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A Study on the Preparation, Characterization, and Application of Spray-Coated Cellulose Nanofiber Films

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Abstract: Spraying cellulose nanofibers (CNF) on a polished stainless-steel plate is a novel approach to the fabrication of free-standing cellulose nanofiber film. Recently, free-standing cellulose nanofiber film has been attracting attention as an alternative to synthetic plastic film. Free-standing/self-standing CNF film can be used as a potential barrier, packaging application, membranes for wastewater application, fabrication of biomedical film for wound repair, and other applications in the fabrication of functional materials. To speed up the production of free-standing CNF film, the spraying process is a considerably intensified method for large-scale production of the film in a rapid manner. Spraying CNF on the stainless-steel plate produces unique surfaces, namely a rough surface exposed to air and a smooth surface on the steel surface. The smooth surface of the film was shiny and provided a platform for utilizing this smoothness for fabricating functional materials such as substrates for flexible electronics and solar cells, etc. This paper summarizes the production of free-standing CNF film via spraying, along with its characterization and application.

Keywords: Spraying; Cellulose nanofibers; Free-standing films; Air permeance; Uniformity; Thickness mapping; Water vapor permeability

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1. Introduction

Plastic pollution is one of the serious threats to the environment in the current scenario. Packaging is the main source of this plastic pollution and it is vital to replace synthetic plastics with biopolymers. Biopolymers are good alternatives for synthetic plastics as it has characteristics such as biodegradability, eco-friendliness, and good mechanical and barrier properties for the development of various functional materials [1]. Recently, cellulose nanofiber (CNF) is gaining a predominant place in the list of biopolymers [2].

Cellulose is the most important bio-renewable and biodegradable biopolymer that is plenty available in nature and acts as an excellent feedstock for the development of various sustainable materials on an industrial scale production [3]. In the past decade, cellulose nanofiber has been used as one of the pioneer feedstock for the development of various functional materials. It is produced by the disintegration and delamination of cellulose fibrils from pulp produced from a variety of green sources like wood, potato, hemp, and flax, with its dimension diameter ranging from 5 to 60 nm and length in several micrometers [4]. Moreover, the smaller dimension in cellulose nanofibrils gives the larger surface of CNF, thus there is a great opportunity for developing more functional materials for various applications [4,5].

The films made from CNF have various outstanding mechanical, optical, and structural properties and these properties contribute to the fabrication of various functional materials such as cellulose nanocomposite [4], micro-fibrillated film [6], inorganic nanocomposite [7], organic transistors and conducting materials [8], and immunoassays and diagnostics materials [9]. Moreover, nano-fibrillated cellulose can be easily tailored in terms of its surface properties and mechanical characteristics. As a result, it is used in the field of photonics surface modifications, nanocomposites, biomedical scaffolds, and optoelectronics [10]. Due to the barrier and colloidal properties of CNF, it is widely used in paper making, packaging, and coatings to enhance its barrier surface, as well as in automotive industries [11]. CNF sheets could also be one of the most promising high-performance functional materials potentially used as filters [12], adsorbents, catalysts [13], cell culture substrates, thermal insulators, and drug carriers [14].

On top of the mentioned properties, these materials are biodegradable and recyclable. Hence, they have the potential to replace some of the synthetic polymeric materials that cause serious environmental problems [1]. However, one persisting problem lies in that CNF film preparation had high energy consumption and high time consumption in the fabrication [15]. CNF films are prepared using vacuum filtration, followed by casting and spray-coating. However, these processes are time-consuming and the films prepared have poor basis weight and thickness [15,16]. Even though these processes could be efficient in the way of producing better quality films, it has constraints in the scaling-up process in large-scale production for technology transfer and commercialization of the free-standing films [15].

In the vacuum filtration method, the CNF film preparation required a high dewatering time, which shows a major constraint for an industrial-scale process. Furthermore, Varanasi and Batchelor [17] reported the rapid preparation of nano-fibrillated sheet using a British handsheet maker in 10 minutes. However, they achieved only a mass per unit area of 57.4g/m^2 and a thickness of 68.9 ± 8.90 μm. Therefore, the current investigation was motivated to develop a rapid and scalable spray-coating technique to produce the nano-cellulose film to replace the time-consuming conventional techniques for cellulose film [18]. Beneventi *et al.* [19] reported the spray-coating of the micro-fibrillated cellulose on the nylon fabric to prepare the nano paper with a maximum mass of the film of 124 g/m^2 with a conveyor speed of 0.5 m/min. However, it failed to explain the uniformity of the film through the thickness distribution and surface morphology. This paper explores the rapid preparation of CNF film using a developed lab-scale spray-coating system to produce high basis weight film.

2. Free-standing CNF film fabrication via spray-coating process

Spraying cellulose nanofibers on the polished stainless-steel plate is a rapid and novel process for the fabrication of CNF wet film [20]. Previously, spraying micro-fibrillated cellulose on 3D (three-dimensional) brass metal was attempted. However, cracks and shrinkage on the film were formed. This was the inspiration to develop a spray-coating process to fabricate CNF film [21]. An improved process involves spraying micro-fibrillated cellulose on the fabric surface, followed by vacuum filtration to remove the excess water in the wet film [19]. The concept of spraying nanofibers has been formed from these concrete works. In addition, spraying provides contour coating and contactless coating with the solid surface, thus the surface topography and morphology of the solid surface do not influence the coating process [15]. It has been reported that the spraying process produces CNF film with high basis weight without any changes in the operation time [20]. The concept of spraying CNF on the fabric and paper substrates was already developed for films and barrier coating on the paper surface [19,22]. This study reveals the spraying of CNF on the stainless-steel plates for the fabrication of films.

The following experimental system on spray-coating to fabricate CNF film was developed, as shown in **Figure 1**.

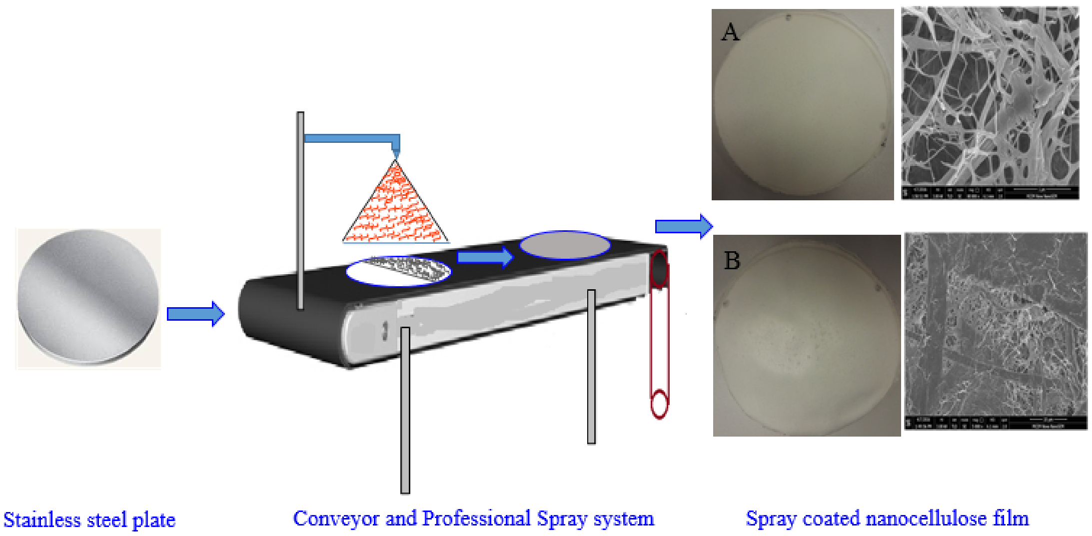

Figure 1. Experimental setup for lab-scale spray-coating system for preparation of nanocellulose (NC) film. (A) Rough surface of the NC film (B) Smooth surface of the NC film

