

Barrier and Mechanical Properties of Spraycoated Nanocellulose laminates on the paper

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Abstract— An excellent barrier property is a critical requirement for the packaging material development. To meet this requirement, the paper substrates were laminated/coated with aluminium and extrusions coated with synthetic organic polymers are the conventional process to produce barrier layers. However, the conventional process has many shortcomings such as cost expensive process, environment disposals issue such as non- biodegradable of aluminium laminated and synthetic polymers. Past decade, various natural biopolymers coated with a paper substrate are an alternate process to produce its barrier layers of polymers on the substrates. Among many coating techniques, the spray coating is a novel technique and has many advantages such as the production of even coating surface in the base sheet and contactless coating with the substrate. This work investigates a laboratory scale spray coating of Cellulose nanofiber (CNF) on paper substrates to enhance their mechanical and barrier properties. Cellulose nanofibre (CNF) is a bio-based nano-material and diameter of fibre from 5-70 nm and length of several micrometres. A laboratory scale spray coating of CNF suspension used from 0.5 to 1.5 wt.% on the paper substrates is developed and the range of achieved grammage and air permeability of coat weight on the paper substrate are 2.9 ± 0.7 - 29.3 ± 6.9 g/m² and 0.78 ± 0.17 - <0.003 $\mu\text{m}^2/\text{Pa}\cdot\text{s}$ at a sprayable concentration from 0.5 to 1.5 wt.% of MFC suspension and the scanning electron microscopy studies of spray coated paper confirms that the surface pores in the paper substrates are filled with sprayed CNF and forms a continuous film on the surface of the substrate that induces a drop in the air permeability of the paper substrate and increases its tensile strength of spray coated paper from 109.7 ± 7 to 131.1 ± 13.4 N. The developed spraying of CNF suspension on the paper substrate has excellent potential to produce a continuous film as a barrier layer on the paper substrate in a roll to roll converting process or a conceptualized spray coating process.

Keywords— cellulose nanofiber, barrier coating, spray coated laminates, air permeability, oxygen transfer rate and mechanical properties.

I. INTRODUCTION

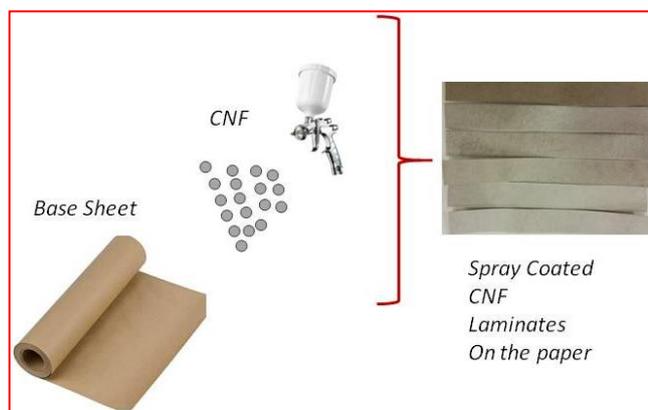
Synthetic Polymer based packaging materials are extensively used because of lowest oxygen and water permeability. However, they have poor recyclability and biodegradability and the waste is harmful to the environment. In conventional process, aluminium foil is used as layer for enhancing barrier properties of the paper board and paper substrates. Neither the plastic materials nor the aluminium is renewable and these composites are difficult to recycle. To resolve this problem, major efforts are being taken the way to identify alternative materials from natural source to improve the barrier performance as well as minimise disposal or recycle problems [1].

Nanocellulose, also known as cellulose nano-fibres or nano-fibrillated cellulose, is made by breaking down cellulose fibres into fibres with diameters ranging from 5-100nm and is as building block for development of new cellulose based functional materials. Recently, cellulose fibrils are used in the development of high strength barrier layers and nanocomposites [4]. Cellulose nanofibre has a potential of renewable, recyclable, compostable and biodegradable alternatives to the synthetic polymer based products [2]. CNF is used as coating materials for enhancing the barrier properties of a base sheet and fabrication of nanocellulose sheet and films [3].

Cellulose is the most important bio-renewable, biodegradable and biopolymer available in nature and is an excellent feedstock for the development of various sustainable functional materials such as coatings, films and

membranes on an industrial scale for production [9]. These bio-based products could provide an outstanding solution to various international problems such as recycling, disposal and incineration of waste. It is produced by disintegration and delaminating of cellulose fibrils from pulp produced from a variety of green sources such as wood, potato tuber, hemp and flax. It has dimension diameter ranging from 5 to 100 nm and is typically several micrometres in length [10]. Moreover, having a smaller dimension and a larger surface, cellulose nanofiber is a great opportunity to be developed more functional materials for various applications [11, 12]. These Cellulose fibrils at micro/nano scale are used to functionalize base sheets by coating or to make freestanding sheets/films and nanocomposites [13].

