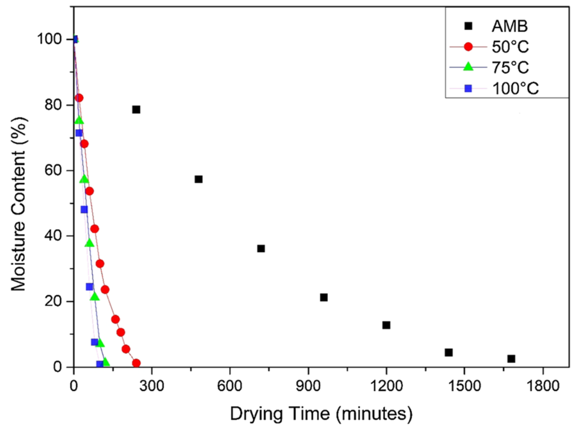


Fig. 1 Moisture content for the NC films dried at 50 °C, 75 °C, 100 °C and ambient temperature

Physical characteristics

As Fig. 2 shows, the NC films dried at ambient temperature and 50 °C, 75 °C and 100 °C had similar physical appearances. The dimensional stability in the films dried at 100 °C and 75 °C was not consistent as the majority of films dried at 100 °C and some of the films dried at 75 °C experienced shrinkage which is evident in the images (Fig. 2c) and (Fig. 2d), captured by a camera and optical profilometer. No sign of shrinkage or wrinkling was observed in the films dried at 50 °C. High temperature although had the advantage of fast drying (i.e. ~ 2 times faster at 100 °C compared with 50 °C) but the dimensional stability