Figure 1 shows the experimental system for spraying CNF on the stainless-steel plate that is kept on the conveyor. In this experimental system, there are two important parameters for tailoring the properties of CNF film. CNF suspension consistency and velocity of the conveyor are the parameters used for tailoring the thickness and basis weight of the films. At a constant velocity of the conveyor, varied CNF suspension can be used in the spraying operation to produce the CNF film. Normally, spraying low-percentage CNF suspension produces film with the lowest thickness and basis weight; whereas film with high basis weight and thickness can be fabricated by spraying high-percentage CNF suspension on the polished metal surface. Similarly, the CNF suspension concentration can be fixed, and varying the velocity of the conveyor can tailor the thickness and basis weight of the film. It means that film with high basis weight and thickness was fabricated at the lowest velocity of the conveyor, during a high amount of suspension was deposited and fibers were concentrated to form high basis weight CNF film. Conversely, thin CNF films were fabricated by spraying CNF suspension on the metal plate at high velocity of the conveyor. In this case, less amount of fibers were deposited on the steel plate to form a thin CNF film due to the fast movement of the conveyor. Apart from these important parameters in the spray system to show the effect on CNF film's properties, the spray distance between the spray gun to the base surface, spray nozzle, and spray gun position indirectly controls the film's properties such as uniformity, thickness, and basis weight [18,20].

In this spray system, CNF wet film can be fabricated and should be subjected to a drying process to remove the excess water. The drying process can be carried out by different methods such as drying the wet film in an oven at a temperature of 105°C and drying the wet sheets in a laminar flow chamber or fume hood with a constant airflow under standard laboratory practice. The dried CNF film can be subjected to various characterization and applications. The dried spray-coated CNF film has compact and two unique surfaces, namely rough surface and smooth surface. The rough surface was exposed to the air when spraying CNF suspension on the metal plates. The smooth side of the CNF film was on the stainless-steel and it has a shiny surface as one of the finishing qualities in this process. The surface smoothness of the CNF film was replicated from the stainless-steel plate. This smooth side of the film was used for the fabrication of numerous functional materials such as substrates for flexible electronics and printed electronics [16].

3. Analysis of spray-coated CNF film and its characterization

Spraying cellulose nanofiber suspension on polished stainless-steel plates is a rapid process for the fabrication of CNF wet film. This method produces a compact film of cellulose nanofibrils having two unique surfaces. The operation time required to form a 15.9 cm diameter CNF film consumes less than a minute and is independent of CNF suspension concentration. However, this method produces a wet CNF film and is subjected to a drying process to evaporate the water in spray-coated CNF suspension. The drying of the spray-coated wet film can be performed by keeping the wet film in an oven at 105^{o}C or air drying in a laminar flow chamber under standard laboratory conditions. The dried film on the stainless-steel plate can be easily peeled from the plate and the CNF becomes free-standing/self-standing films for various applications [20].

The spray-coated CNF films are displayed in **Figures 2** and **3**. The spraying of CNF on circular and square stainless-steel plates was performed to fabricate the circular and square sheets, respectively. The operation time of spraying the CNF suspension to form a 15.9 cm diameter film was less than a minute. Unlike vacuum filtration, the CNF concentration for film fabrication was independent of their operation time in the spraying process. The spraying of CNF on the metal surface produces an ultra-smooth film for various applications [18].

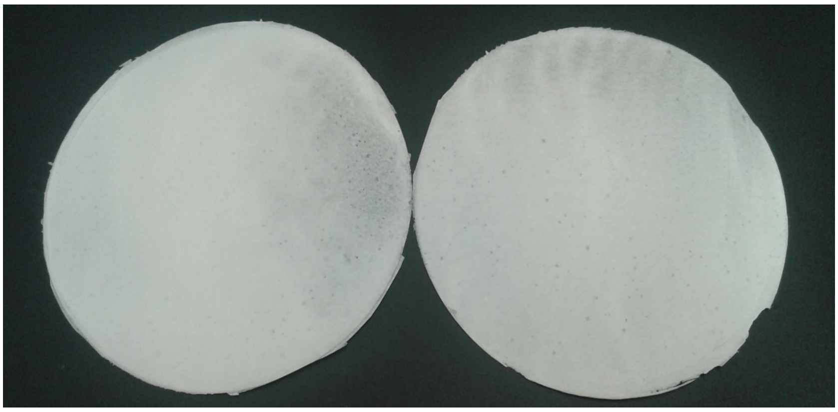

Figure 2. Circular sheets of CNF film produced via spray-coating process

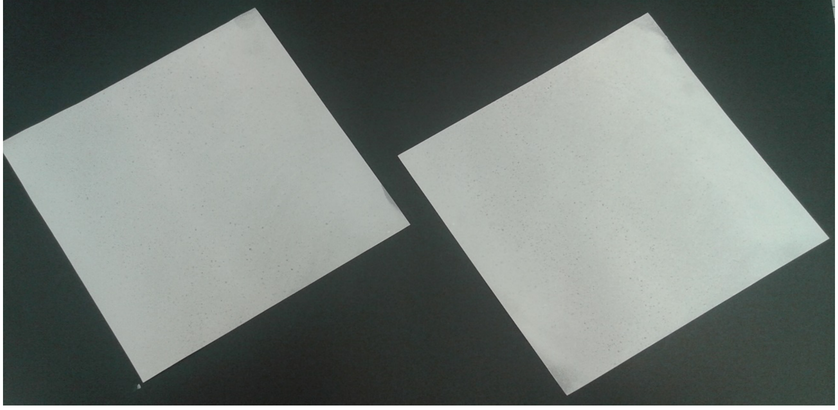

Figure 3. Square sheets of CNF film produced via spray-coating process

The cross-section SEM (scanning electrode microscope) micrograph of CNF film prepared via spray-coating is presented in **Figure 4**. The SEM micrograph confirms the complex cellulose nanofibrils layers intertwined through the hydrogen bonding between the hydroxyl groups of the CNF. This also increases the tortuosity of the film and demonstrates this effect on the barrier performance of the CNF film [16].