Graphical Abstract



The nomenclature for cellulose nanofiber has not been reported in a consistent manner in the previous scientific investigations. It is also called as micro fibrillated cellulose (MFC), nano-fibrils, micro-fibrils and nano-fibrillated cellulose (NFC). Nano-cellulose fibres are isolated and processed from wood via various chemical, enzymatic, and/or mechanical treatments. Due to nano size of fibres, it possesses various outstanding properties, such as high aspect ratio, high specific strength, flexibility, large specific surface area, and thermal stability, combined with biodegradability and biocompatibility. These properties could make nanocellulose suitable for a wide range of applications, such as nanocellulose film [14], reinforcing phase in composite materials [15], barriers in packaging [16], rheology modifiers for suspensions [17], filters for virus removal and water treatment technologies [18,19], flexible platforms for biomedical applications [20] and printed electronic applications [21].

It has been proved that the films/sheets and its nano composites made from nanocellulose and coated with fibre substrates increased the barrier and mechanical properties [22]. Due to outstanding multifunctional barrier properties such as oxygen and water transfer rate, it has the potential for application of packaging materials for foods [23].

Barrier materials required low gas and water permeability to protect the contents from the external influences and to preserve the flavour and nature of the packaged product.

The barrier properties of paper-based packaging can be tailored by applying layer of either synthetic or natural polymer using coating process. The previous studies confirmed that cellulose based coating on the paper based substrates substantially improved their barrier and surface properties [24, 25]. The cellulose nanofiber could be applied either on the paper or paperboard by several techniques such as solvent casting, dispersion coating, foam coating, bar and blade coating, and vacuum filtration. Figure 1 show different coating processes for paper applications and their details are updated in Table 1 in the supplementary information.

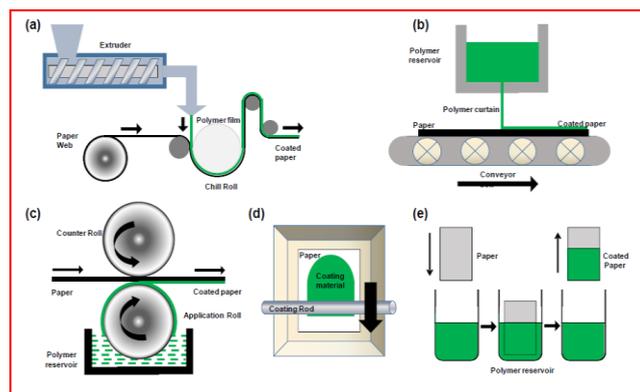


Figure 1: Different coating processes for paper applications, (a) extrusion coating; (b) curtain coating; (c) size press coating; (d) bar coating; and (e) dip coating. (Vibhore Kumar Rastogi et al, Coatings, 2015, 5, 887-930)

Extrusion coating method is a common technique for coating synthetic polymers, such as poly-ethylene. It provides a continuous processing, uniform coating, minimal pinholes and cracks in the surface of base sheet, and solvent-free application. However, it has shortcomings in the coating performance such as being unable to produce a high coat weight to achieve the necessary barrier properties, instability of the polymer during melting stage and coating speed and efficiency. It is only suitable for coating of thermoplastic polymers. Given that cellulose nanofiber is not thermoplastic it can only be suitable as a coating formulation by either dissolving cellulose nanofiber in a suitable solvent (i.e., solvent coating/casting), or dispersing the polymer in solvent (i.e., dispersion coating) [26, 27].

In dispersion and solvent coating methods, low coat weights around 10g/m² can be used to achieve the barrier layers of the base sheet, but sometimes two layers are mandatory to coat to eliminate the surface pinholes and achieve a sufficient water vapour performance. In this case, post coating process is expensive process including evaporation and drying of the coated surface [1]. Aulin et. al. reported that the preparation of carboxymethylated micro-fibrillated cellulose (MFC) films by dispersion-casting from aqueous dispersions and by surface coating on base papers and confirming that the oxygen permeability of the sheet

prepared via dispersion coating and air permeability were reduced. [22]

Curtain coating is where a uniform coating is applied and found sufficient to cover the entire surface with better gas and water vapour barrier properties. In size pressing, the coating is not covering the paper surface completely and does not provide expected barrier properties. Bar Coating and rod coating techniques provides a better control over the thickness of the coating layer, but can only be used at the laboratory or pilot scale [28]. Dip Coating is the quick testing performance of coating at laboratory scale. However, the coating thickness of the base sheet is difficult to control and hence always find practical application at the laboratory scale [1]. Due to its film forming capacity, nanocellulose could be used as coating layer on the base sheet which enhances strength and barrier functions of the base sheet. It proved that nanocellulose would be a potential coating material [1].

Size press is not able to significantly alter papers properties as the MFC coat weight barely reached 4 g/m² resulting from ten successive MFC layers on the base sheet. The bar coating of MFC on the paper board was found not to substantially enhance its barrier properties, however did increase stiffness while reducing compressing strength. [30, 31]

The nanocellulose with multilayered resin coated on the paper board is proved to decrease the water vapour permeability of paper board. The coating is performed by the dispersion coating process or lithographic printing [35].

Micro-fibrillated cellulose (MFC) and shellac were coated on the paper and paper board using a bar coater or a spray coating technique to enhance its barrier properties of the substrates. The coating performance is evaluated by the decreasing barrier properties of the sheet through decrease of the air permeance of the paperboard and papers with a multilayer coating of MFC and shellac. Furthermore, the oxygen transmission rate decreased several logarithmic units and the water vapour transmission rate reached values considered as high barrier in food packaging (6.5 g/m²/day) [36].