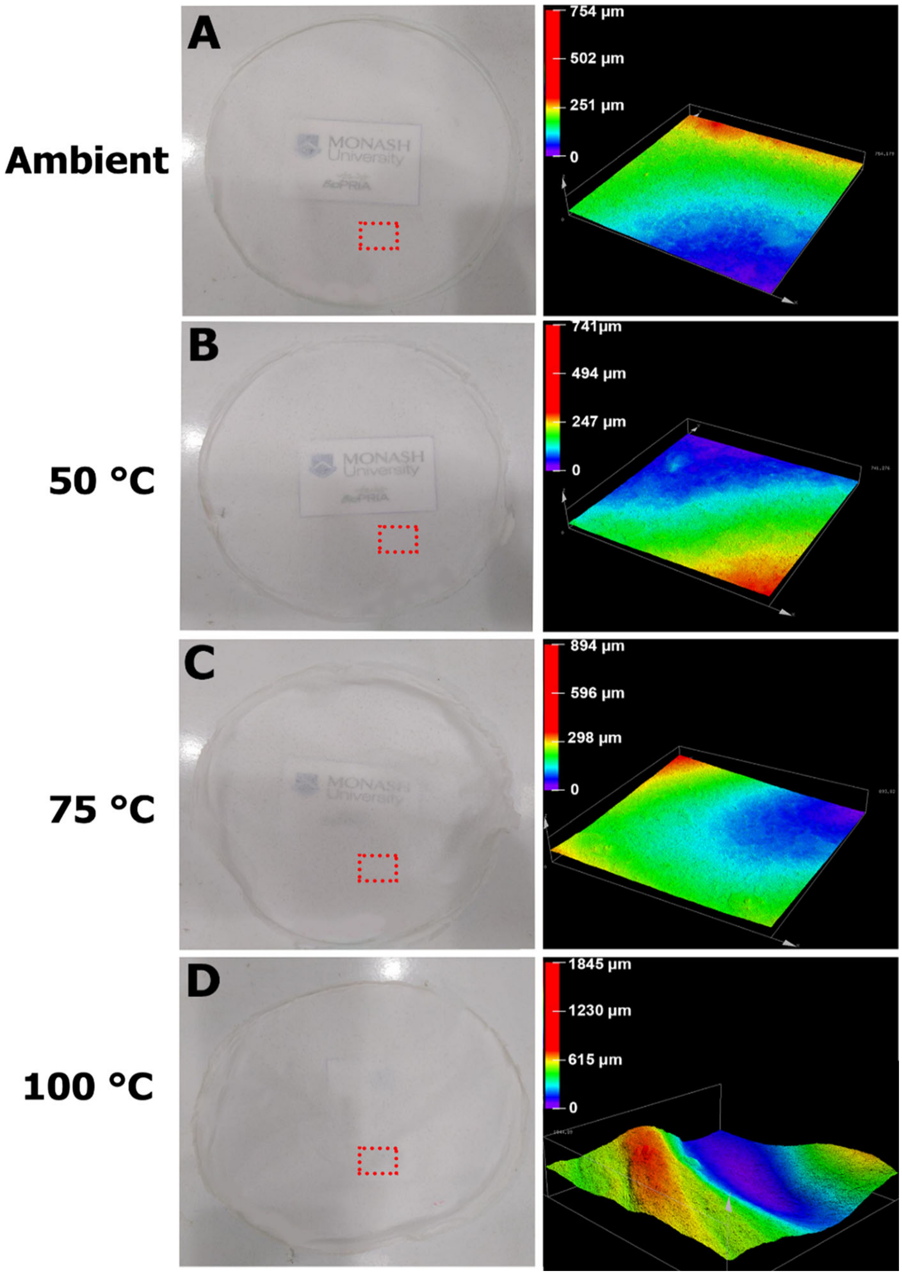


Fig. 2 NC films with the dimensions of 16 cm × 16 cm dried under varying temperatures of **a** Ambient, **b** 50 °C, **c** 75 °C and **d** 100 °C. Topography images in the right column

could not be guaranteed. Heggset et al. (2017) suggested that the NC films start to degrade at 110 °C which explains the deformation of NC films when dried at high temperatures in this study (Heggset et al. 2017).

The SEM images showed that the NC films produced at 100 °C acquired a fibrous structure consisting of micro and nanosized fibres (Fig. 3d). Unlike the NC film at 100 °C which possessed complete fibrous structure, the films dried at 50 °C (Fig. 3b) and 75 °C (Fig. 3c) had fibres compacted in the NC matrix, formed from the NC fibres fused together. Similarly, the ambient dried film (Fig. 3a) mainly consisted of NC matrix with few micro fibres embedded in its structure. Moreover, to some extent, the agglomeration of the NC fibres was also observed for the films produced at 100 °C. The NC images at different magnifications are also provided in the supplementary data (Figure S1-S4).

The transparency of the ambient and heat dried NC films was investigated using a UV–vis spectrophotometer and the results are displayed in Fig. 4. The films produced onto stainless steel plates formed a nanofibrillar network during water evaporation. This is a complex process including mass transfer and phase separation competing with each other (Sun et al. 2018). As the NC fibres are quite larger in size, they tend to tangle together, forming clumps and making it hard for light to transmit through the films. The transmittance was almost the same for the films dried at ambient temperature and at 50 °C. The lower temperature enhances the inter-fiber bonding of the

