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Preparation of cellulose nanofibre laminates on the paper substrate via vacuum filtration

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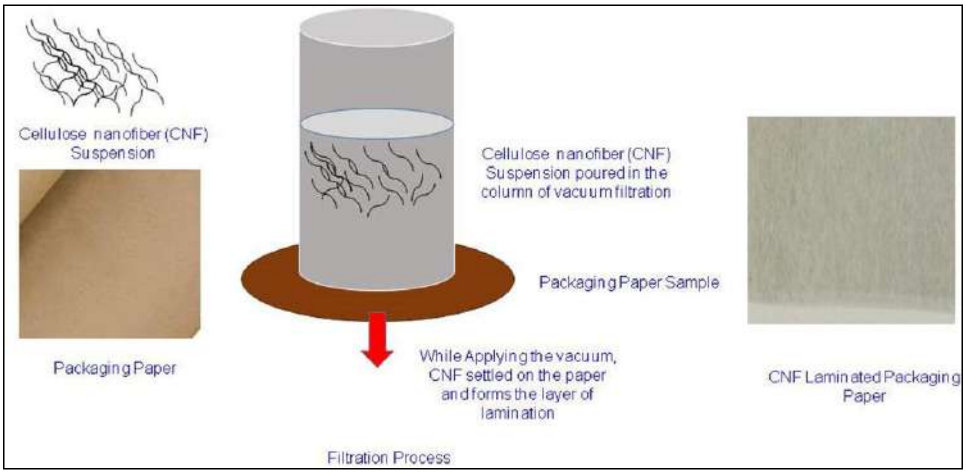
Abstract
Green Chemistry is the plan of synthetic items and cycles that lessen or wipe out the utilization and age of substances risky to people, creatures, plants and the climate. It is an advanced science that arrangements with the use of natural agreeable synthetic mixtures and materials in the different parts of our life like modern uses and numerous others. The start of green science is considered as a reaction to the need to diminish the harm of the climate by man-made materials and the cycles used to deliver them. Green science could incorporate anything from diminishing waste to discarding waste in the right way. Science assumes a crucial part in deciding the personal satisfaction. The synthetic substances industry and other related ventures supply us a gigantic assortment of fundamental items, from plastics to drugs. Be that as it may, these ventures can possibly genuinely harm our current circumstance. Green science in this way serves to advance the plan and productive utilization of naturally harmless synthetics and substance processes. This load of focuses will be examined in this article.

Keywords: Cellulose nanofiber, coating, lamination, vacuum filtration and air permeability

Introduction



Cellulose microfibers such as paper are an ecofriendly substrate used for developing packaging material. It is bio-renewable, biodegradable and biopolymer abundantly available in nature and a potential feedstock for functional materials such as coatings, films and membranes [1]. Cellulose based substrate can be good alternative for synthetic plastics and potential for solving global issues such as such as recycling, disposal and incineration of waste. However, the paper substrate has large surface pores and allowing a considerable amount of air and water vapour resulting poor barrier performance.

Paper and paper board generally used as packing and wrapping materials for packaging. The necessity of coating these substrates is required for improving their barrier performance against air, oxygen and water vapour. The coating of paper and paper board with wax, aluminium and synthetic plastics were already in practice. However, these coats on the substrates were not biodegradable and not recyclable and remain a threat to the environment. Recently, the coating of paper with cellulose nanofiber is an attempt for promoting the barrier performance of the paper substrates.

The production of cellulose nanofiber was carried out by disintegration and delamination of cellulose fibrils from pulp produced from a variety of green sources such as wood, potato tuber, hemp and flax. The diameter of CNF varies from 5 to 100 nm and is typically several micrometres in length [2]. These Cellulose fibrils at micro/nano scale are used to functionalize paper substrates by coating [3].

Cellulose nanofibre is also called as micro fibrillated cellulose (MFC), nano-fibrils, micro-fibrils and nano-fibrillated cellulose (NFC). CNF are isolated and processed from wood via various chemical, enzymatic, and/or mechanical treatments. Due to nano size of fibres, it has high aspect ratio, high specific strength, flexibility, large specific surface area, and thermal stability, combined with biodegradability and biocompatibility. Because of these properties, cellulose nanofiber is more suitable for a wide range of applications, such as film [4], reinforcing phase in composite materials [5], barriers in packaging [6], rheology modifiers for suspensions [7], filters for virus removal and water treatment technologies [8, 9], flexible platforms for biomedical applications [10] and printed electronic applications [11]. The CNF films/sheets and its nanocomposites and coated with fibre substrates increased the barrier and mechanical properties [12]. Due to outstanding multifunctional barrier properties such as oxygen and water transfer rate, it has the potential for application of packaging materials for foods [13].