Although above mentioned conventional processes offers some advantages, they poses serious limitations such these methods should as they are done in batches and/or are not capable of producing high coat weight on the base sheet to achieve barrier properties of the sheet. Therefore, spraying is a potentially promising approach for the preparation of nanocellulose sheet and coating of nano cellulose on the base sheet [29, 38].

Spraying of cellulose nanofiber is an alternative technique for making nano-fibre sheets that has been used to produce either continuous self-standing films by spraying on to a fabric or to produce composite laminates by spraying onto a

base sheet [6, 7]. Spraying has some significant advantages such as contour coating and contactless coating with the base substrate. The topography of the surface of the base substrate does not influence on the coating process. It is a novel technique for creating barrier film on the base surface rapid manner. Beneventi et. al. reported that the laboratory scale spray coating of micro fibrillated cellulose on different kinds of paper substrate enhances the barrier and mechanical properties of the spray coated sheet [6]. However, after spraying, they used vacuum filtration which is like the conventional paper making process to remove the excess water. Therefore, it leads to time consuming process.

The spray coating of cellulose nanofibre on the base surface could produce sheet with enhanced barrier and mechanical properties. Based on the laboratory performance of spray coating, the nanocellulose sheet could be prepared rapidly and coating nanocellulose on the base sheet quickly to create barrier layers. Therefore, it has potential for scale up in a continuous mode. The aim of this research is to develop spraying as a rapid and flexible method to coat cellulose nanofibre on the base sheet. This paper deals production of CNF barrier layers via the spraying process and their characterization of barrier layers via evaluating its mechanical and barrier properties.

II. MATERIALS AND METHODS

The nomenclature for cellulose nanofiber has not been reported consistently in the literature. As well as cellulose nanofiber (CNF), it is also called micro-fibrillated cellulose (MFC), cellulose nano-fibrils, cellulose micro-fibrils, nano-fibrillated cellulose (NFC) and Nanocellulose (NC). In this paper, we use CNF as the generic term for the cellulose nanomaterials used. The CNF used was supplied from DAICEL Chemical Industries Limited (Celish KY-100S) at 25% solids content. DAICEL CNF (Celish KY-100S) has cellulose fibrils with an average diameter of ~ 73 nm with a wide distribution of fibre diameter, a mean length of fibre around 8µm and an average aspect ratio of 142 ± 28. DAICEL KY-100S is prepared by micro fibrillation of cellulose with high-pressure water. The crystallinity index of DAICEL cellulose nanofiber was measured to be 78%. CNF suspensions were prepared using by diluting the original concentration of 25 wt. % to 0.25 wt.% to 1.5 wt. % with de-ionized water and disintegrating for 15,000 revolutions at 3000 rpm in a disintegrator.

Evaluation of Viscosity for CNF suspension:

The viscosity of the cellulose fiber suspension is evaluated to find the sprayable concentration of CNF suspension. CNF sample was used at consistencies ranging from 0.25 to 2.0 wt. %, prepared by diluting the original concentration of 25 wt. % with distilled water and mixing for 15,000 revolutions in a disintegrator. The viscosity of the CNF suspension was evaluated by the flow cup method which evaluate the process of coating fluid flow through an orifice to be used

as a relative measurement of kinematic viscosity expressed in seconds of flow time in DIN-Sec.

Spraying cellulose nanofiber suspension on the paper substrates:

Cellulose nanofiber supplied from DAICEL Chemical Industries Limited (Celish KY-100S evaluation) was used for spraying operation for coating purpose. The domestic spray gun is used for spraying nanocellulose on the base sheet. The spray pattern is elliptical and the distance between spray nozzle and paper substrate is 20 ± 2 cm. The coating of nanocellulose on the paper substrate is one layer. The spray coated sheet is dried in the air drying under standard laboratory conditions. The experimental set is shown in Figure 02.

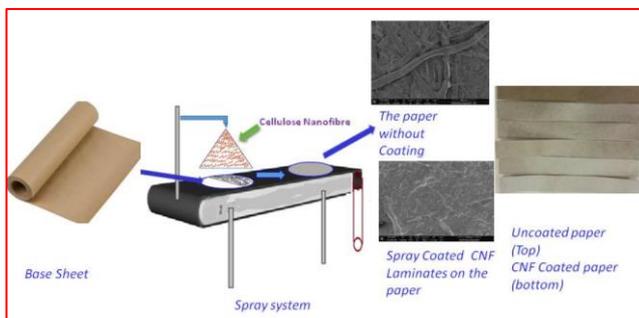


Figure 02 – Spray Coating Experimental Set up for producing CNF barrier layers on the paper.

Drying of spray coated CNF laminates on the paper substrates:

The spray coated CNF on the paper substrates were dried in the open air with specific care in the standard laboratory conditions. The dried CNF layers on the paper substrates used for various characterizations such as surface topography, basis weight, thickness, barrier properties and mechanical properties.