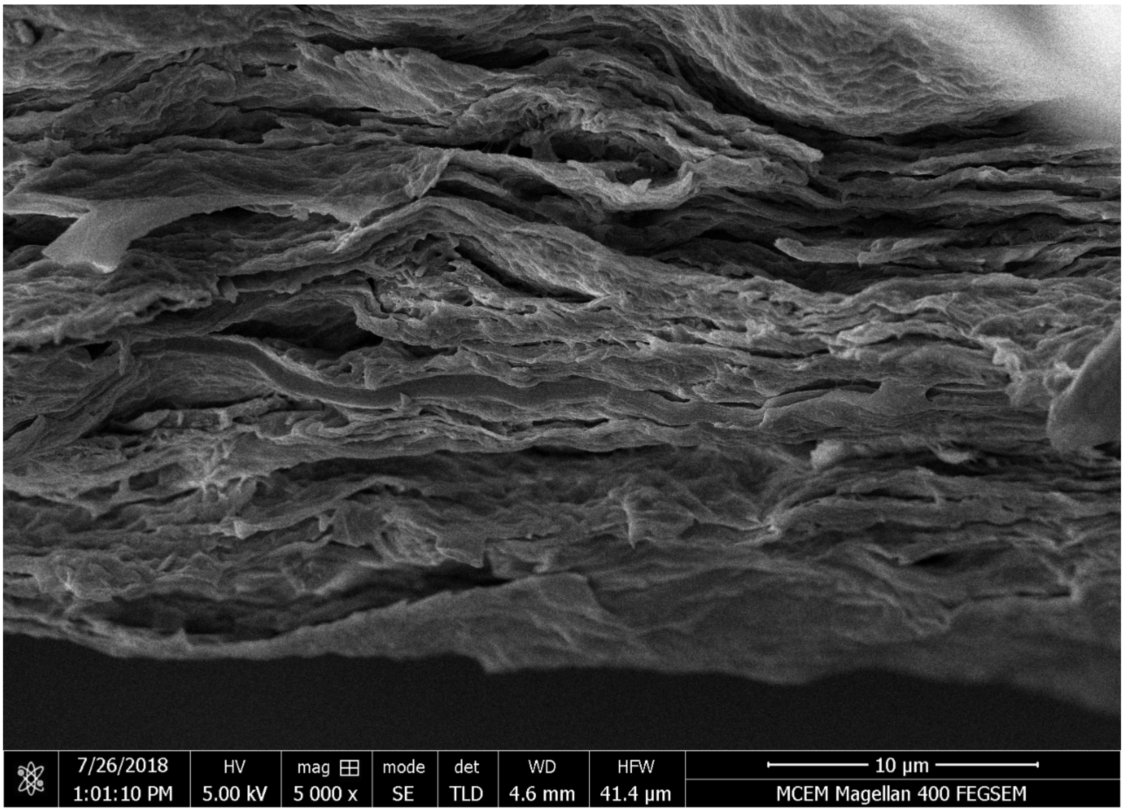

Figure 4. Cross-sectional view of spray-coated CNF films

The comparison of SEM micrographs of the spray-coated CNF film and the CNF film prepared via vacuum filtration is shown in **Figure 5**. The CNF film prepared via spraying is compact and rough on the free side and smooth on the other side. The rough surface of the film is highly porous due to various sizes of fibers distribution. The smooth side of the film is shiny and its smoothness is replicated from the surface of the stainless-steel plate [18,20]. The mechanism of the replication of the smoothness from the stainless-steel plate remains obscure [16]. The rough and smooth surfaces of the spray-coated CNF film are important in the fabrication of various functional materials [16]. For example, in the construction of flexible electronics and printed electronics, the conductive ink on the cellulose substrates should penetrate well into the surface of the substrates. To achieve this, sufficient roughness/smoothness of the substrates is required for spreading the conductive ink. Similarly, the roughness and smoothness of the film can be used in the fabrication of solar cells [16].

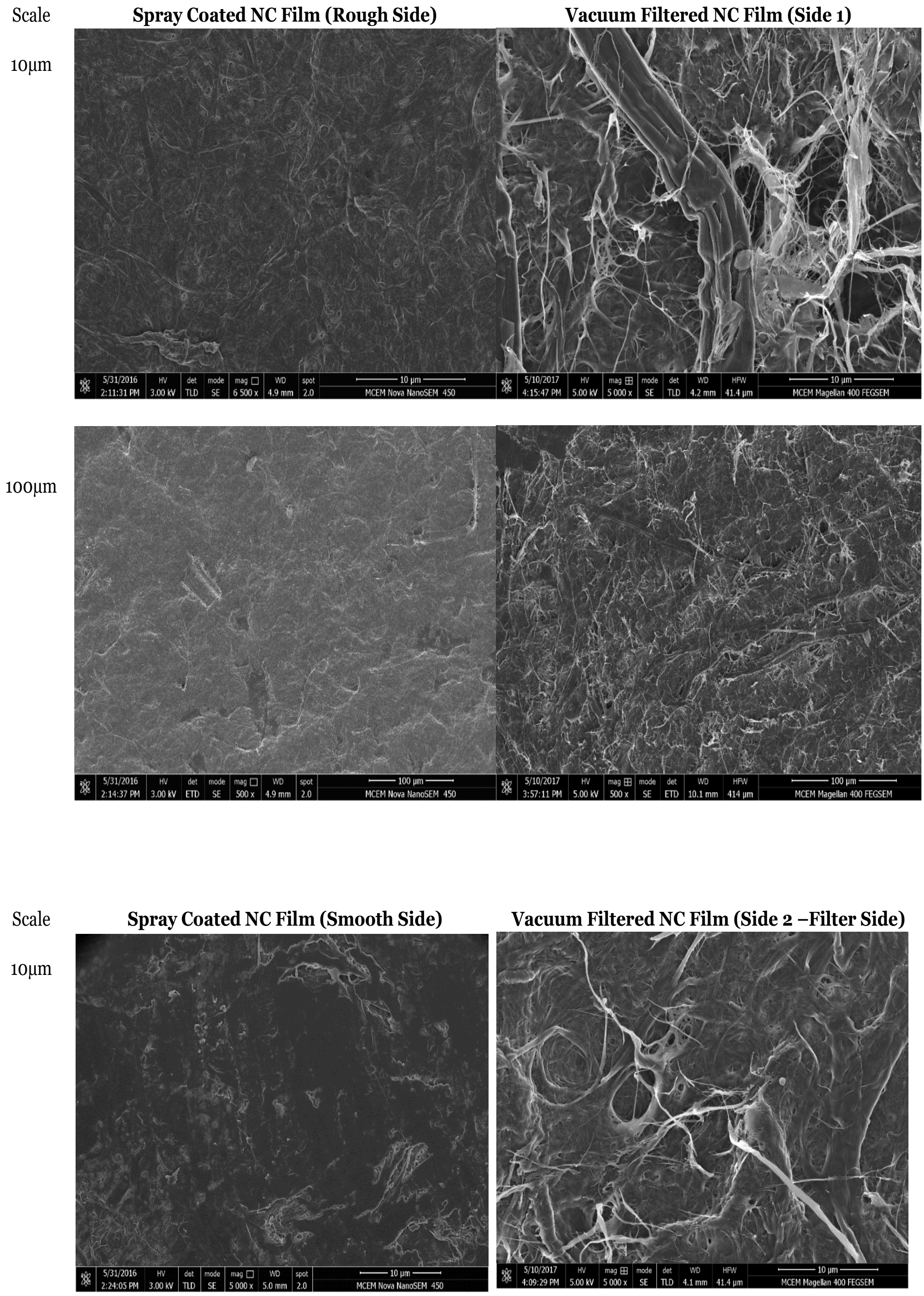

Figure 5. SEM micrographs of the spray-coated CNF film and the CNF film prepared via vacuum filtration

4. Surface roughness of free-standing CNF film

As discussed earlier, the surface roughness of the film is one of the important criteria for the construction of functional materials. The surface roughness of the CNF film was evaluated by optical profilometry.