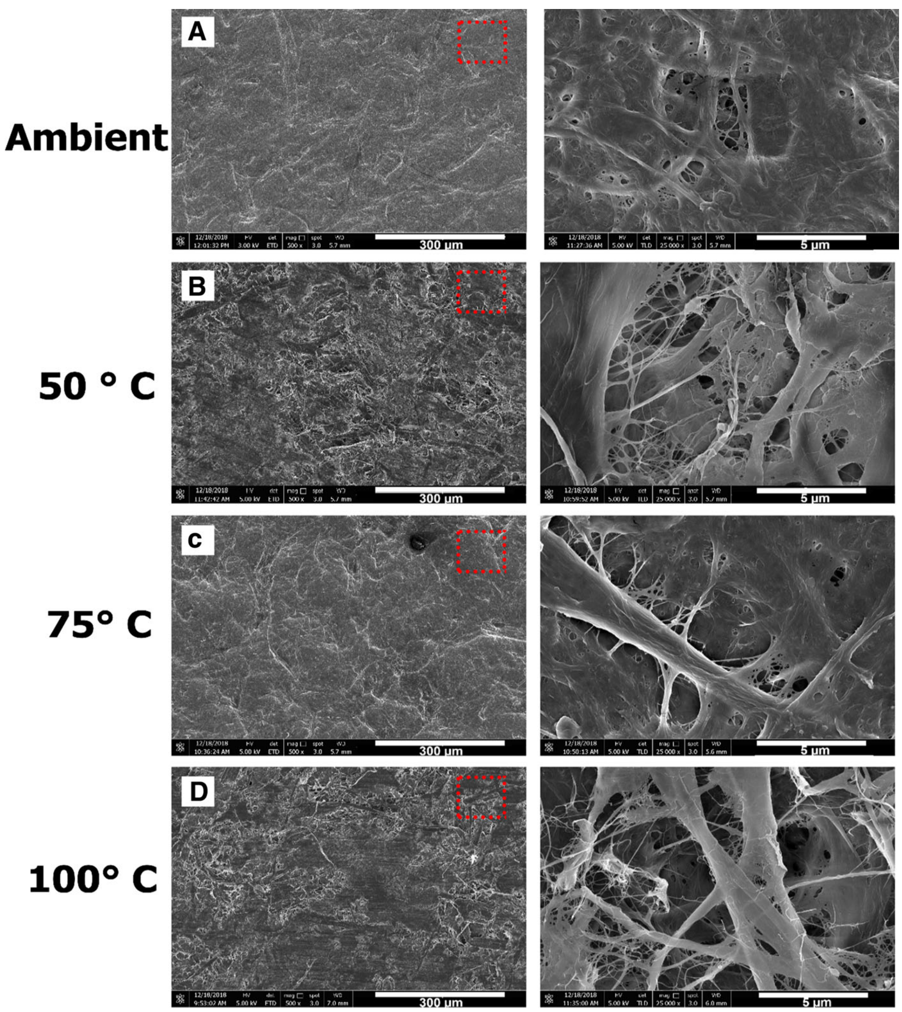


Fig. 3 SEM images (LHS: 500 ×, RHS: 25,000 ×) of the NC films dried at **a** ambient, **b** 50 °C, **c** 75 °C and **d** 100 °C

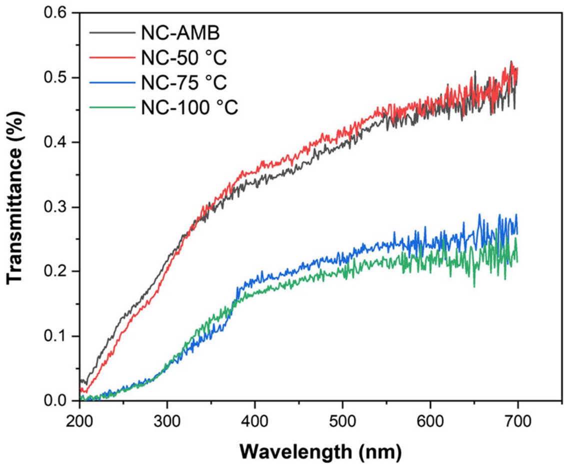


Fig. 4 Effect of temperature on the transmittance of the NC films

fibres and results in less dense structures (Shimizu et al. 2014). However, increasing the temperature beyond 50 °C has reduced the transmittance of the films by 50–60%. Temperatures above 100 °C may cause some structural changes to NC films (Zhang et al. 2017). As NC suspension was dried, a number of air voids remained inside the sheet and these voids are directly proportional to temperature i.e. higher the temperature, the greater the number of air voids (Hsieh et al. 2017). These air voids also cause scattering of light and reduce the transparency.