SEM Investigation of Spray coated CNF on the paper substrates:

Sample Preparation:

The spray coated paper (4mm X 4mm) is fixed on the stab using carbon tape and blown with Nitrogen to remove the any dust or any loose material on the sample and then coated with Iridium with a maximum thickness of $10 \mu\text{m}$. Moreover, the iridium coated samples are blown off with Nitrogen to remove any dust and loose materials on the sample before loading into the FEI-NOVA Nano SEM 450 (Jisheng Ma, 2015).

Parameters for Scanning Electron Microscopy:

Nanocellulose is a biodegradable and delicate material in nature and highly susceptible to high accelerating voltage. Therefore, the parameters for collecting micrograph are optimized. The surface morphology and topography of the spray coated paper was characterized using FEI-NOVA Nano SEM 450.

Mode 1: This mode is used for collecting the low resolution micrograph at $100 \mu\text{m}$ and this micrograph is ideal for investigating the survey of the surface of the nanocellulose coated sheet and the roughness of the coated surface. The optimized parameters for high voltage and spot size are 3 KV and 2.00 respectively. The working distance and aperture size are 5 mm and 6 (30 mm).

Mode 2: This mode is used for collecting the micrograph at $1 \mu\text{m}$ and $10 \mu\text{m}$ (high resolution (UHR) imaging) and this micrograph is ideal for investigating the fibre orientation and size of the fibres and pores in the surface of the spray coated surface. The optimized parameters for high voltage and spot size are 3 KV and 2.00 respectively. The working distance and aperture size are 5 mm and 6 (30 mm).

Basis weight of the CNF Coating on the paper substrates:

The basis weight (g/m^2) of spray coated CNF laminates on the paper substrates was calculated by dividing the weight of the sheet, after 4 hours drying in the oven at a temperature of $105 \text{ }^\circ\text{C}$, by the paper area.

Thickness of the CNF coating on the paper substrates:

The thickness of the spray coated CNF laminates on the paper substrates was determined using a Thickness Tester Type 21 from Lorentzen & Wettre AB, Stockholm, Sweden. The thickness was evaluated at fifteen points and averaged. The thickness was measured according to TAPPI T 411, 2015.

Barriers Properties of Spray coated CNF laminates on the paper substrates:

Air Permeability

The air permeance of dried NC films was measured with an L&W air permeance tester with an operating range from 0.003 to $100 \mu\text{m}/\text{Pa}\cdot\text{s}$. The mean value of air permeance evaluated from 3 different areas of each NC film was reported. The Technical Association of the Pulp and Paper Industry (TAPPI) standard T 460 is used to measure the air permeance of the films.

Oxygen Transfer Rate

The air permeability data was used to calculate the oxygen transfer rate of the CNF coated samples. It was evaluated by the air permeability data divided by 4 and gives the OTR. The sample calculated has been added in the supplementary information.

Surface properties

The surface roughness of the spray coated CNF laminates on the paper substrates was evaluated by Parker surface instrument.

Mechanical Properties

The strength of spray coated CNF laminates on the paper substrates were evaluated by an Instron model 5566 using test specimens of 100 mm length and 15 mm width, conditioned for 24 hours at 23°C and 50% RH before dry tensile testing based on the Australian/New Zealand Standard AS/NZS 1301.448S-2007. All thickness and tensile tests were done at 23°C and 50 % RH. The samples were tested at a constant rate of elongation of 10mm/min. The Tensile Index (TI of the samples was calculated from the tensile strength (expressed in Nm-1) divided by basis weight (gm-2). The mean value was obtained from six to seven valid tests and the error bars in the plots indicate standard deviation.

III. RESULTS AND DISCUSSION

Paper is widely used in packaging applications and is biodegradable and therefore perfectly safe for the environment. The hydrophilic nature of cellulose limits the water vapour-barrier properties and oxygen barrier properties of paper. To mitigate these limitations, paper is often associated with other materials, such as plastics, wax and aluminium, for their good barrier properties. However, these materials suffer from serious environmental issues, as difficult and inefficient to recycle. Recently, nanocellulose based materials has been considered as an alternative to produce eco-friendly barrier materials. Existing techniques to prepare cellulose nanofibre films/sheets/composites are commercially not feasible and expensive. Therefore, other cost effective and readily implementable methodologies are required to achieve cellulose nanofiber barrier layers. In the present work a novel approach is developed using spray coating technique to produce materials with excellent barrier properties.

Among many coating techniques, the spray coating has many advantages such as the production of even coating surface on the base sheet and contactless coating with the substrate. A laboratory scale spray coating of cellulose nanofiber suspension on a paper substrate was developed. When the nanocellulose suspension concentration was varied from 0.5 to 1.5 wt. %, coat weight is increased from

2.9 ± 0.7 to 29.3 ± 6.9 g/m². As a result, the air permeability of composite was decreased 0.78 ± 0.17 to < 0.0030 $\mu\text{m/Pa.s}$. Scanning electron microscopy studies of spray coated paper confirms that the surface pores in the paper substrates are filled with sprayed nanocellulose and forms a continuous film on the surface of the substrate. These are the probable reasons for the reduction of air permeability of composites. Brown Sheet as a Substrate used for coating CNF as barrier laminates for improving the barrier performance of the packaging paper.