Vacuum Filtration is the conventional technique for making the sheets and coating on the paper substrates. Using the method, The CNF coating on the paper substrates was carried out in this paper. The barrier and mechanical properties of CNF laminates of the paper substrates were evaluated.

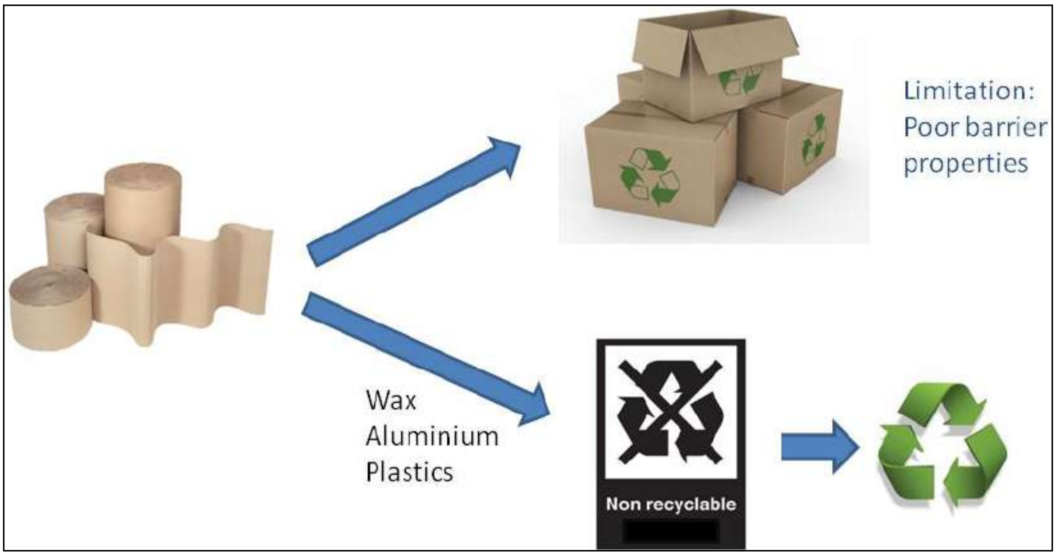


Fig 1: Limitation of coating of papers and paperboards

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 (CNF), cellulose nano-fibrils, cellulose micro-fibrils, nano-fibrillated cellulose (NFC) and Cellulose nanofiber (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 0.2 wt. with de-ionized water and disintegrating for 15,000 revolutions at 3000 rpm in a disintegrator.

CNF Lamination on the paper via Vacuum Filtration:

The disintegrated CNF suspension was diluted to 0.2 wt.% CNF suspension for preparing CNF laminates on the paper. The 600 ml of 0.2 wt.% CNF suspension was used to prepare 60g/m^2 basis weight of coat on the paper and it was 159 mm diameter coating on the paper. Based on the volume of CNF suspension, the coat weight of CNF lamination on the paper substrate can be varied from 5 g/m^2 to 60 g/m^2. CNF Lamination on the paper substrates were also prepared using the conventional vacuum filtration method using British Hand Maker (BHM) as reported in (Varanasi & Batchelor, 2013). 600 ml of CNF suspension with 0.2 wt.% concentrations was poured into a cylindrical container having a 150 mesh filter at the bottom and packaging paper should be placed above the 150 mesh and filtered to form a wet CNF lamination on the packaging paper.

Drying of Vacuum Filtered CNF Lamination on the paper

The wet CNF film coating on the paper substrates was carefully separated using blotting papers and subsequently dried at 105^{0}C in a drum drier for around 10 minutes. The basis weight (g/m^2) of each CNF lamination on the paper substrates was calculated by dividing the weight of the film, after 4 hours drying in the oven at a temperature of 105 °C, by the film area.