The optical profilometry image of the rough side of the CNF film prepared via spray-coating is shown in **Figure 6**. The rough side of the film was highly porous with high surface roughness on the film. This was due to various fiber distributions in cellulose nanofibrils. The mean roughness on the rough side of the CNF film was found to be 1654 nm and the RMS (root mean square) value of surface roughness on this side was reported to be 2087 nm. The optical profilometry image of the smooth side of the CNF film is displayed in **Figure 7**. The Ra (average roughness) and Rq (root-mean-square roughness) values from the image confirm that the surface was very smooth and shiny. The Ra and Rq values were evaluated to be 278 nm and 389 nm, respectively. The optical profilometry images of the free and filter sides of the CNF film prepared via vacuum filtration are shown in **Figures 8** and **9**. Ra and Rq on the free side of the vacuum-filtered CNF film were 2150 nm and 2673 nm, respectively. Similarly, Ra and Rq on the filter side of the vacuum-filtered CNF film were 3015 nm and 3751 nm, respectively. When compared with the surface roughness of CNF film prepared from vacuum filtration, the spray-coated CNF film has a smoother and less porous surface [18,20].

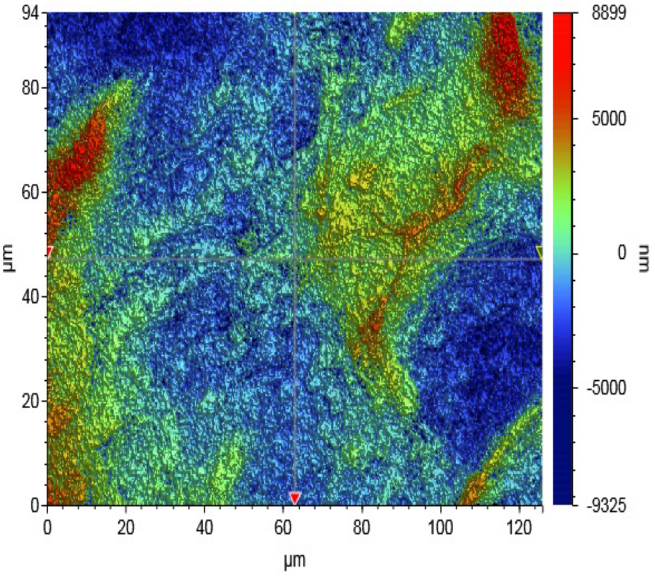

Figure 6. Optical profilometry image of the rough side of CNF film via spray-coating

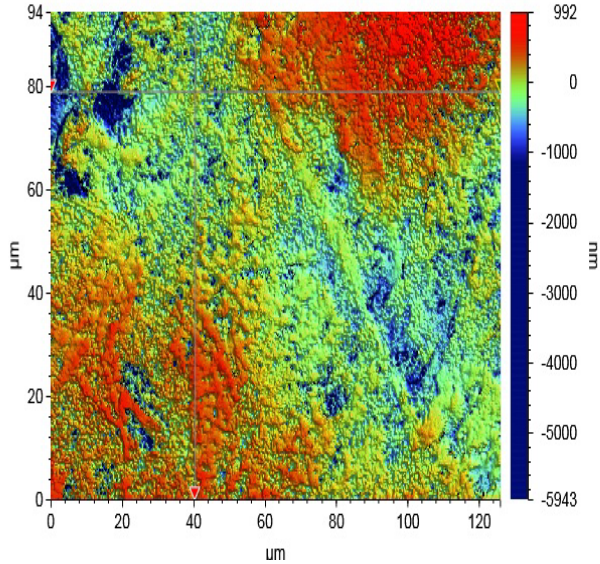

Figure 7. Optical profilometry image of the smooth side of CNF film via spray-coating

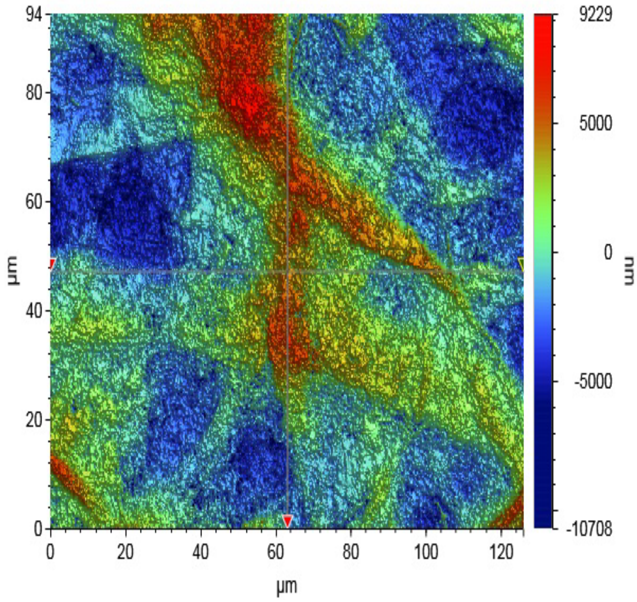

Figure 8. Optical profilometry image of the free side of CNF film via vacuum filtration

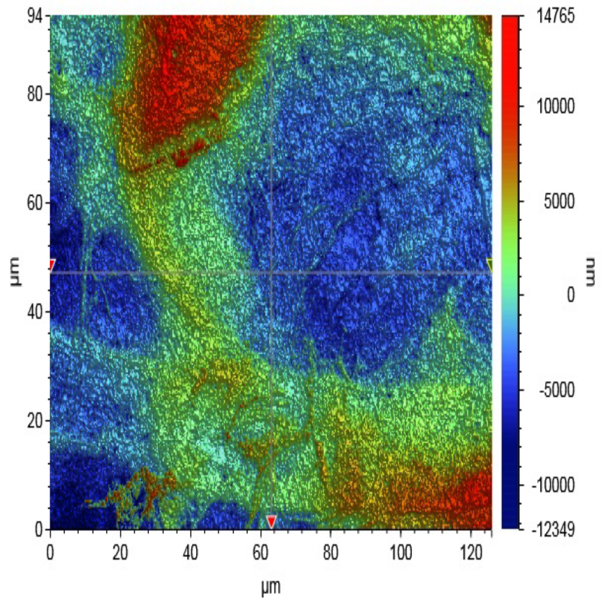

Figure 9. Optical profilometry image of the filter side of CNF film via vacuum filtration

5. Atomic force microscopy (AFM) studies

The AFM studies on the spray-coated CNF film were conducted to analyze the nanoscale surface roughness of the rough and smooth surfaces of the film. The visual examination of the spray-coated CNF film showed roughness on the free side of the film and smoothness on the metal side. In addition, the smooth side was very shiny as one of the finishing qualities of the film. Based on the AFM micrographs, the RMS surface roughness of the CNF film was evaluated to be 51.4 nm on the rough side and 16.7 nm on the smooth side in an inspection area of 2 µm x 2 µm. For the vacuum-filtered CNF film, the RMS surface roughness was found to be 102.3 nm on the free side and 70.64 nm on the filter side in the same area of inspection. In this way, the nanoscale roughness of the CNF film was evaluated and these surfaces were implemented for the construction of printed and flexible electronic substrates. **Figures 10** and **11** show the surface roughness of the CNF film and the RMS values evaluated from these AFM micrographs [16].