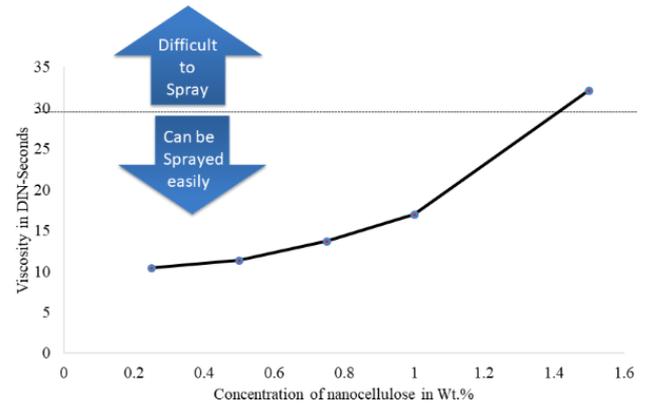


Figure 03 Viscosity of CNF suspensions

Figure 03 confirms the limitation of CNF suspension to be sprayed on the paper substrates. Effect of suspension concentration on the viscosity of cellulose nanofibre suspension is given in figure 3. It increased with solid/ fibre content in the suspension. The efflux time < 30 sec confirms the sprayable concentration of CNF for coating operation. The viscosity of 1.5 wt. % CNF suspension is 32.18 ± 0.94 DIN Sec predicted by dip cup method. It is quite challenging to predict the efflux time and viscosity of CNF suspension beyond the concentration of 1.5.Wt. %. It is reported that CNF suspension could form a gel like structure and behave shear thinning rheology even at low concentration of CNF in the suspension. The viscosity of CNF suspension increase with increasing concentration of fibres content in the suspension [44]. The rheological properties of CNF suspension is influenced by fibre morphology, orientation and aggregation. It is also reported that the viscosity of suspension increases with fibre aspect ratio and becomes substantially higher for high suspension concentration [45] When the MFC suspension concentration is higher than 2.00 wt. %, MFC suspension has lost its fluidity, becomes like stiff gel and also viscosity is higher than 32.18 DIN Sec after dispersion in water after disintegration of fibres. Onwards of this concentration above 2 wt. %, MFC suspension behaves as a viscoelastic fluid and formation of network of entangled cellulose fibrils which causes gel-like behaviour [46][47]. The spraying such high solid content suspension is really challenging because more chance of clogging the nozzle. Furthermore, the high shear force is required to pump and spray the high fibre content of the slurry.

Effect of Suspension Consistency on basis weight:

The figure 4 shows the effect of suspension concentration on the coat weight. Using lab scale spray coating, the maximum of 25-35 g/m² on the base sheet is spray coated with concentration of 1.5 wt. % of Micro fibrillated cellulose. At this concentration of spray coating of CNF on the base sheet, it forms film over the surface and this film acts as barrier materials.

Effect of basis weight of the coating on the base sheet on Air Permeability

The figure 05 shows the effect of coating weight on the base sheet on air permeance. The basis weight of the coating on the base sheet increased with suspension concentration of CNF and after 1 wt. % CNF concentration, the barrier properties of the coated sheet is enhanced. Additionally, the air permeance of the spray coated sheet drastically reduced from 3.5 to < 0.003 μm/Pa. sec.