The Daicel NC used in this study is speculated to be cotton derived produced via high pressure homogenization (Hideno et al. 2016; Jang et al. 2015). The mean diameter of these commercial grade NC fibres was around 73 nm (Varanasi et al. 2013), while their suspension showed a zeta potential of − 22.3 mV (Raj et al. 2017). It is noteworthy that the transmittance characteristics of the NC films will change considerably with the composition of the NC fibres. Lignin containing NC fibres and thermomechanical pulp exhibited less transparency while lignin free fibres and bleached hardwoods were highly transparent (Ang et al. 2019; Spence et al. 2010). Similarly, the process involved in the production of the fibres may also effect the optical characteristics. For example, the films produced prepared by drying and compression were much more densely packed, but these films were not optically transparent, while the films produced by filtration showed quite high transparency (Dufresne 2013). Additionally, homogenization may cause the defibrillation of the fibres to a considerable extent and result in higher opacity (Spence et al. 2010). Techniques such as polishing to reduce surface scattering, formation of composites, and tuning the chemistry of the material have been used to improve the optical properties of the NC films (Niu et al. 2018).

Basis weight and thickness

Figure 5 represents the average basis weight and the thickness of the films at varying drying temperatures. The heat of drying had no effect on the basis weight and thickness of the resultant NC films. The films had an average basis weight of 37 ± 0.9 g m^{-2} with the thickness of 64–74 µm. The basis weight could be increased by decreasing the velocity of the conveyor or increasing the suspension concentration (Shanmugam et al. 2018). Although there was no statistically significant change in the basis weight of the films, a decreasing trend for the weight loss and thickness is observed with the increased evaporation rate or increased drying temperature. These findings are in accordance with the results reported in the literature (Greener et al. 2018; Vasconcellos and Farinas 2018). The basis weight was also found to be directly proportional to the thickness of the resultant films (Shanmugam et al. 2017).

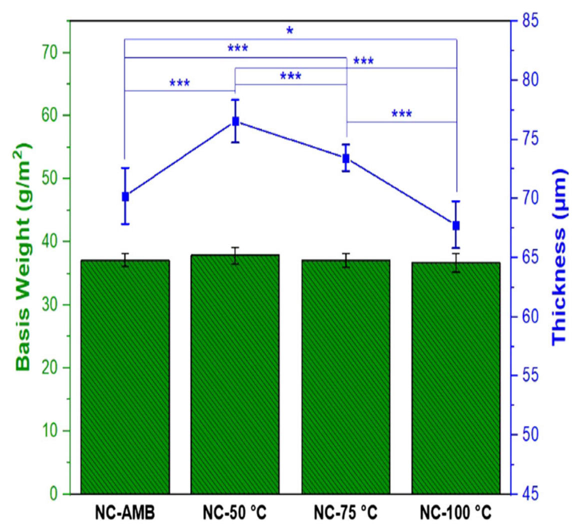


Fig. 5 Effect of temperature on the basis weight and thickness of the NC films. Values represent the confidence intervals, n = 5

Mechanical characteristics

The tensile index results are shown in Fig. 6. The average tensile index for the fully dried NC films was found to be 53 ± 5 Nm g^{-1} for average basis weight of 37 ± 0.9 g m^{-2}. No statistically significant difference was observed between the tensile index results from the heat-dried films and the ones dried at ambient temperature, indicating the heat drying had no significant effect on the mechanical characteristics of the NC films. Beneventi et al. (2014) produced NC films via spray depositing and their films showed quite similar results with average tensile index values in the range of 45–60 Nm g^{-1} for the basis weight of 30 ± 5 g m^{-2}. The NC films produced by Shanmugam et al. (Shanmugam et al. 2018) via the same technique have shown a tensile index of approximately 65 Nm g^{-1} for the basis weight of ~ 45 g m^{-2}. However, it is worth mentioning that the decrease in basis weight could reduce the films' tensile properties (Beneventi et al. 2015). Moreover, some of the NC films produced by vacuum filtration have shown the same tensile index values as the spray deposited films (Shanmugam et al. 2017, 2018). Recently, Ang et al. (2019) reported that the mechanical characteristics of the films can be improved by treating the fibres in a homogenizer or refiner. The tensile index values achieved by above-mentioned treatments was in the range of 100–120 Nm g^{-1} for the basis weight of ~ 60 g m^{-2}, however, these aspects will adversely affect the energy consumptions.