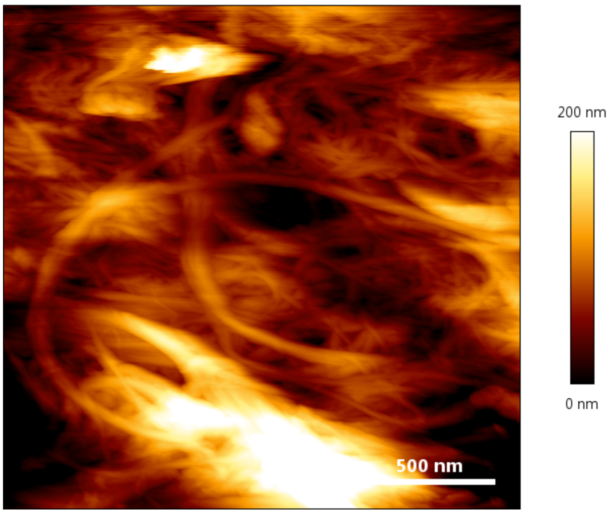

Figure 10. Rough surface of the spray-coated CNF film

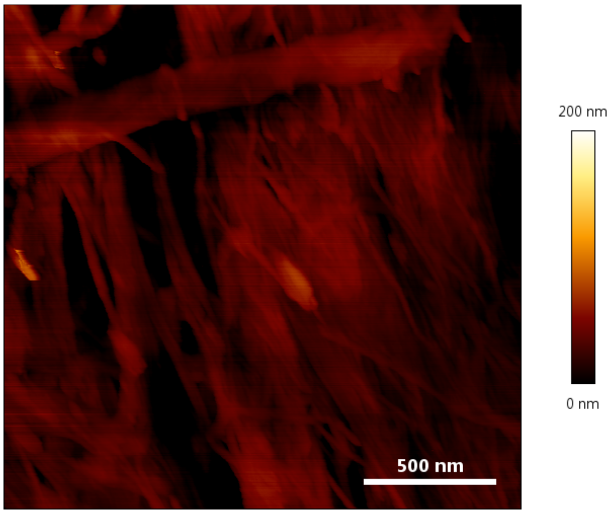

Figure 11. Smooth surface of the spray-coated CNF film

6. Thickness mapping of CNF film via spray-coating

The thickness of the film is one of the main parameters for controlling the barrier performance of the film. The thickness mappings of the spray-coated CNF film and the vacuum-filtered CNF film are presented in **Figures 12** and **13**. The thickness mapping of the CNF film was evaluated from 1.5 wt.% CNF film via spray-coating 1.5 wt.% CNF on the stainless steel plate. The basis weight of the film produced by vacuum-filtering and spray-coating, respectively, is 100.5 ± 3.4 g/m^2 and 95.2 ± 5.2 g/m^2. Vacuum-filtering consumes a substantially longer dewatering time of 15 minutes to form the film. In spray-coating, the operation time to form the film is independent of CNF suspension concentration. Even after accounting for the little variation in basis weight, the spray-coated CNF film is somewhat thicker when compared to the vacuum-filtered film. The apparent densities of the vacuum-filtered and spray-coated films were 793 and 834 kg/m^3, respectively. Additionally, the thickness of the spray-coated film is distributed across a somewhat larger range. Based on **Figures 12** and **13**, it seems that the spray-coated CNF film has better uniformity and is comparable with vacuum-filtered CNF film [20].

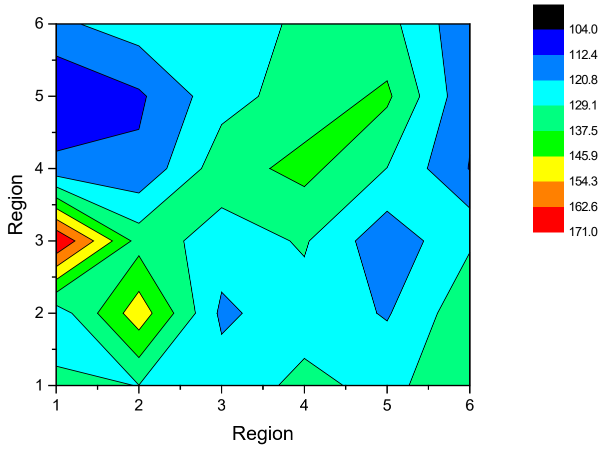

Figure 12. Thickness mapping of spray-coated CNF film

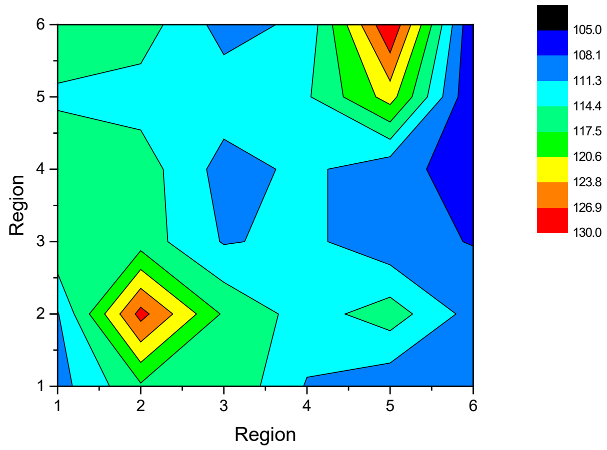

Figure 13. Thickness mapping of vacuum-filtered CNF film

7. Thickness and basis weight of the film

The linear relationship between the thickness and basis weight of the CNF film via spray-coating is shown in **Figure 14**. It demonstrates that the CCD (Central Composite Design) design model may be used to scale up the spraying process since it matches the actual experimental data well. The thickness and basis weight of the CNF film were tailored by spraying CNF suspension from 1 wt.% to 2 wt.%. The operation time for spraying CNF suspension was independent of the fiber content in the CNF suspension. The following models have been developed to scale up the process. These models reveal that the basis weight and thickness of the CNF film were found to be highly influenced by the CNF suspension concentration as opposed to conveyor speed and spray distance based on the test findings.

Basis Weight = –64.45 + 122.43 × CNF Suspension concentration –17.28 × Velocity of the conveyor +0.34 × Spray distance

Thickness = –0.106 + 0.111 × CNF Suspension concentration –0.000017 × Velocity of the conveyor –0002 × Spray distance

These are linear models confirming the direct relationship between the thickness and basis weight of the CNF film.