Fig 2: CNF lamination of Packaging Paper using vacuum filtration

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 blowed with Nitrogen to remove the any dust or any loose material on the sample and then coated with Iridium with a maximum thickness of 10µm. Moreover, the iridium coated samples are blowed 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

Cellulose nanofiber 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 µm and this micrograph is ideal for investigating the survey of the surface of the cellulose nanofiber 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 1µm and 10µm (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 °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 CNF laminated paper substrate was measured with an L&W air permeance tester with an operating range from 0.003 to 100 µm/Pa.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.

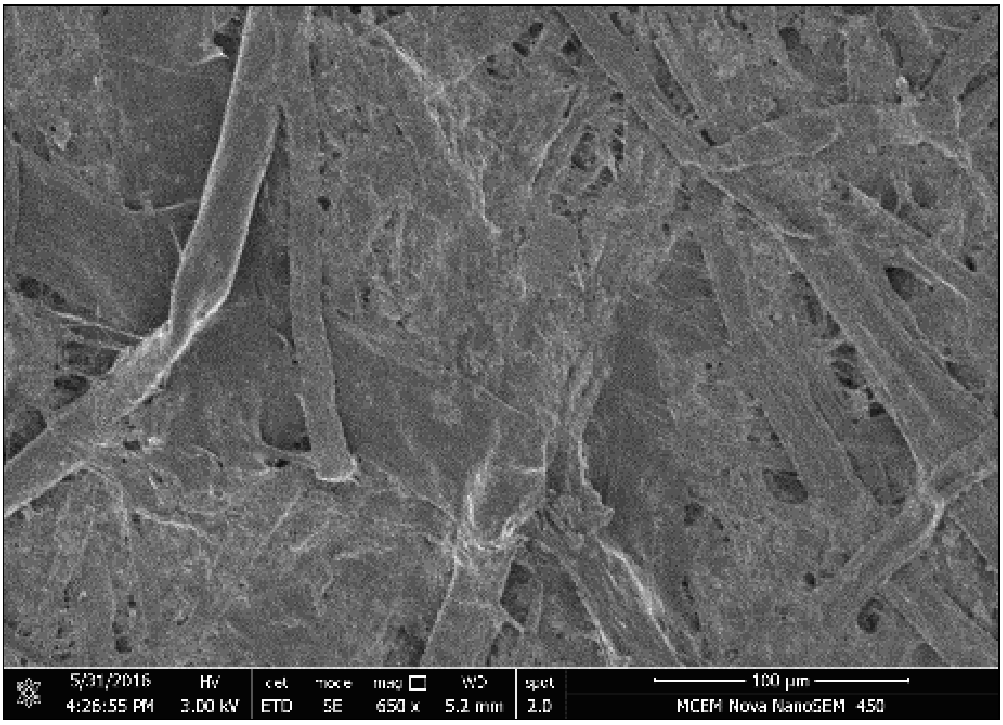
Oxygen Transfer Rate

The oxygen transfer rate of the sample can be calculated from their air permeability data. The OTR was evaluated by 4 times of the air permeability of the samples.