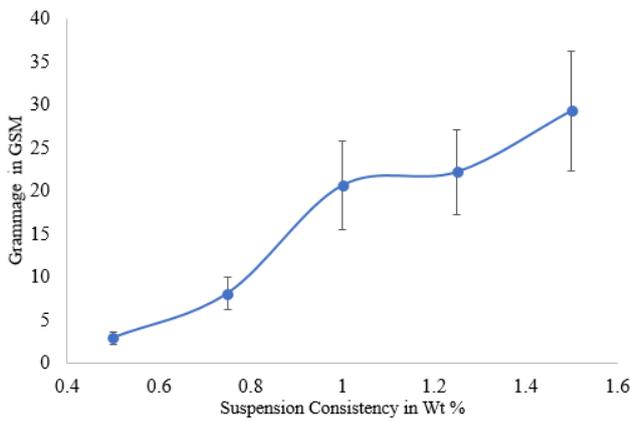


Figure 04 Plot of coat weight on the base sheet against suspension consistency

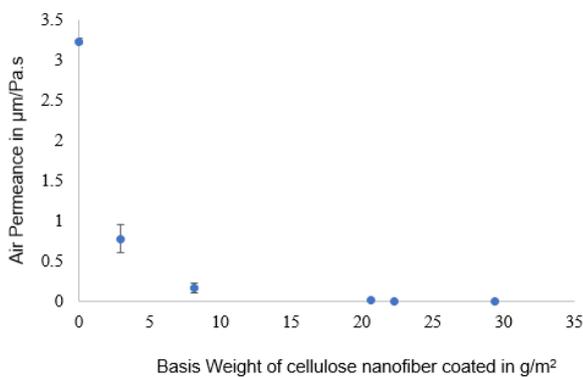


Figure 05 – Basis weight of CNF coat vs air permeance of the substrates

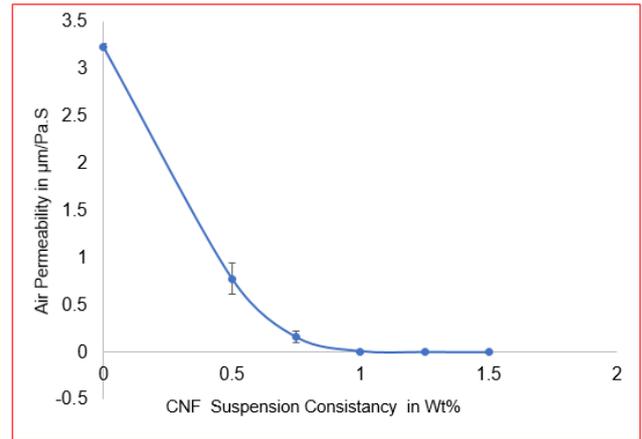


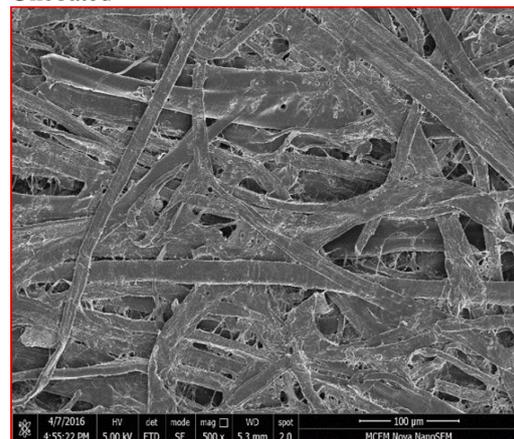
Figure 06 – Air permeability vs suspension consistency of CNF

Figure 06 – Air permeability vs suspension consistency of CNF

Figure 06 confirmed the effect of CNF suspension consistency on the air permeability of CNF coated paper. The plot proved that 1 wt. % CNF coating on the paper gives completely impermeable against air. This suspension consistency of CNF comes under sprayable conditions for coating on the paper. At this 1 wt. % Coating of CNF, CNF not only fills the surface pores of the papers and forms a barrier layer or laminates on the paper. This film can be acting as barrier to air and other gaseous substances. Below 1 wt. CNF coating, CNF can fill the surface pores of the paper only and beyond 1 wt.% CNF coating, Film formed on the paper acts as barrier layers. The limitations in spray coating of 1.25 wt. % and 1.5wt.% CNF on the paper was separation of barrier layers from the paper after drying

IV. SEM STUDIES ON COATED SURFACE

Uncoated



CNF Coated

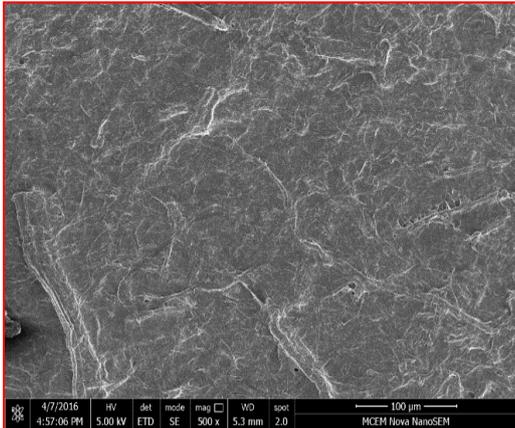
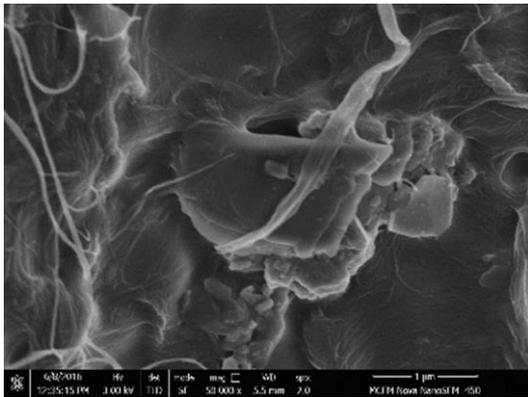


Figure 07 SEM micrographs of uncoated and 1.25 Wt% CNF coated Paper

High Magnification



Low magnification

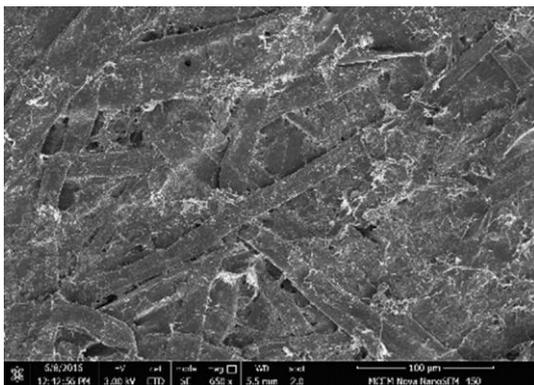


Figure 08 High and Low magnifications of 1.25 Wt.% CNF coated Paper

The Figure 07 shows the micrograph of the spray-coated paper with 1.25 Wt. percentage of the micro-fibrillated cellulose at low magnification. The micrograph (100µm) shows the deposited cellulose fibres clumps and fibres on

the surface of the base sheet. It also confirmed the different size of the fibre entangled with cellulose fibres clumps on the surface. Moreover, the micrograph (100µm) confirms the complete coverage of micro-fibrillated cellulose coating formulation on the base sheet. When compared to the micrograph (100 µm) of the uncoated paper, the coated paper showed that the coating formulation filled many surface pores and void space between the cellulose fibres. The 1µm micrograph confirms the filling surface pores by the cellulose nanofiber and forming the barrier film on the paper substrates.