Cellulose at the nanoscale is connected by hydrogen bonds formed between hydroxyl groups, resulting in a high-strength material. At higher temperature of around 110 °C, the degradation of the fibres starts (Heggset et al. 2017). As 100 °C is near to the temperature where the initiation of fibres' degradation might occur, a decreasing trend in the tensile index values of NC films is observed. The NC fibres usually remained stable at temperatures less than their degradation temperature, resulting in tensile index values of NC films in a quite similar range at 50 °C and 75 °C (Heggset et al. 2017). Sehaqui et al. also suggested that for the oven drying (below degradation temperature), the mechanical properties can be slightly improved over ambient drying with the shorter preparation time (Sehaqui et al. 2010).

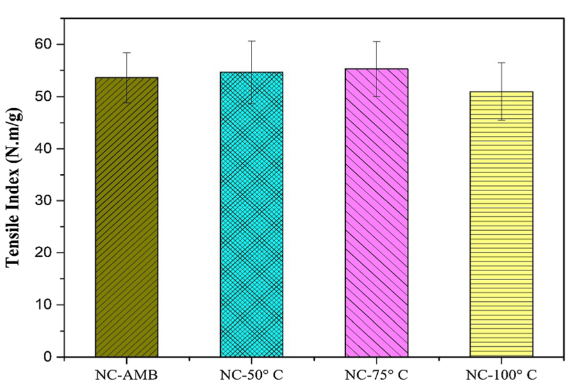


Fig. 6 Average tensile index of the NC films at varying temperature. Values represent the 95% confidence intervals, n = 5

Moisture transmission properties

The moisture barrier performance of the NC films is shown in Fig. 7. The WVP of heat-dried NC films showed a slight decrease in values compared with the films dried at ambient temperature. The higher WVP values of the films produced at ambient conditions are attributed to the drying of the suspensions at high relative humidity (Mo et al. 2014). The hydrogen bonds were formed between the hydroxyl groups of NC fibres chains and water. The water acted as a plasticizer and resulted in swelling of the films. The swelling phenomenon is directly proportional to the relative humidity of the films i.e. higher the humidity of ambient air, more the swelling of films. As a result, water vapour transported more readily across the

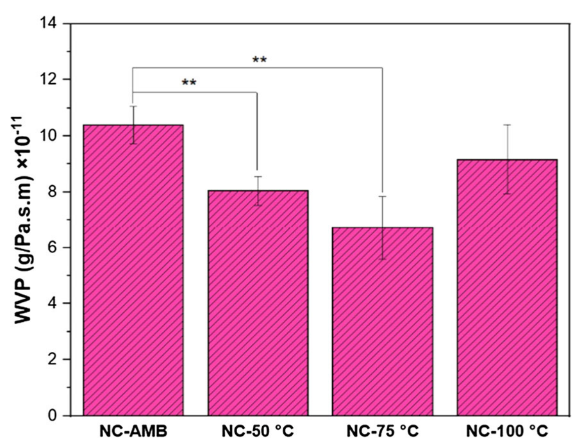


Fig. 7 Moisture barrier properties of the NC films **a** Water vapour transmission rate, **b** water vapour permeability. Values represent the 95% confidence intervals, n = 4