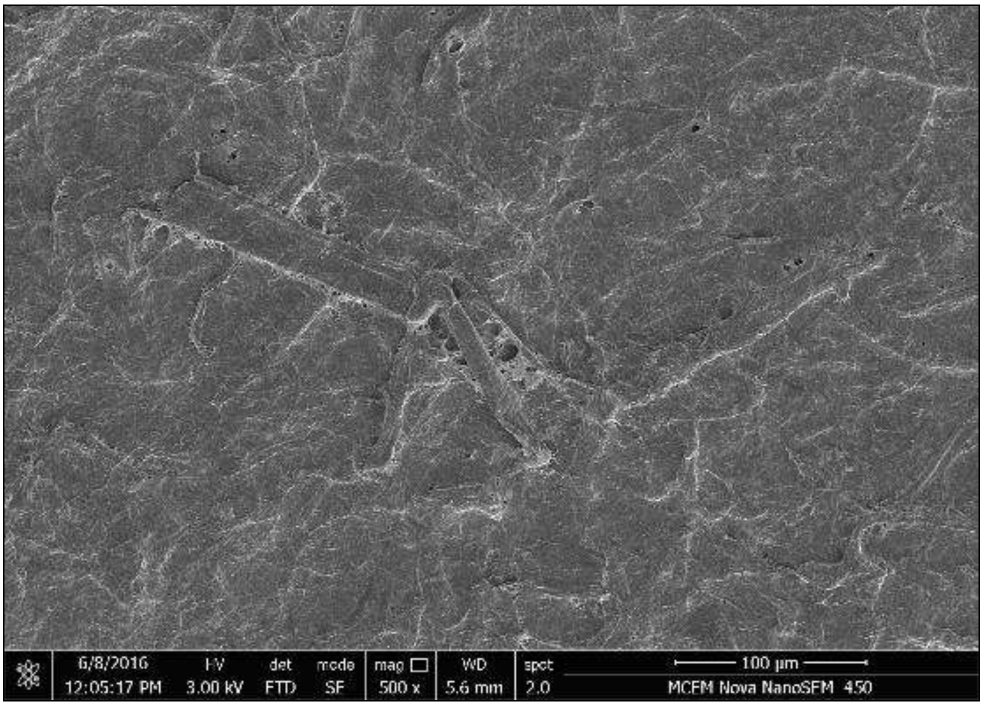
Results and Discussion

Most of food packaging materials are conventionally produced from synthetic plastic or the cellulose substrates such as paper and paper boards coated with petroleum based polymers. In practical approach, these materials provide good barrier performance with good mechanical strength. However, these packaging materials has non renewable source, high cost index relaying on the crude oil price, no chance or poor recyclability and its great impact in environment. To overcome this global issue, packaging materials from natural polymers has been utilized to replace the synthetic plastic packaging materials. Cellulose nanofiber is a novel carbohydrate polymer fibrillated from cellulose fiber extracted from pulps. It is reported that cellulose nanofibre has good mechanical and barrier properties with good biodegradability in the environment. Cellulose nanofibre has web like structure containing crystalline part producing tortuous path in the fibrous matrix. As a result, NC film has good barrier properties against air and water vapour and oxygen. The past results concluded that the NC film has good barrier properties and suitability for packaging application.

SEM Investigation of Unlaminated and CNF Laminated Paper



Uncoated Paper



CNF Laminated Paper

Fig 3: SEM micrographs of the uncoated and CNF laminated paper

Figure XX shows the SEM micrograph of CNF laminates of the paper and confirm that the CNF covers the surface pores of the paper and covers the paper via barrier layer. This CNF barrier layers on the paper improves the barrier performance against air and oxygen. The lamination of the paper with CNF improves their mechanical properties. The surface pores in the uncoated paper were open and reason for poor barrier performance. During the vacuum filtration, cellulose nanofibres are settled and retained on the paper and fills the surface pores and gradually formed the barrier layers on the sheet.

Air Permeability of CNF Laminated Paper

It is observed that CNF lamination produced on the paper substrate via vacuum filtration gives a promising result such as a considerable reduction of air permeability of the sheet composite. At lower CNF coating on the paper, it informed that the CNF fills the surface pores excellently via vacuum filtration. In vacuum filtration, the cellulose fibres are suspended in the watery suspension and slowly settled down and filling the surface pores and forming the CNF layers on the paper substrates. The film of CNF on the paper acts as barrier against air and oxygen. At higher CNF coat on the paper, the surface of the paper completely covered with CNF film and results in impermeable barrier against the air and oxygen.