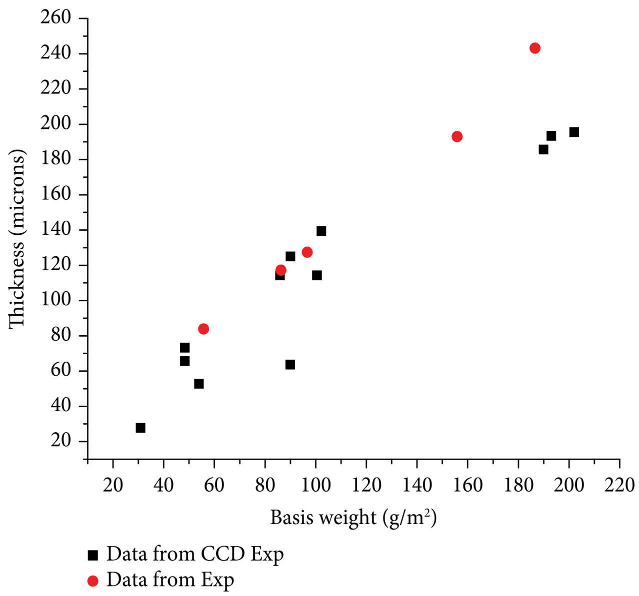

Figure 14. Linear relation between thickness and basis weight of spray-coated CNF film

From the linear models, there were two important parameters controlling the thickness and basis weight of the CNF film. The CNF suspension concentration and velocity of the conveyor in the experimental setup were the deciding parameters for tailoring the CNF film's properties [23].

8. Mechanical performance of spray-coated CNF film

The tensile index of the spray-coated film and its comparison with vacuum-filtered film is displayed in **Figure 15**. The modified configuration of the experimental spray-coating system produces CNF film that has higher tensile indices than the CNF film produced via vacuum filtration. This is because the high uniformity of CNF film was fabricated in the modified configuration of the spray-coating system. Generally, spraying CNF suspension on the polished metal plate was controlled by numerous parameters like CNF suspension consistency, and process variables in the spray system such as spray distance, nozzle diameter, type of spray system, and sprayability of CNF suspension. These parameters indirectly control the uniformity of the film, which is linked to the barrier and mechanical properties of the film [18].

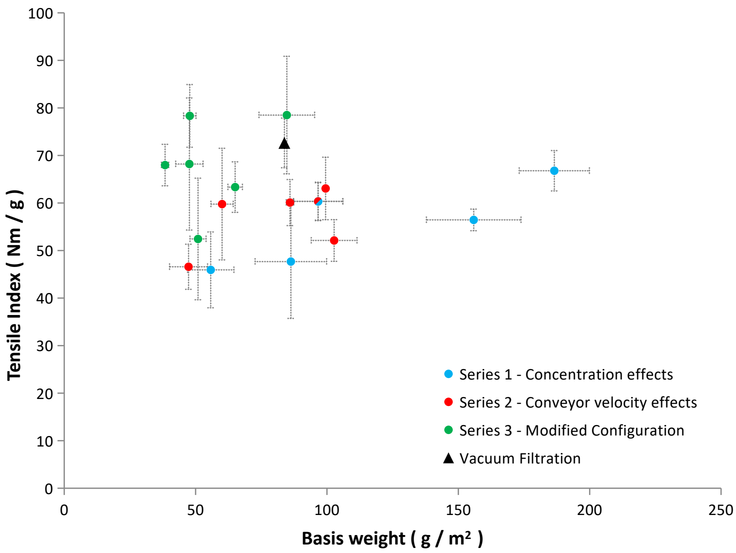

Figure 15. Tensile index of spray-coated cellulose nanofiber film and its comparison with vacuum-filtered film

9. Cost and environmental analysis

The free-standing CNF film has not been commercialized so far and it is in the development and research stage. The patents on free-standing CNF films and its composite produced via various methods have been increased. The cost of Daicel KY 100S from Daicel Chemical Pvt Ltd was 2500 AUD per 50 kg of nanocellulose. The cost of the spraying system was around 4000 AUD for the construction of an experimental system to spray CNF on the polished metal surface. The size of the film is 220 mm x 220 mm for square sheets and 159 mm for circular sheets. The basis weight of the film was assumed to be 100 g/m^2. For 1 kg of KY 100S, 10 sheets can be fabricated and each sheet consumes 5 AUD for the fabrication. The operation and maintenance costs were not considered in this study. The operation time for the fabrication of 220 mm x 220 mm square sheets and 159 mm circular sheets was less than a minute. When compared with Vacuum Filtration (VF), a laboratory version of the paper-making machine, spray-coating is a process intensified for the fabrication of free-standing cellulose nanofiber films and their composites. In spraying method, it involves a few steps such as spraying CNF suspension on the metal plates followed by drying under a standard approach. It requires less fixed capital and operating and labor costs. Conversely, vacuum filtration involves many steps such as agitation of CNF suspension, mixing of CNF suspension, dewatering, couching, sheet removal, drying, and then pressing. It confirms that filtration requires good fixed capital and high operating and labor costs. Spraying operation can be integrated with other coating methods such as roll-to-roll (R2R) to produce a high production rate of CNF film. Thus, the cost of CNF film will be reduced for commercializing in the market.

Under controlled composting circumstances, the biodegradability and compostability of nanofibrillar cellulose-based (NFC) products such as films, concentrated NFC, and paper products incorporating NFC were assessed. All the NFC products that were evaluated met the criteria for biodegradability outlined in European Standard EN 13432. NFC even increased the biodegradability of paper added with 1.5% NFC. The modified pilot-scale composting test EN 14045 was used to assess disintegration during composting. In three weeks of composting, NFC films entirely decomposed, and NFC did not affect how easily paper products containing NFC degraded. Through a bioluminescence test using *Vibrio fischeri*, ecotoxicity during the biodegradation of NFC products in a compost environment was assessed. For any of the samples, there was no evidence of acute toxicity [24].

10. Comparison with other coating processes and validation

Spraying cellulose nanofibers on polished stainless-steel plates is a rapid process to form a compact CNF film. It is a new process that should be compared with other conventional methods to confirm the efficacy of the spray-coating method [25]. The current spraying method has the capacity to handle the CNF suspension from 1.0 wt.% to 2.0 wt.% to produce the thickness and basis weight of CNF film from ~60 µm to ~200 µm and ~55 g/m^2 to ~199 g/m^2 [20]. The performance of the spray-coating process can be improved by the high-performance spray system, which can handle the CNF suspension of more than 2.00 wt.% [16,18]. Additionally, the rheology modifier such as Montmorillonite (MMT) clay can be added to the CNF suspension for spraying to avoid any interruption in forming a spray jet for the fabrication of the films [16]. When compared to spraying micro-fibrillated cellulose (MFC) on the 3D structures [21], this method is quite reproducible and produces the CNF film without any cracks and homogenous film with a high degree of uniformity [18]. The earlier method of spraying MFC on brass 3D structure produces the MFC film with the basis weight and thickness from 59 to 118 g/m^2 and 46 to 68 µm respectively. The spray-coated 3D structure consists of cracks and wrinkles formed on the surface. However, the reported method handled high solid MFC suspensions such as 4.5 wt.% and 9 wt.% MFC suspension, resulting in the formation of a disturbed spray jet and results in cracks and wrinkles on the film. High MFC suspension behaves as a gel-like fluid and loses the sprayability by the spray systems [21].