swollen film than the oven-dried film. The moisture barrier performance of the films dried at 75 °C was slightly better in comparison to the films dried at 50 °C. However, the observed differences were not statistically significant. The water absorbed for the drying of the films, while keeping the relative humidity constant decreased with the rise in temperature indicating that less water was bound at higher temperatures (Chinma et al. 2015). This is an indication of lower moisture transmission rate for the films dried at 75 °C. Moreover, as affirmed from the SEM images, the NC films produced at 50 °C and 75 °C mainly consisted of a compacted matrix with a few pores in the structure. This resulted in a lower water vapour permeability for NC films produced at 50 °C and 75 °C as compared with the films produced at 100 °C and at ambient conditions. The increased WVP values for the films dried at 100 °C might be attributed to the initiation of degradation of NC fibres (Heggset et al. 2017). Additionally, as observed from SEM images, agglomeration of fibres took place to some extent at 100 °C, which may reduce the tortuosity of the path (Wolf et al. 2018), increasing the moisture transmission.

The WVP of the films produced in this study were higher than the conventional packaging materials owing to the hydrophilic nature of the NC (Nair et al. 2014). However, this performance can be improved by compositing NC with other compatible materials (Garusinghe et al. 2018). The moisture transmission characteristics for ambient dried NC films was in the range of the values reported by Rodionova et al. (2011). Additionally, Shanmugam et al. (2019) also reported quite similar results for the WVP values of the NC films.

Environmental aspects

Excessive energy in the production of materials is a serious environmental concern that needs to be addressed. The reduction in the embodied energy of the materials is extremely important to reducing environmental damage. For this reason, the embodied energy of the NC films was calculated and compared with the conventional polyethylene terephthalate (PET) films. The total energy emissions per virgin and recycled NC films are displayed in Table 2. Results indicate that the embodied energy to produce a virgin NC film is slightly higher than that of conventional PET films. These high energy emissions are due to the production and extraction of NC itself that utilizes energy-inefficient process and making it unappealing for future commercialization.

Energy for production of NC fibres

The most common method to produce NC is mechanical fibrillation that requires up to 30,000 kWh tonne^{-1} (Nakagaito and Yano 2004). Using various pre-treatment methods such as carboxymethylation, TEMPO-mediated oxidation, and enzymatic hydrolysis, the energy requirements has been significantly reduced to approximately 2000 kWh tonne^{-1} (Isogai et al. 2011). However, long oxidation time, toxicity, corrosiveness, cost and difficulty in catalyst recovery are a few of the major limitations in using above-mentioned pre-treatment methods (Lavoine et al. 2012). The high-energy consumption could be overcome by either utilizing alternative energy-efficient processes, recycling or incorporating the NC with waste streams and environmentally benign materials. Recently, Varanasi et al. (Varanasi et al. 2018), investigated that producing the NC from vegetable sources could be a more energy-efficient and sustainable method. The mechanical treatment of vegetable residues has reduced the energy consumption to approximately 8500 kWh for the production of 1 tonne of NC, while at the same time eliminating the use of any chemicals. This could be a huge advance since producing the NC from waste would further reduce the energy requirements.

Embodied energy and oven-dried NC films

Additionally, the drying of NC films in an oven has increased energy usage. The life cycle analysis of NC and PET films is illustrated in Fig. 8. Initially, the virgin NC films (either heat dried or ambient dried) have higher embodied energy compared with the PET films. It would be highly energy inefficient if the virgin NC films were being disposed-off without recycling. When recycled, the NC films have significant advantages over the PET films, as the recycling process requires much less energy.