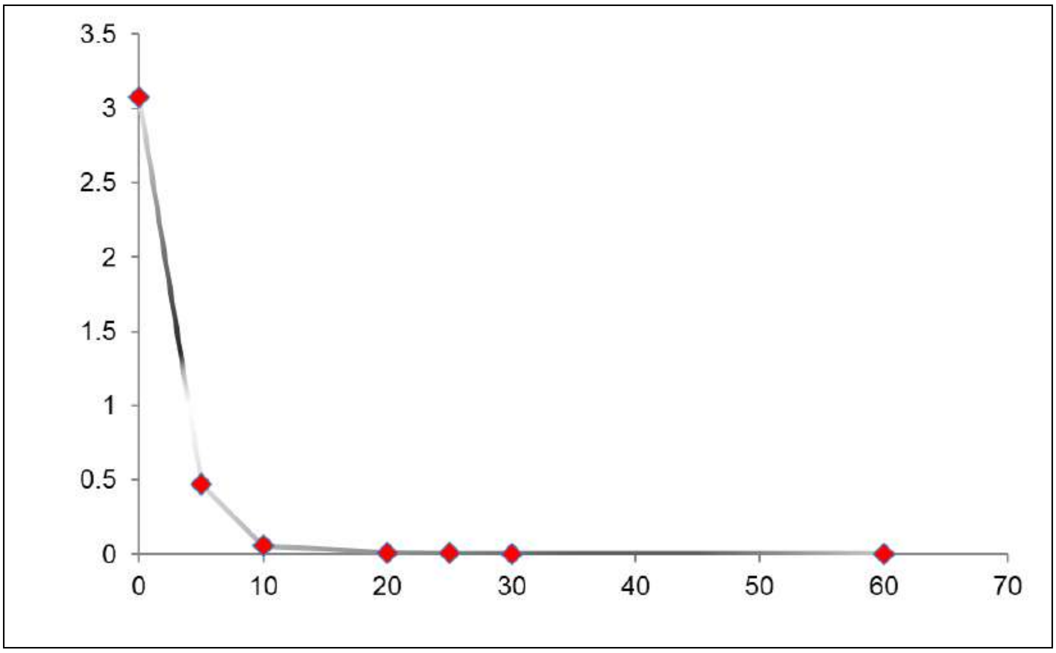


Fig 4: Plot between Air permeability (µm/Pa.S) and Coat weight (g/m^2)

Figure 04 shows the effect of CNF coat on the paper substrate and reveals that the air permeability of CNF laminates on the paper substrates decreased with increasing coat weight. The air permeance of the uncoated paper is evaluated to be 3.075 µm/Pa.S and 5 g/m^2 coat weight of CNF on the paper give an air permanence value of 0.472±0.209 µm/Pa.S. At this coat weight, CNF slowly blocks the surface pores and forms thin layer on the paper surface. As a result, the paper becomes good barrier against the air and oxygen. At the coat weight of 10 and 20 g/m^2, the paper surface completely covered the considerable thickness of the CNF and good barrier coating on the paper. As a consequence, the coated paper becomes the impermeable against the air and oxygen. The limitation of the instrument for measuring air permeance was < 0.003 µm/Pa.S.