Similar to the current spray-coating method, the earlier method of spraying MFC on the nylon fabric was attempted to fabricate the free-standing CNF film. In this method, spraying MFC on the nylon fabric was time-consuming (10 to 20 minutes), and then the wet sprayed film was subjected to water removal from CNF suspension by applying vacuum, which is similar to vacuum filtration. The time consumed for filtration was around 15 to 90 seconds, followed by vacuum drying under standard temperature. The spray-coated MFC film produced from this spraying process has a basis weight varied from 13.7 g/m^2 to 124 g/m^2 with the thickness of the film varied from 10 µm to 72 µm. It was also reported that the imprints of nylon fabric were marked on the spray-coated MFC film [19]. Spraying was more efficient in the fabrication of free-standing CNF film when compared with solvent casting, vacuum filtration, and hot pressing. These processes were problematic in the evaporation or removal of solvent from CNF film and time-consuming, with limitations in the basis weight of the film. Spin coating is a laboratory approach for the fabrication of free-standing thin CNF film for the study of biomolecule interaction. It is not a scalable method due to the removal of water from the suspension via spinning to form ultra-thin films. This method can be used to coat the substrates for laboratory-scale studies. R2R coating is another approach for the fabrication of CNF films that has the capacity for large-scale production of the film. In this method, CNF was coated on the pre-treat substrates such as plastic films. The spreading of CNF on the substrates was a challenging task. After being peeled from the substrates, it was coated and dried under pressing. The basis weight of the CNF film can be achieved from 0.1 to 400 g/m^2 [15].

Given this analysis, spraying CNF on the base substrates is more advantageous in the fabrication of free-standing CNF film. Spraying on the stainless-steel plates produces films with unique surfaces (mainly smooth surface on the steel side) and it can be used for the fabrication of functional materials. When compared with other coating processes, the operation time for spray-coating to fabricate a 15.9 cm diameter film in the current practice was less than a minute and it was independent of CNF suspension concentration. The integration of R2R coating with a spray system is another approach for the large-scale production of free-standing CNF films.

11. Applications of spray-coated CNF films

Spray-coated CNF films have been utilized in various fields and applications as a substrate for developing functional materials. **Figure 16** shows various applications for CNF films in different fields. Due to the rapid process of spraying, it can be used as a barrier material to replace synthetic plastics in the packaging sector. Generally, cellulose nanofibers have outstanding oxygen barrier properties, which are better than that of synthetic plastics. However, the water vapor permeability of CNF was not equalized with the water vapor barrier performance of synthetic plastics. Spraying CNF on the metal plates produced a film with compactness, acting as a good barrier against water vapor and its performance was better than the vacuum-filtered film and comparable with synthetic plastics. Furthermore, the water vapor barrier of the CNF film was improved by incorporating nano-inorganics/ antimicrobial inorganics into the cellulose nanofibril matrix in the CNF suspension [15].

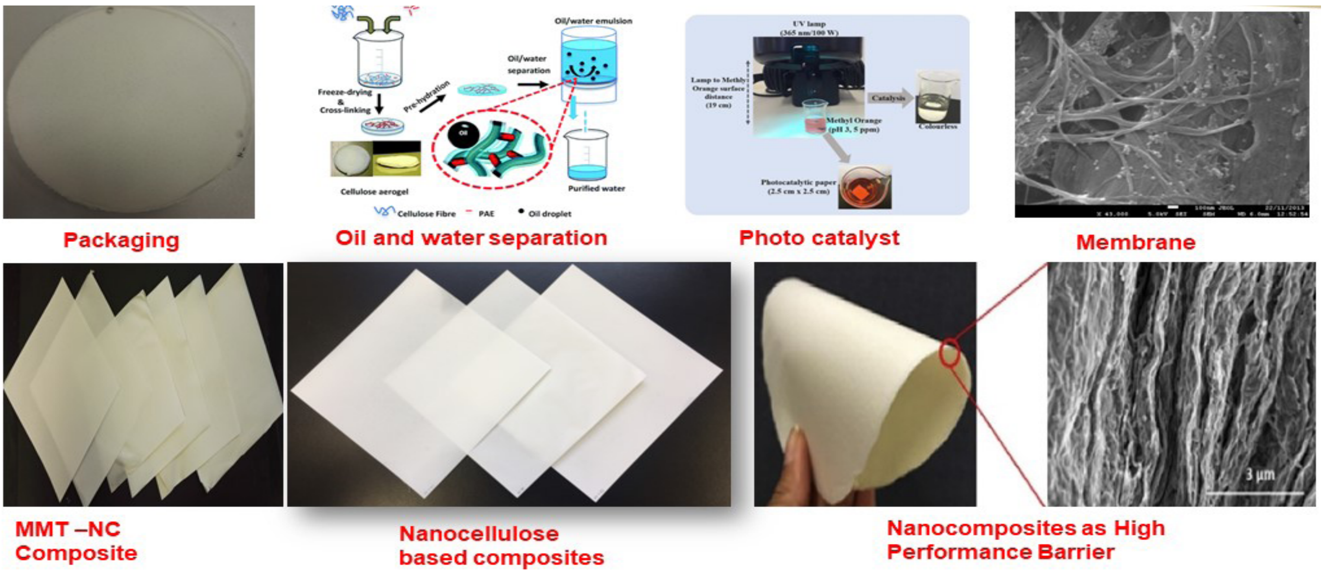

Figure 16. Applications of cellulose nanofiber films via spraying and filtration

In the case of the fabrication of cellulose nanofiber-Montmorillonite (CNF-MMT) composite, the time taken for dewatering in the vacuum filtration process was exponentially increased and consumed more than 3 to 24 hours depending on MMT content in cellulose nanofiber suspension. To mitigate this problem, spray-coating has been implemented to fabricate CNF-MMT composite and the operation time for spraying CNF-MMT suspension was independent of the MMT concentration in CNF suspension. The spray-coated CNF MMT composite can be a good barrier material for replacing synthetic plastics. The anti-microbial inorganics incorporated in cellulose nanofiber suspension were fabricated as a composite via the spray-coating process. This free-standing composite can be used as anti-microbial packaging and bioactive packaging. Similarly, the free-standing CNF film prepared via spraying can be used as the membrane for wastewater treatment. In this composite, titanium dioxide was also incorporated in the film and it becomes a photocatalyst for wastewater treatment applications [15].

The membranes were also developed from spray-coated CNF film to separate the oil and water mixture. In addition to that, various composites from CNF-inorganics can be fabricated via the spray-coating process for various applications. The spray-coated CNF films have unique surfaces of rough surface and smooth surface. The smooth surface of the CNF film can be used for the development of printed and flexible electronics. **Figure 17** shows the CNF film as a substrate for printed electronics and flexible electronics [16].