V. OXYGEN TRANSFER RATE

Figure 09 shows the effect of CNF coating on the oxygen transfer of paper substrates through the evaluation of air permeability. The oxygen permeability of the paper substrate has been reduced drastically with coating of CNF on the paper substrates. As discussed earlier, CNF fills the surface pores of the paper substrates and reducing the air passage or oxygen permeance via blocking the surface pores with CNF. The figure 09 shows the high oxygen transfer rate of water wetted paper via spraying. During the wetting of the paper, the fibres are loosened and resulting the widening of the surface pores in the paper substrate, resulting the high air passage across the paper.

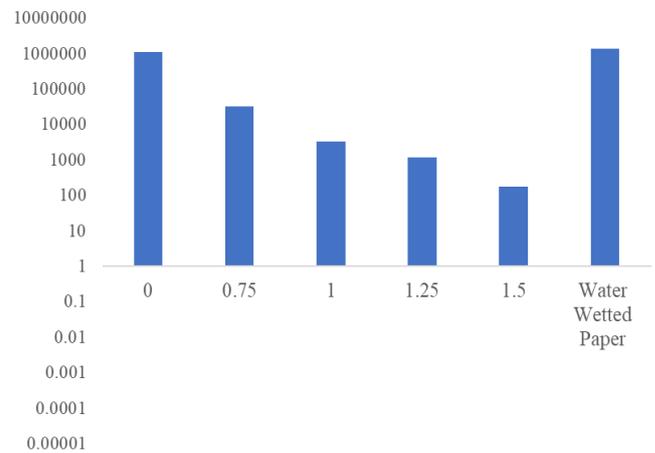


Figure 09 – Oxygen Transfer Rate of CNF coated packaging paper

Mechanical Properties

Figure 10 shows tensile strength of CNF coated paper and the effect of CNF coating has been investigated. The coating of CNF on the paper normally increased the strength of CNF laminates on the paper. Figure 10 concludes that an increase of strength of CNF laminated paper was seen, however, there was drop in strength when increased CNF concentration for coating. There was CNF barrier layer formed on the paper at higher CNF coating and these layers are not adhesive on the paper and separated from the paper. When specimen keep in Universal Testing Machine

(Instron), The CNF layer has been broken down and the paper was broken under pulling in the measurement of tensile strength in Instron Instrument. This phenomenon has been observed in the stress – strain curve of the CNF laminates added S2 in Supplementary information. The figure of this case S05 -Sample Breakage during Mechanical Testing has been shown in Supplementary information. Up to coating of 0.75 wt. % of CNF on the paper, the mechanical strength of CNF laminated paper was improved. It has been shown in Figure 11.

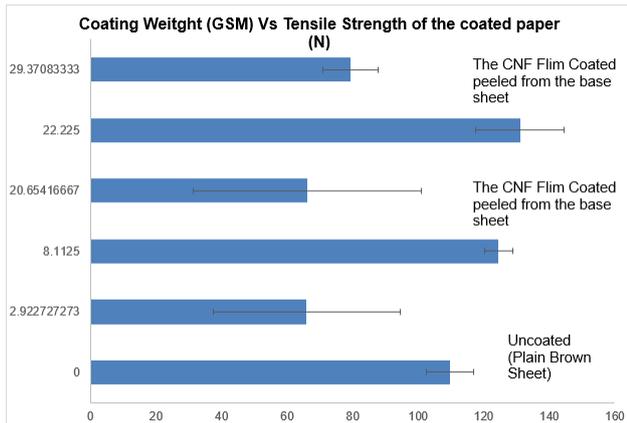


Figure 10- Tensile strength of CNF coated paper

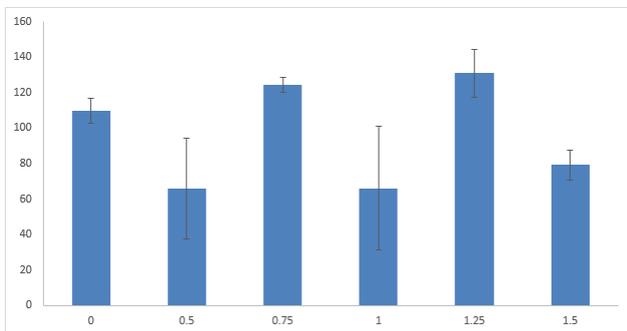


Figure 11 -Suspension Consistency Vs Tensile Strength of the coated paper

Surface Properties

Figure 12 shows the effect of suspension consistency on the surface roughness of the paper. At lower concentration of CNF, the surface roughness of the CNF coated paper does not change with the uncoated paper. At higher concentration, the surface roughness of the coated paper increased with coating percentage.