Oxygen Transfer Rate

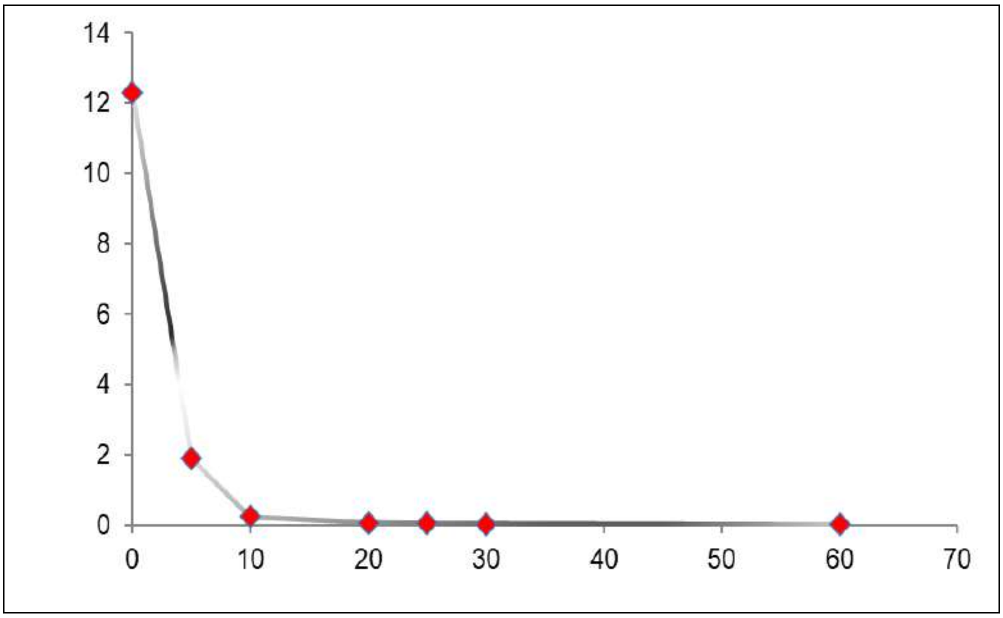


Fig 5: Plot between OTR (cc/m^2.S.Pa) vs. CNF coat weight on the paper

Figure 05 reveals the effect of CNF coat weight on the OTR of the CNF laminated paper. These values are evaluated from air permeance data. It shows that the OTR decreased with increased coat weight on the paper. Normally, Cellulose nanofibre has already lowest oxygen permeability comparable with synthetic polymers. CNF has the structure of complex tortuous pathway due to compact and various cellulose nanofibrils distributions. As a result, the diffusion pathway for air and oxygen was delayed and given good barrier potential. Figure 06 gives the plot between OTR (cc/m^2.day.Pa) vs. CNF coat weight on the paper. This is another version of the figure 5 confirming that CNF coating on the paper decrease the oxygen transfer rate.

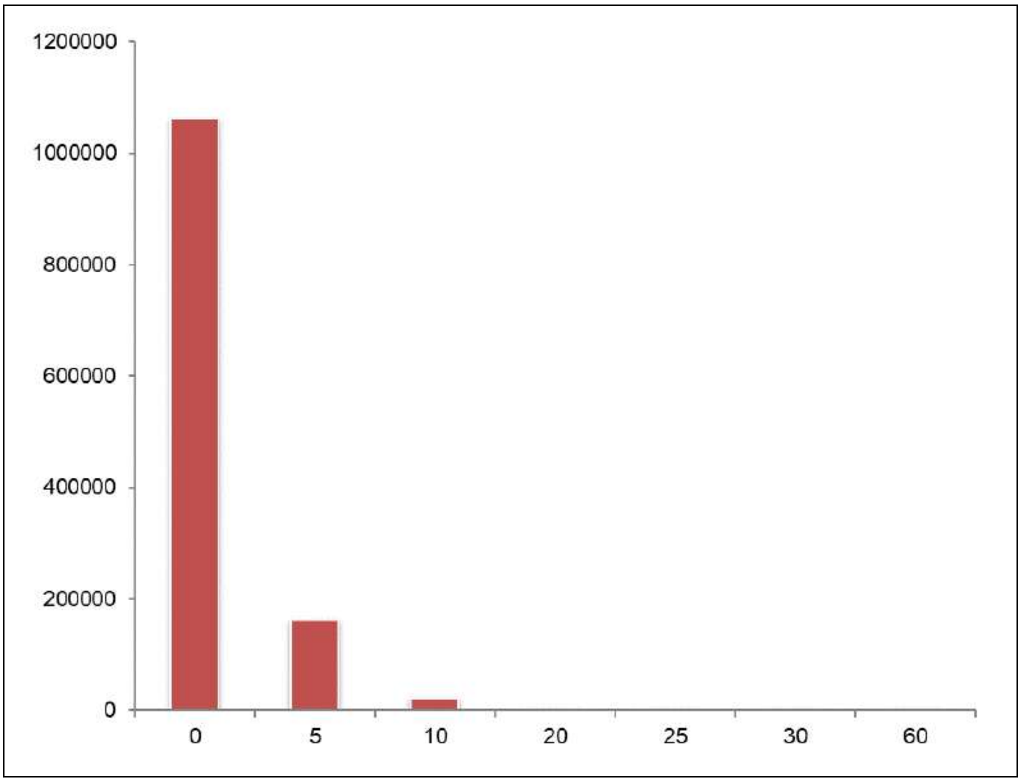


Fig 6: Plot between OTR (cc/m^2.day.Pa) vs. CNF coat weight on the paper

Conclusion

Due to plastic pollution, the biopolymers coating of papers and paper board is required to improve the barrier performance of the paper and mechanical strength of the paper. CNF coating on the paper is an achievable method for improving their barrier performance. CNF is a renewable bionanomaterial having high mechanical strength, good barrier properties against the water vapour and oxygen, large specific surface area and high aspect ratios. In addition to that, it has good biodegradability and biocompatibility. Generally, CNF consists of amorphous region and crystalline region in the cellulose nanofibrils matrix. These configurations in CNF produce the tortuous pathway for water vapour and oxygen. The CNF coating on the paper substrate via vacuum filtration is a conventional approach for CNF lamination on the paper substrates for promoting their barrier properties. The air permeance of the CNF laminates on the paper substrates have been reduced with increasing the coat weight. Given this correspondence, the vacuum filtration is a conventional method for producing CNF barrier layers on the paper substrates.

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