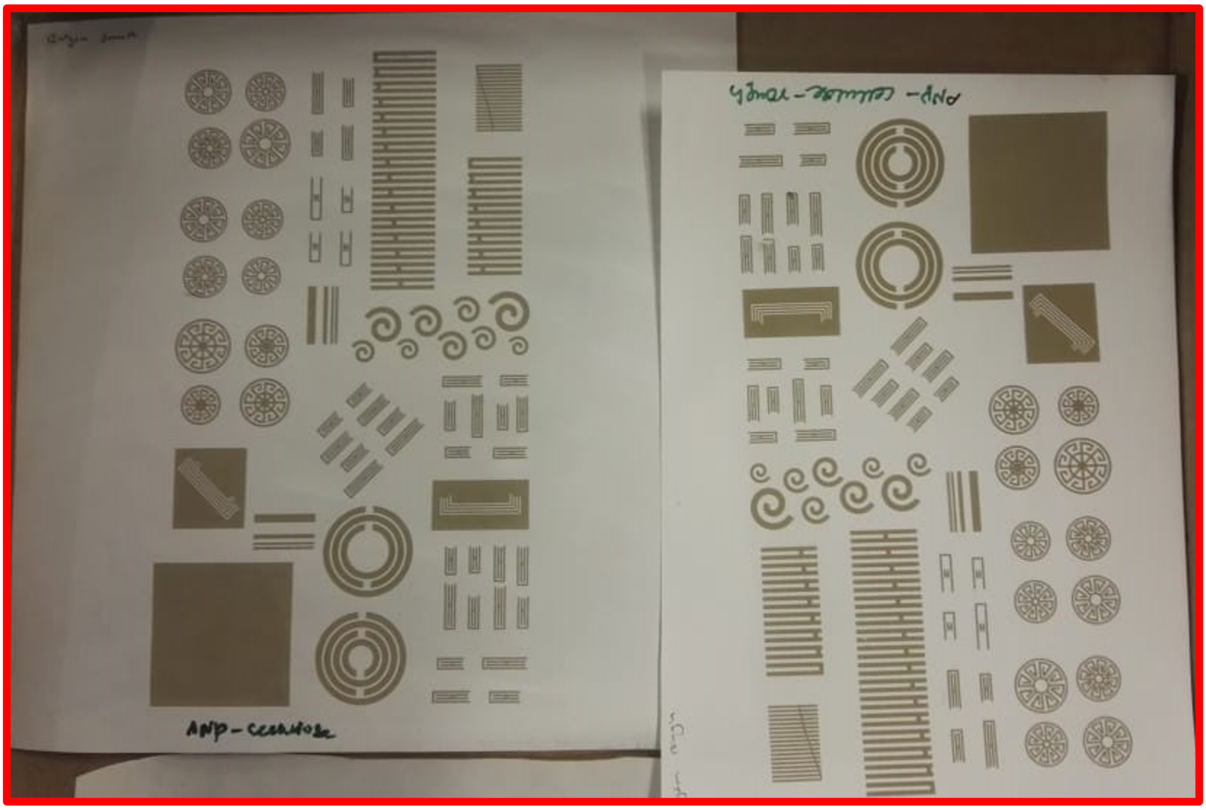

Figure 17. The printed circuits on the spray-coated nanocellulose films

The spray-coated CNF film can be used as a base biomaterial for the development of tissue engineering material and drug-delivery vehicles. The silver nanoparticle and MMT were coated on the spray-coated CNF film via a laboratory spraying method to develop a drug-delivery vehicle composite for the treatment of wounds. The silver nanoparticle present on the surface of CNF film can eradicate the pathogens at the wound site and CNF film can act as a template for skin regeneration. In addition to spray-coating to prepare free-standing CNF films and composites, this methodology can be used for developing CNF barrier layers on the paper and paper board substrates to enhance their barrier potential against air and water vapor. Furthermore, the spraying CNF suspension was implemented to coat the CNF layers for membrane development in water treatment applications [15].

12. Spray-coated CNF film in packaging

The most important application of spraying CNF on the base surface was the fabrication of CNF film, which can be used as a barrier material and a good alternative for synthetic plastics. **Figure 18** demonstrates the capability of the spray-coated CNF film as a reliable water vapor barrier and its comparison to synthetic plastics. However, the barrier efficacy of the packing film against water vapor is also determined by its thickness. Due to this, the film's water vapor permeability—a number that was determined by normalizing the thickness of the film with its WVTR (water vapor transmission rate) values—was used to characterize the performance of the water vapor barrier. **Figure 18** also compares synthetic plastics with spray-coated CNF film in terms of WVP (water vapor permeability). This graphic demonstrates how comparable the WVP of spray-coated CNF film is to synthetic polymers. Beyond this benefit, CNF is an environmentally benign nanomaterial with the ability to break down in the environment [25].

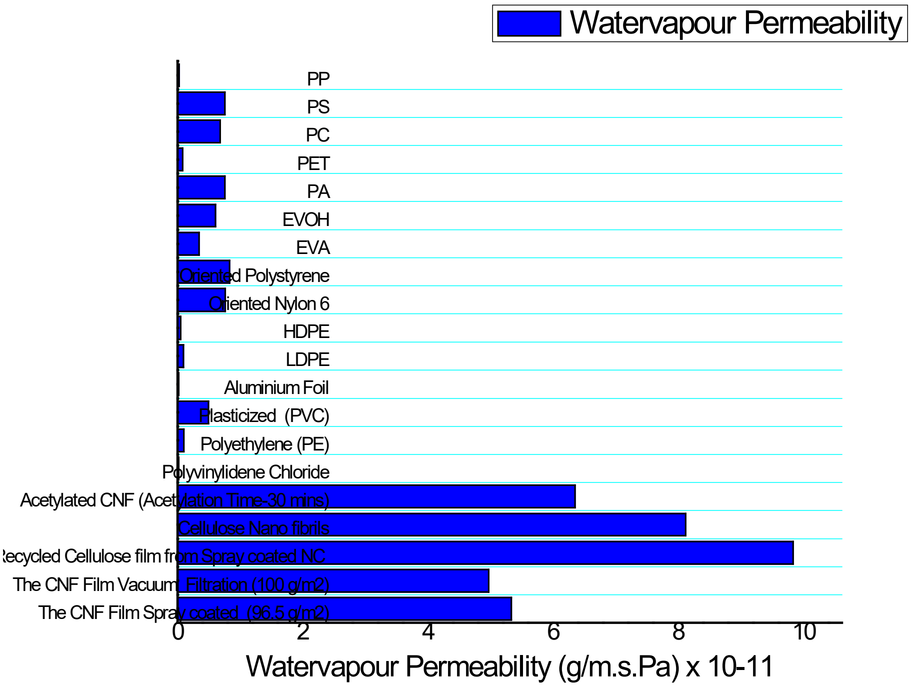

Figure 18. Water vapor permeability of CNF film and its comparison to synthetic plastics

13. Conclusion

To meet the speed of production of plastic film, a rapid process is required to fabricate the free-standing film of cellulose nanofibers/nanocellulose material. To fulfill this need, spraying/spray-coating is a rapid process to fabricate the CNF film in a free-standing manner/self-standing sheets for various applications. The current spraying process rapidly produces the free-standing film, with an operation time of less than a minute in forming the spray-coated wet film. However, the drying of spray-coated wet film consumes more than 24 hours in an air drying process under standard laboratory conditions and a couple of hours in an oven at 105^{o}C under standard practice. Unlike the vacuum filtration process, the operation time and film formation time of the spraying process were independent of CNF suspension concentration and had a potential for scaling up. The spray-coated film has unique surfaces such as rough and smooth surfaces, and these surfaces lead to the development of various functional materials such as packaging, membranes, and drug-delivery vehicles.

Disclosure statement

The author declares no conflict of interest.

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