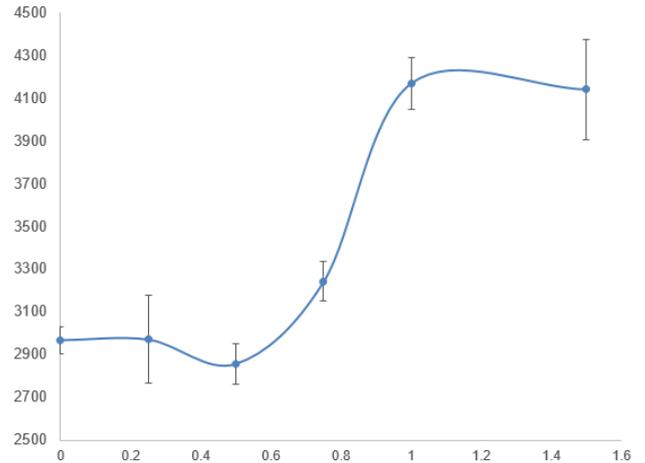


Figure 12 Suspension Consistency Vs Surface Roughness

Scale Up Studies.

The spray coating of cellulose nanofiber on the paper using a laboratory spray system confirmed promising results for scale up. However, these experiments have to be carried out batch wise. The integrated Dow web coater with professional Wagner spray system is developed for preparing similar spray coated CNF in a continuous mode.

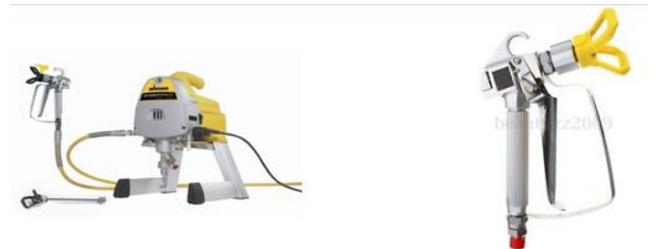
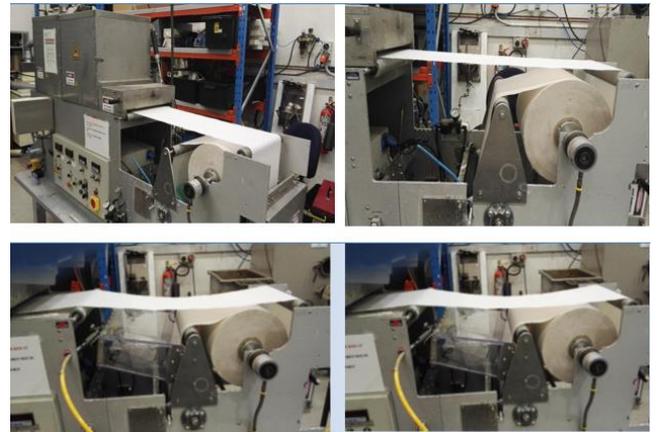


Figure 13 Spray system can be mounted in Dow web coater.

The suspension (CNF) sprayed on the base sheet was 1.5 Wt. % using this integrated Dow web coater with spray system. The CNF suspension from 0.5 to 1.00 Wt. % is also sprayed on the base sheet, due to continuous movement, the coating fluid is not stagnant on the base sheet and poor

adsorption of the brown paper. In the experimental setup for continuous process coating, the spray coated paper is stick to the other side of the paper when rewinding in the Dow web coater. It affects the coating surface on the base sheet. This problem arises due to the improper drying of the coated sheet in the dow web coater. Another limitation is dripping of the coating fluid from the base sheet due to moving from downward to upward in the Dow web coater. The spray gun is planned to locate on top of web near to the infrared heater 1 in the Dow coater. This rearrangement might help to prevent the dripping of coating fluid on the base sheet and coating could be retained on the base sheet. The preliminary results confirm that the spraying of cellulose nanofibre on the base sheet allows the complete retention of CNF suspension. The laboratory spray coating confirms that this technique enhanced the barrier properties of the base sheet. For continuous spray coating of CNF on the base sheet, the Dow web coater integrated with professional spray gun is used. In this experimental setup, the speed of the web controls the coat weight of CNF on the base sheet. Preliminary studies on Dow web coater showed that spraying is a feasible technology to increase the barrier and mechanical properties of the base sheet.

VI. CONCLUSION

Spraying CNF on paper substrates is a novel approach for increasing the barrier and mechanical properties of the paper. The operation time of spraying CNF for forming barrier layers on the substrates consumes less than one minute. Spray coating can capable of high suspension consistency for producing either thin or thick barrier layers on the paper substrates. Sprayed CNF could either fill or form the barrier layer on the surface pores of the paper substrates. As a consequence, the air permeability and oxygen transfer rate of the CNF laminated paper substrates decreased with increased CNF suspension coating. Simultaneously, the mechanical properties of the paper were tailored by the CNF lamination produced via spray coating. To conclude, the spraying process is a fast process for creating barrier layers on the paper substrates for improving their barrier and mechanical characteristics.

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