Table 2 Total embodied energy per NC films, (A) Ambient dried (B) 50 °C

Process/material	Embodied energy MJ kg^{-1}	Embodied energy per film (MJ)
(A)		
Pulp	50 (Arvidsson et al. 2015)	0.16
Pulp to CNF	72 (Arvidsson et al. 2015)	0.24
Disintegration of CNF suspension	0.0675 (Wernet 2019)	0.0037
Spray pump	0.0756 (Wernet 2019)	0.0042
Belt conveyor	0.101 (Wernet 2019)	0.0056
Ambient drying	0.64 (Nadeem et al. 2017)	0.028
Total		0.44
Recycling		
Embodied energy (virgin CNF film)		0.44
Disintegration		0.0037
Spray pump		0.0042
Belt conveyor		0.0056
Ambient drying		0.028
Total R1		0.48
Total R2		0.52
Total R3		0.56
(B)		
Pulp	50 (Arvidsson et al. 2015)	0.16
Pulp to CNF	72 (Arvidsson et al. 2015)	0.24
Disintegration of CNF suspension	0.0675 (Wernet 2019)	0.0037
Spray pump	0.0756 (Wernet 2019)	0.0042
Belt conveyor	0.101 (Wernet 2019)	0.0056
Heat drying		0.06
Total		0.47
Recycling		
Embodied energy (virgin CNF film)		0.47
Disintegration		0.0037
Spray pump		0.0042
Belt conveyor		0.0056
Heat drying		0.06
Total R1		0.54
Total R2		0.61
Total R3		0.68

Embodied energy and recycled NC films

The ease of recyclability and biodegradability are the distinguishing features of NC films. The dried CNF films could easily recycled in accordance with TAPPI standard T205 SP-02. Briefly, the contributing steps in producing a recycled NC film are disintegration followed by spraying and drying. Recently, it has been shown that recycled NC films could retain much of its barrier and mechanical properties (Shanmugam et al. 2019). In contrast, the recycling of conventional PET films is either difficult or requires high energy consumption (Jackson 2006). Thus, NC films when recycled, regardless of the production method, have shown a lower increase in embodied energy in comparison with the recycled PET films. After the

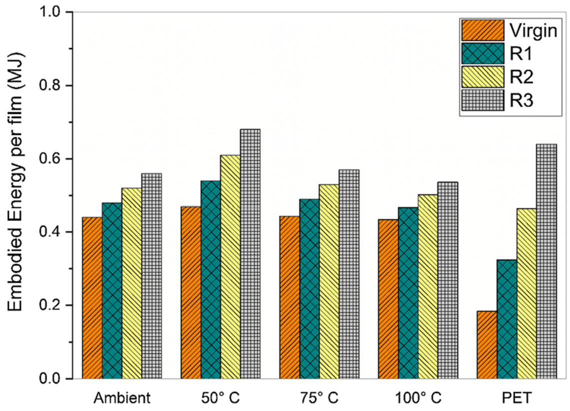


Fig. 8 Life cycle analysis comparison of the NC and conventional PET films

third recycling, the embodied energy for ambient dried and heat dried NC films at 75 °C and 100 °C have shown lower embodied energy as compared with the recycled PET films. However, in order to achieve the same result for the films dried at 50 °C, a higher number of recycling times are needed. Thus, producing the NC films (considering its recycling aspects) is likely to be a more viable and energy-efficient process now, with the NC advantages increasing in the coming years.

Conclusion

Rapid and environmentally friendly production of NC-based films is demanded for packaging applications to minimize the petroleum-based wastes. The heat dried spray deposited NC-based films can be a promising candidate for such purpose. Herein we investigated the effects of heat drying on the properties of the NC-based films produced via spray deposition process. Drying has been an issue in commercialization of this production method and the present results demonstrate that the films could be heat dried in approximately 2 h in comparison with more than 36 h needed under ambient temperature. This study also shows almost similar mechanical characteristics with slightly better moisture barrier performance as compared with the ambient dried films. Moreover, the heat drying has a significant effect on the energy emissions of these films, but due to its recycling advantage, the NC films have shown an overall low embodied energy. The results here will provide a building block for large-scale production of NC-based films to replace the environmentally unfriendly packaging and other high-energy consuming materials.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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